BEÜ Fen Bilimleri Dergisi
10 (3), 1083-1092, 2021

BEU Journal of Science
10 (3), 1083-1092, 2021

Araştırma Makalesi / Research Article

The Assessment of H_2S Emission from Araç Stream, in Karabük

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Abstract
Odor pollution is an important type of pollution that is subject to complaints in terms of quality of life and health. Since the odor perception is a sensorial process that occurs naturally with breathing, it is difficult to avoid when exposed. H_2S is one of the main sources of odor pollution and wastewater reservoir, estuarine and polluted river or waterways are among the most suitable areas for H_2S formation. The aim of this study is to determine the changes in H_2S concentrations on Araç Stream. Therefore, in this study, H_2S concentrations were determined instantaneously by using portable handheld gas monitors in air samples from the surface of Araç Stream at 15 points within the boundaries of Karabük city, and at the same time, pH and temperature values were determined. According to results, there were statistically significant differences in the H_2S and pH values between the sampling points ($p < 0.05$). H_2S concentration decreased in the order according to the sampling points as SP10< SP1< SP11< SP2< SP12< SP3< SP13< SP14< SP7< SP8< SP9< SP15< SP4< SP5< SP6. The highest level was determined after the industrial zone and the lowest levels were determined at the furthest point from the city center and industrial facilities. The results were shown in a map by using ArcMap program of ArcGIS software package.

Keywords: Odor pollution, H_2S, Araç Stream, Karabük.

Karabük Araç Çayı kaynaklı H_2S Emisyonunun Değerlendirilmesi

Öz
Koku kirliliği, yaşam kalitesi ve sağlık açısından şikâyet konusu olan önemli bir kirlilik türüdür. Koku algısı, nefes alma ile doğal olarak oluşan duyusal bir süreç olduğundan, maruz kaldığında kaçınılması zordur. H_2S, koku kirliliğinin ana kaynaklarından biridir ve atık su rezervuarı, nehir ağzı ve kirli nehir veya suyolları H_2S oluşumu için en uygun alanlar arasındadır. Bu çalışmanın amacı Araç Çayı üzerindeki H_2S konsantrasyonlarındaki değişiklikleri belirlemektir. Bu nedenle bu çalışmada Karabük ili sınırları içerisinde 15 noktada Araç Çayı yüzeyinden alınan hava örneklerinde portatif el tipi gaz monitörleri kullanılarak anlık H_2S konsantrasyonları belirlenmiş ve aynı zamanda pH ve sıcaklık değerleri belirlenmiştir. Sonuçlara göre örnekleme noktaları arasında H_2S ve pH değerlerinde istatistiksel olarak anlamlı farklılıklar vardı ($p < 0{,}05$). H_2S konsantrasyonu, örnekleme noktalarına göre sırasıyla SP10< SP1< SP11< SP2< SP12< SP3< SP13< SP14< SP7< SP8< SP9< SP15< SP4< SP5< SP6 olarak azalmıştır. En yüksek seviye sanayi bölgesinden sonra, en düşük seviyeler ise şehir merkezi ve sanayi tesislerine en uzak olan noktada belirlenmiştir. Sonuçlar ArcGIS yazılım paketinin ArcMap programı kullanılarak bir haritada gösterilmiştir.

Anahtar kelimeler: Koku kirliliği, H_2S, Araç Çayı, Karabük.

1. Introduction

Many people experience difficulties in terms of health and living comfort both in the workplace and in residential areas when exposed to different levels of hydrogen sulfide (H_2S) emissions that is the main sources of odor [1, 2]. H_2S is one of many airborne pollutants emitted by different sources such as rayon production, livestock feedlots, hog production facilities, meat processing plants, wastewater treatment

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Recieved: 30.03.2021, Accepted: 12.08.2021

plants, pulp and paper industry, oil and gas refining processes [3], concentrated animal feeding operations [4], wastewater sources [5], river estuary [6] and polluted river or streams [7].

Water, one of the major components of life-support environment that is part of earth is vital necessity for ecosystem [8]. However, decrease in the quality of water resources is one of the most crucial problems that negatively affect human health and quality of life. In general, water resources are polluted as a result of anthropogenic activities due to the discharge of untreated domestic and industrial wastewater, population growth, pesticides and fertilizers, organic and inorganic wastes, urban development and poor water resource management systems [7, 9, 10]. Especially the discharge of sewage system and industrial wastewater directly to the receiving water sources reduces the water quality and causes odor problems.

H_2S is one of the most important compounds that cause odor and has very serious health effects. H_2S is a colorless, flammable gas and has a strong odor of rotten eggs [3, 11]. In nature H_2S produced by decaying organic matter, is formed by the reduction of sulfate to hydrogen sulfide gas with bacteria under anaerobic conditions in septic or sewer systems [11]. H_2S can cause eye irritation, upper respiratory tract irritation, respiratory failure, and histopathological changes in the nasal cavity. It may also affect the nervous system, cardiac and vascular function, and blood pressure [3].

Since the odor perception is subjective, the odor threshold of H_2S varies between 0.01-0.3 ppm. While 1-5 ppm H_2S is a moderately offensive odor level, it may cause nausea, tearing in the eyes, headache, or sleep loss with prolonged exposure. Eye damage, indigestion and loss of appetite may occur if exposed to 20-50 ppm H_2S on a few days. Severe eye damage and lung irritation, sudden loss of consciousness within 30 minutes; and death can occur within 4-8 hours when exposed to a concentration of 500 ppm H_2S [12, 13]. Inhalation of H_2S concentrations greater than 1000 ppm may be fatal in both humans and animals within seconds or minutes [11].

It is a fact that due to the developing industry and diversified economic activities, increasing population, the spread of irrigated agriculture, the increase in chemical fertilizers and other pesticides, both the amount of water and the water quality will be affected more negatively in the future. Thus, in this study, the H_2S pollutant formed in Araç Stream in Karabük was evaluated. The pollution load that increases due to the human activities of the stream reaches its maximum level at the Karabük outlet. Iron and steel plants, rolling mills, wastes from other industries, pesticides, domestic wastes and untreated wastewater cause the increase in pollution in the stream [14].

In addition, the pollution in the stream has caused fish deaths and annoying odors to reach serious levels [15]. However, since there is no study in the literature that determines the pollution level in Araç Stream, especially in terms of H_2S, this study will fill this gap. For these reasons, the aim of this study is to determine the H_2S concentration by sampling on the surface of the Araç Stream at 15 different points and perform spatial distributions of H_2S by ArcMap program of ArcGIS software package.

2. Material and Method

2.1. Study Area

The parts of the Araç and Soğanlı Streams within the borders of Karabük constitute the study area. Araç Stream subject to this research is located in North of Turkey Westen Black Sea region, between South slope of Kure Mountains and North slopes of Ilgaz Mountains [14]. The source of the Araç Stream is the northern slopes of the Ilgaz Mountains. The total length of the Araç Stream is 150 km, the length within Karabük is 73 km and flow rate is 18.714 m^3/sec. Soğanlı Stream originating from the southwest of Gerede. merges with Araç Stream in Karabük and takes the name Yenice River. The total length of Soğanlı Stream is 70 km, the length within Karabük is 32 km and flow rate is 28.066 m^3/sec [16].

2.2. Sampling Strategy and Analysis of H_2S

It was thought that H_2S formation would be high in September due to the fact that the air temperature is above seasonal values. For this reason, although it is seen as the weak point of the study, the samples were collected one time on 14-15 September 2020. A total of 13 samples were collected from the 35 km long section of the Araç Stream. On the other hand, 2 samples were collected from Soğanlı Stream at a distance approximately 5 km before merging with Araç Stream. A total of 15 samples were collected at distances ranging from 0.5-10 km. Sampling points (SP) are shown in Figure 1. Air sample was taken from the water surface by using apparatus with a 25 cm diameter as a flux chamber, and highly sensitive digital portable measuring devices with 550 ml/min flow rate (GEOTECH BIOGAS 5000 - Portable Gas Analyzer) were used for H_2S measurement. Measurements were carried out using portable pH meter to determine the water pH value. Water temperatures were determined using a mercury thermometer. All measurements were carried out instantaneously at the sampling points and recorded. The results were shown in a map by using ArcMap program of ArcGIS software package.

Figure 1. Sampling points

2.3. Statistical Analyzes

IBM SPSS 22 statistic Data Editor was used in the statistical analysis of the results obtained. The Levene trial was used for normal distribution and homogeneity of deviations, so that the mean values of the H_2S (ppm) variables taken at various pollution points along the Araç Stream and the statistical differences between pollution points are revealed. Following this, an ANOVA test was performed as there were more than two groups. Differences between measured H_2S values at 15 different points were tested using Duncan's multiple comparison analysis. In the analyses, the α significance level was evaluated at 0.05.

2.4. Spatial Distribution

Geographical information system programs such as ArcGIS provide visual information about pollution sources and impact areas. ArcMap program of ArcGIS software package.was used to create a digital map by using values of H_2S, pH and temperature. The coordinates of the sampling points were recorded while the samples were collected to create the distribution map. Grid, a raster GIS file format, was used to record the interpolation using the data at the sampling points. Topographic maps, satellite images and similar data of the study area were collected using GIS software GeoMedia and ArcGIS. Thus, an updatable database was created by entering the database [17].

3. Results and Discussion

H_2S is typically produced as a result of the degradation of sulfur-containing organic materials by bacterial activity [18]. However, parameters such as pH, temperature, BOD, COD and DO are important variables in this process. Dissolved oxygen is necessary for organisms in metabolic processes. The amount of oxygen required by organisms in the aerobic metabolic process is represented by BOD, and the amount of chemical oxygen required for the decomposition of organic matter by chemical reaction is represented by COD. COD is also used to estimate the amount of organic matter in water [19]. In aquatic environments such as coastal areas and ports close to cities, low dissolved oxygen levels are observed due to the biological oxygen demand resulting from seasonal temperature changes [20]. The low dissolved oxygen level leads to the conversion of the aerobic environment to the anaerobic environment [19], which is a suitable environment for the formation of H_2S. Temperature is an important parameter in the H_2S formation process due to its significant effect on DO and bacterial activity. As the temperature increases, the volatility of the liquid increases and at the same time the activities of bacteria in the anaerobic environment increase. It also contributes to the anaerobicity of the environment as oxygen is less soluble at high temperatures.

Sulfate ion is one of the important anions found in natural waters and wastewater. Since sulfate is used as an electron acceptor for biochemical oxidations catalyzed by anaerobic bacteria in the absence of dissolved oxygen and nitrate, the sulfate ion is reduced to HS^- ion and S^{-2} ion under anaerobic conditions to form hydrogen sulfide (Equation 1-3).

$$SO_{4-}^{2} + \text{organic matter} \xrightarrow[\text{bacteria}]{\text{anaerobic}} S^{-2} + H_2O + CO_2 \quad (1)$$

$$S^{-2} + H^+ \leftrightarrow HS^- \quad (2)$$

$$HS^- + H^+ \leftrightarrow H_2S \quad (3)$$

Different pH levels affect the equilibrium reaction between H_2S, HS^- and S^{-2}. For example, if the pH level is 9 and above, most of the reduced sulfur is in the form of HS and S^{-2} ions, and the amount of H_2S is low. On the contrary, if the pH level is 9 and below, the equilibrium shifts towards the formation of un-ionized H^2S and the amount of H_2S increases [21].

Therefore, in order to determine H_2S, pH and temperature values, on-site measurements were carried out instantaneously triplicate at each point. Measurement results were given in Table 1.

Table 1. Values of H_2S, pH ve Temperature

Points	Coordinates		Description	H_2S (ppm)	pH	T (ºC)
	Latitude	Longitude				
SP1	41.2035	32.9045	There are no industrial facilities or heavy pollutant sources	1.00	7.71	19.00
SP2	41.2151	32.7703	There are no industrial facilities or heavy pollutant sources but within the residential areas	2.33	8.15	25.00
SP3	41.2136	32.7571	There is a residential area, restaurant and shopping center. Agricultural activities are also carried out	3.00	8.52	24.00
SP4	41.2158	32.7372	It is close to the residential area and poultry farm activity	3.67	8.41	25.00
SP5	42.2219	32.7129	There is no residential area and pollution source	4.00	7.81	23.50
SP6	41.2219	32.7129	It is located after the industrial site, textile production and residential area	5.33	8.10	24.00
SP7	41.2115	32.6646	There is intensive greenhouse activities, residential area and dormitory building	3.33	8.24	28.00
SP8	41.2078	32.641	There are not residential areas however, there are university and small iron and steel production facilities	3.33	8.19	24.50
SP9	41.2006	32.6302	There are not many residential areas, however greenhouse cultivation activities are carried out	3.33	7.85	24.00
SP10	41.1652	32.6434	It is located before the city center of Karabük and the big iron and steel industry facility	0.67	8.10	24.00
SP11	41.1893	32.6192	It is located after the heavy iron and steel making industry	2.00	8.17	24.00
SP12	41.1923	32.6084	It is the first point after the merger of Soğanlı and Araç Stream	2.67	8.17	27.00
SP13	41.172	32.583	Planned residential area is the majority in this area and there is no sources of serious pollution	3.00	8.01	25.00
SP14	41.1691	32.579	It is located before an integrated meat facility	3.00	8.03	27.00
SP15	41.1594	32.5592	It is located after the wastwater treatment plant	3.33	7.90	24.50

According to the results, the lowest concentration of H_2S were obtained at SP1 and SP10 as 1 ppm and 0.67 ppm respectively. SP1 was at the beginning position of the Araç Stream at the Karabük provincial border. When this point was investigated, it was seen that there were no industrial facilities or heavy pollutant sources nearby. SP10 was located before the city center of Karabük and the big iron and steel industry facility. In addition, it is thought that this point remained cleaner due to the presence of a dam 3.22 km before this point. The fact that both points were cleaner than the others is expected as they did not interact with the pollutant sources.

The third lowest H_2S concentration was at SP2, approximately 10 km from SP1. Although there is no industrial facility and heavy pollutant source between both points, SP2 is within the residential areas. At this point, the H_2S concentration increased 133% compared to the first point. SP3 is 11.26 km from SP1 and 1.26 km from SP2. This point is located in the area where facilities such as residential area, restaurant and shopping mall are located. In addition, agricultural activities are carried out around it. At this point, H_2S concentration increased by 29% and 200%, respectively, compared to SP2 and SP1. SP4 is 13.05 km from SP1 and 1.79 km from SP3. SP4 has a higher potential for pollution than the previous point, as it is close to the residential area and poultry farm activity. At this point, H_2S concentration increased by 22% and 267%, respectively, compared to SP3 and SP1. SP5 is 15.43 km from SP1 and 2.38 km from SP4. When examined between two points, it is seen that there is no residential area. In addition, there is no factor to increase the pollution level. Therefore, there is no serious difference between the two points. At this point, H_2S concentration increased by 9% and 300%, respectively, compared to SP4 and SP1. Sampling point 6 is located after the industrial site, textile production and residential area. SP6 is 18.06 km from SP1 and 2.63 km from SP5. The highest concentration of H_2S was determined at the this sampling point. At this point, H_2S concentration increased by 33% and 433%, respectively, compared to SP5 and SP1. SP7 is 21.52 km from SP1 and 3.46 km from SP6. It is seen that there is intensive greenhouse activities, residential area and dormitory building in this area. SP7 has H_2S lower concentration of than the previous point. At this point, H_2S concentration decreased by -38% and increased by 233%, respectively, compared to SP6 and SP1. SP8 is 25.19 km from SP1 and 3.67 km from SP7. There are not residential areas however, there are university and small iron and steel production facilities. SP8 has the same concentration of H_2S as the previous point. At this point, H_2S concentration increased by 0% and 233%, respectively, compared to SP7 and SP1. SP9 is 26.42 km from SP1 and 1.23 km from SP8. Although there are not many residential areas, greenhouse cultivation activities are carried out. In addition, landscaping were made on along the stream in this area. This situation causes no change in SP9 has the same concentration of H_2S as the previous point. At this point, H_2S concentration increased by 0% and 233%, respectively, compared to SP8 and SP1. SP11 is located after the heavy iron and steel making industry, and SP11 is 4.52 km from SP10 on the Soğanlı Stream. This sampling point is the second point on the Soğanlı Stream and the last sampling point before joining the Araç Stream. It is seen that the H_2S concentration at this point is higher than SP10. At this point, H_2S concentration increased by 199%, compared to SP10. SP12 is 28.02 km from SP1, 5.18 km from SP10. At SP12 that is the first point after the merger of Soğanlı and Araç Streams H_2S concentration is 2.67 ppm. This result is higher than SP11 (2.00 ppm) on the Soğanlı Stream however, lower than the SP9 (3.33 ppm) on the Araç Stream. SP13 is 3.94 km from SP12. Planned residential area is the majority in this area and there is no sources of serious pollution. At this point, H_2S concentration increased by 12% compared to SP12. The distance between SP14 and SP13 is 0.5 km. Between these two points there is only an integrated meat facility. Although an increase in pollution is expected with this facility, fortunately there is no difference in concentration (3.00 ppm) between the two points. The distance between SP15 and SP14 is 2.18 km. The last sampling point is located after the wastwater treatment plant. At this point, H_2S concentration increased by 11% compared to SP14.

The H_2S in gaseous form on the surface of Araç Stream passed into the atmosphere and dispersed. The spatial distribution map was prepared to show affect area of H_2S using ArcMap program of ArcGIS software package. (Figure 2). As seen in Figure 2, the effect of topography and meteorological conditions is not seen in distribution map. Meteorological, topographic and emission information are used in the air quality modeling system created with the mathematical simulation of real weather conditions in the AERMOD program. With the modeling, the change in the concentrations of the pollutants emitted from the source can be seen according to the meteorological and topographic changes in the study area [22]. Therefore, it will be beneficial to use the AERMOD program to create a more precise distribution map.

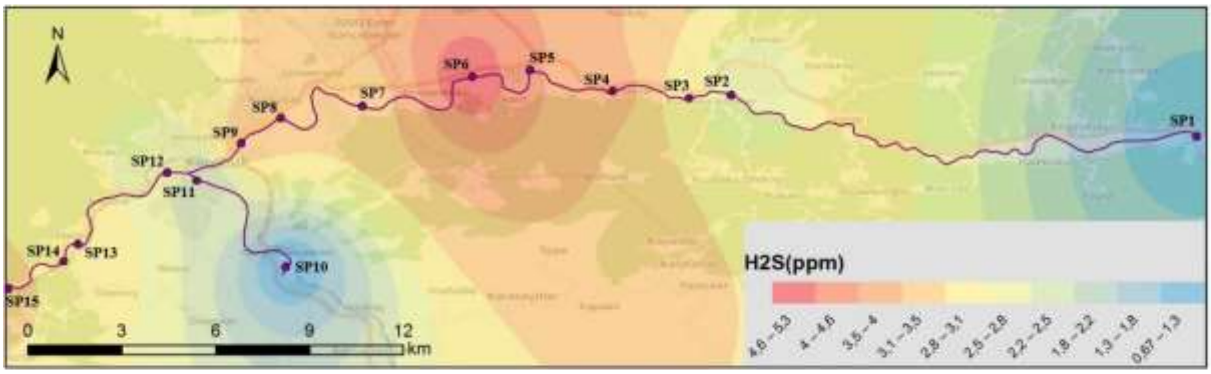

Figure 2. Distribution map of H_2S

Based on the Levene test, the hypothesis that the variances were equal was confirmed, F(14.30)=256.155, P=0.673 (p>0.05) (Table 2). This is because the mean values of the H_2S (ppm) variables of different pollution points along the Araç Stream and the statistical differences between the pollution points. In the ANOVA test of group differences, the difference between points was very significant at level P<0.001 (Table 3). In Duncan's multiple comparison analysis, there is no significant difference between the mean H_2S levels at SP7, SP8, SP9 measured at 15 different points. And SP3, SP13, SP14 points, there is no significant difference among H_2S levels. However, it is clear that there is a significant difference between the other points measured according to the Duncan test and between points SP3, SP7, SP8, SP9, SP13, SP14 (Table 4).

Table 2. Levene Statistic Homogeneity Test

H_2S (ppm)			
Levene Statistic	**df1**	**df2**	**Sig.**
0.789	14	30	0.673

Table 3. ANOVA Testing of Levels of H_2S (ppm) between Points

	n	**df**	**F**	**Sig.**
Between Groups	15	14	256.155	0.000
Within Groups	3	30		
Total	45	44		

Table 4. Duncan multi comparison test

Points	**SP1**	**SP2**	**SP3**	**SP4**	**SP5**
H_2S(ppm) Mean±Std	1±0.05	2.33±0.2	3±0.15	3.67±0.2	4±0.1
Groups	h	f	e	c	b
Points	**SP6**	**SP7**	**SP8**	**SP9**	**SP10**
H_2S(ppm) Mean±Std	5.33±0.1	3.33±0.1	3.33±0.2	3.33±0.1	0.67±0.02
Groups	a	d	d	d	i
Points	**SP11**	**SP12**	**SP13**	**SP14**	**SP15**
H_2S(ppm) Mean±Std	2±0.1	2.67±0.1	3±0.05	3±0.1	3.23±0.1
Groups	g	e	e	e	d

P<0.05

According to the statistical analysis, statistically significant differences were found between the sampling points in H_2S and pH values using the SPSS Statistics program (p <0.05) but were not found for temperature. This result shows the effect of the properties of the sampling points on H_2S and pH. Spatial distribution map of temperature and pH values is shown in Figure 3 and 4.

Figure 3. Temperature values (*°C)

Figure 4. pH values

It has been stated in the study by Wiener et al. that many rivers empty into the Mexican port and the Gulf of Mexico is one of the world's largest oil producing areas. It was also emphasized in the study that H_2S formation is affected by the oxygen content of the water [20]. It has been reported in the study by Isworo et al. that Indonesian State Oil Company activity produces waste that allows pollution of the Donan river. The values of the pollutant parameters were determined to reveal the pollution level of the study area. BOD, COD, DO levels were found in the range of 5.5-7.2 ppm, 33.64-33.73, 6.01-5.90 ppm, respectively. In addition H_2S concentration was determined as 0.2 ppm [19]. In the study conducted by Ogbemudia and Ita [6] in Lower Stubbs Creek, Qua Iboe River Estuary, Nigeria air quality was assessed with samples taken from 5 different points using portable hand held gas monitors for atmospheric gases. The concentration of H_2S ranges from 0.6 ppm to 0.9 ppm, with an average of 0.77 ppm. H_2S values in both studies were found to be lower. In study by Moreno-Silva et al. [5] different H_2S emission sources were evaluated and simulated in order to determine the odor effect of the Munas reservoir in Colombia. Flux chamber was used in the study and it was figured out that in critical conditions Munas reservoir can cause an atmospheric concentration of 4 ppm H_2S in reservoir-surrounding urban region. This result was in the H_2S level range (0.67-5.33 ppm) in present study due to the similarity of the method and study area. In another study by Antai et al. [23] changes in air quality was assessed in wet season in Eleme, Rivers State, Nigeria and the average concentration of H_2S was recorded as 0.37 ppm. Because the study was conducted in the rainy season, lower results may have been obtained. In the study conducted by Novita et al. [7], the water quality of the Bedadung River in Jember Regency, East Java, Indonesia was evaluated with samples taken from 5 different points, and the average concentration of H_2S was determined as 0,035 mg/dm^3. In a study conducted in 1998, the average sulfur concentration in river waters was found to be 180 mg/L. The irritating odors caused by especially H_2S continued until the first half of 2001 where streams carried untreated domestic and industrial wastewater to İzmir Bay [24, 25]. Rim-Rukeh found H_2S concentration in the vicinity of dump site fires in the range of 3.4 to 7.7 ppm [26]. The average H_2S concentration in present study is also within this range.

Since H_2S formation is affected by water or wastewater properties such as BOD, COD, DO, pH and temperature, as well as environmental conditions such as untreated industrial or domestic wastes, and use of area such as industrial, residential or agricultural, different or close concentrations of H_2S can be detected in each study. As different land uses change the originality of the land structure [27], anthropogenic activities can also disrupt the structure of water and air. In this study area, there are many different anthropogenic activities such as agriculture, iron and steel industry, vehicle repair, institutional, domestic and livestock. For example, livestock is one of the most important sources of H_2S. Animal waste can be a source of H_2S as a result of the degradation of biosolids, including manure and feed [28]. In addition, wastewater greatly contributes to the formation of H_2S as a result of decomposition of organic matter by microorganisms in anaerobic conditions [29]. Moreover, industrial activities can also cause H_2S pollution [30], especially in food, rubber and leather industries, odor pollution arising from raw materials and processes can be observed [31]. Although it is at a very low level compared to other e emission sources, solid waste landfills are among the most important H_2S sources [32]. Finally, H_2S is also associated with agricultural processes [33] due to the decay of the organic structure of agricultural wastes in aqueous environments such as streams. Therefore, different or close concentrations of H_2S were determined both between sampling points and other studies depending on anthropogenic activities.

4. Conclusion and Recommendations

In this study, the differences in atmospheric H_2S concentration along Araç Stream were determined. In addition, the spatial distribution of H_2S emissions from Araç Stream was shown in this study.

- It has been observed that closeness of residential areas, industrial sites, agricultural and industrial activities cause an increase in H_2S concentration.
- Statistically significant differences were found between the sampling points in H_2S and pH values using the SPSS Statistics program ($p < 0.05$), but were not found for temperature.
- In pariticular, it was observed that the H_2S level was higher at the sampling points after the places where the untreated water was given to the Araç Stream.
- It was determined that the measurements were above the odor threshold value.
- In addition, since the H_2S concentraions are generally between 1-5 ppm, prolonged exposure may adversely affect health and quality of life.
- Since there are no topographic and meteorological effects in the spatial distribution map by ArcMap program of ArcGIS software package, it will be useful to use programs such as AERMOD to create a more precise distribution map.

H_2S has not been measured in Araç Stream before. For this reason, it can be used as an important data source in the evaluations to be made about whether the pollution load will increase in the future.It is also expected to contribute to the literature as a data source that can be used for comparison in similar studies conducted in different streams.As a result, it was observed that H_2S was spread from the Araç Stream but was not effective in residential areas due to atmospheric dispertion. However, it is obvious that it may cause problems in the future unless the necessary measures are taken. Therefore, tight control should be applied and sources of pollutants given to the Stream should be prevented.

Author's Contributions

All authors contributed equally to the study.

Statement of Conflicts of Interest

There is no conflict of interest among the authors.

Statement of Research and Publication Ethics

The authors declares that this study complies with Research and Publication Ethics.

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