

Sources of adsorbable organic halogens (AOX) in sludge of Gaza

B. Shomar *

Institute of Environmental Geochemistry, University of Heidelberg, Im Neuenheimer Feld 236, 69120 Heidelberg, Germany

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Abstract

Average concentrations of adsorbable organic halogens (AOX) in sludge from the Gaza Strip reached 600 mg kg^{-1} which currently represents a major limiting factor for sludge application in agriculture. This study aims to identify the main sources of AOX in Gaza where the area is small with limited industrial activities. The results showed that the AOX in effluent sludge is formed mainly inside the treatment plant. Although the plant receives AOX wastewater of only $85 \mu\text{g l}^{-1}$, this value increased five fold inside the plant to an average of $400 \mu\text{g l}^{-1}$ in the effluent wastewater. On the other hand, the sludge from the first sedimentation pond showed an AOX concentration of 213 mg kg^{-1} and increased in the final sedimentation pond to 500 mg kg^{-1} . The AOX concentration in three month old sludge was 130 mg kg^{-1} . The textile and detergent industries are the major AOX producing industries in Gaza, with an average AOX of $40000 \mu\text{g l}^{-1}$ in their effluent wastewater. These values do not represent a significant AOX pollution load to the treatment plant because these industries are very limited and their effluents are diluted before entering the municipal treatment plant. Industrial activity decreased in 2006 to less than 70% of the previous four years. Consequently, the AOX in the industrial effluent decreased from 150000 to $40000 \mu\text{g l}^{-1}$ over the same time period. Sludge from these industries showed 1300 mg kg^{-1} AOX for the past four years and only 400 mg kg^{-1} in 2006. Moreover, the AOX in the influent wastewater discharged to the treatment plant decreased from 320 to $85 \mu\text{g l}^{-1}$ for the same period. This study revealed that the major AOX is formed inside the treatment plant and especially in the anaerobic facilities. There was no correlation between the AOX concentrations in the sludge and the wastewater at the same location. Moreover, there was no correlation between the AOX in sludge/wastewater and dissolved oxygen, pH and chloride in that treatment plant.

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1. Introduction

In his review, Müller (2003) distinguished between the AOX “adsorbable organic halogens” in water and the AOX-S18 in sludge where A stands for “adsorbed” and not for “adsorbable”. The latter term includes the most important water insoluble organochlorine product: polyvinyl chloride (PVC). Adsorbed organohalogenes, such as those in sludge, are hydrophobic and therefore not adsorbable. Mineralogenic halogens (X) are neither organic nor adsorbable, and therefore by definition not AOX.

More than 3700 organohalogen compounds, mainly containing chloride or bromide but a few with iodide and

fluoride, are produced by living organisms or are formed during natural abiogenic processes, such as volcanoes, forest fires, and other geothermal processes (Gribble, 2003). Keppler and Biester (2003) showed that the natural occurrence of AOX in soil and peat lands could reach values up to 5 and 2000 mg kg^{-1} , respectively.

AOX represents a wide range of substances that are defined by the binding of a halogen containing chemical to activated carbon (Grimvall and de Leer, 1995). AOXs include chemicals of differing structures and toxicological profiles. Consequently, relevant toxicological endpoints vary cannot be described without further specification of AOX (Planquart et al., 1999). The chemical found in the AOX of in different sewage sludges or wastewaters can be quite diverse depending on the origin of the samples. Use of low-cost chlorous cleaners (chloramine T, chloramine

* Tel.: +49 6221 546 009; fax: +49 6221 545 228.

E-mail address: bshomar@ugc.uni-heidelberg.de

BARS, sodium hypochlorite, chloric lime) can create AOX in wastewater (Sebb, 1980). Both chlorination and ozone treatment may lead to the formation of AOXs called trihalomethanes (THM). Bromine derivatives can also be formed when the water contains small amounts of bromine (Grohmann, 1991).

Although the environmental significance of AOX is controversial, many jurisdictions have adopted strict regulatory requirements limiting its discharge (Liu et al., 1997). The German regulation limits AOX in sludge to 500 mg kg⁻¹ for use in agriculture (UMK-AG, 2000; Langenkamp and Part, 2001). Sauerbeck and Leschber (1992) reported that these limits are a purely precautionary and not based on scientific evidence of immanent toxicity. However, a large number of AOX causing substances show a significant eco-toxicity (Höfl et al., 1997). While concentrations of AOX in sludges do not give information about the absence or presence of hazardous substances, they could serve as a marker for levels of anthropogenic compounds, some of which may be persistent pollutants, in soil (Leschber, 1992). In the context of soil contamination, it is noteworthy that some organic halogens may be transformed in the soil to more toxic compounds such as vinyl chloride, which is a known human carcinogen (Salkinoja-Salonen et al., 1995; AURAS, 2001).

Due to the scarcity of water resources in the Gaza Strip, it is crucial to control the quality of wastewater for reuse. Additionally, using sludge as fertilizer would likely provide benefit to the agricultural sector. Recent studies by Shomar et al. (2004, 2005) found that more than 85% of sludge samples collected in 2001, 2002, 2003 and 2004 from the wastewater treatment plants in Gaza had average AOX levels of 600 mg Cl kg⁻¹. This level exceeds the standards of all industrial countries for sludge to be used in land applications and thus poses chemical limitations for sludge applications in Gaza. The studies also found that the upper 40 cm of soil were the affected by wastewater and sludge.

Several industries, such as the manufacture of polyvinyl chloride (PVC) and waste incineration, are important sources of AOX worldwide. Several studies showed that the amount of PVC in wastes influences levels of AOX (Mertens, 1996; Litz, 1998; Mertens, 1999). The increasing use of industrially produced synthetic organohalogen (mainly chlorine) compounds has led to the accumulation of these compounds and their derivatives in the environment. This can lead to serious adverse health and environmental effects (Müller, 2003). DDT, and its metabolites and many other pesticides detected in the water and the soil of Gaza may serve as examples (Shomar et al., 2005).

The industrial sector in Palestine is presently underdeveloped. Most industries are concentrated in the city of Gaza and in the northern areas, grouped in two main industrial estates, Gaza Industrial Estate (GIE) and the smaller Beit Hanoun Industrial Estate (BIE). Several industries are scattered among residential areas. The industries with greatest potential for environmental pollution are the agricultural/farming and food processing, chemical

manufacturing, tanning, textile manufacturing and washing, and electroplating and metal finishing industries. Industries in Gaza are light, each employing 5 to 100 individuals (Shomar et al., 2004).

The main objective of this study is to identify the sources and the build up of elevated AOX in the sludge in Gaza.

2. Study area, materials and methods

2.1. Location of the study area

The study includes wastewater and sludge sample from the central wastewater treatment plant (WWTP) of the Gaza Strip, which lies to the southwest of Gaza City, and from four light industries that discharge wastewater to the plant. The plant was originally constructed in 1977 as a two-pond treatment system. In 1986, it was expanded to a capacity of 12000 m³ day⁻¹ with the construction of two additional ponds. A project in 1994 rehabilitated the plant without increasing capacity. In 1999, with USAID funding, the plant was expanded to a capacity of 32000 m³ day⁻¹ and consisted of anaerobic ponds, an aerated pond, biotowers, an effluent polishing pond, disinfection, effluent pump station/force main and sludge drying beds (Fig. 1). The current flow to the plant is about 42000 m³ day⁻¹ from Gaza City and parts of Jabalia.

2.2. Description of the treatment plant

The treatment process at Gaza WWTP (Fig. 1) starts with pumping the wastewater to a single inlet chamber. Flows from the inlet chamber pass to the first sedimentation

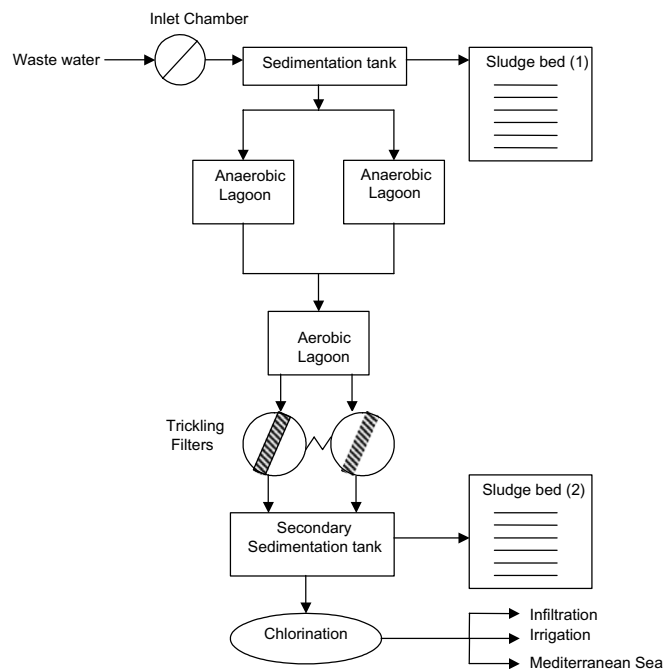


Fig. 1. Major units of Gaza wastewater treatment plant.

tank, and then pass to two anaerobic lagoons. The two anaerobic lagoons have a combined capacity of 46 400 m³ working parallel or in series. The retention time at average daily flow is 2.9 days. From the anaerobic lagoons, flows pass to a single aerobic lagoon of 43 700 m³ capacity with a retention time of 2.73 days. From the aerobic lagoon, effluent passes to two trickling filters then to a secondary sedimentation tank, then to a pumping station where flows are pumped to a disinfection unit (chlorination) which is currently not working. About 70% of the treated effluent flows to the Mediterranean Sea, 26% infiltrates to the groundwater, and 4% is used to irrigate a farm of two hectares that cultivates olive and citrus trees. Sludge is removed through a 150 mm suction pipe draped along the existing 3:1 slope to a suction pump. The pump discharges sludge to an anaerobic holding pond with a capacity of 10 700 m³. Sludge treatment facilities are almost absent and the sludge produced (450 000 ton year⁻¹) is removed from the ponds and exposed to the sun and then accumulated and transferred to the central solid waste dumping site.

2.3. Description of light industries

AOX is a measurement for halogenated organic compounds and is an important parameter for the characterization of industrial wastewaters. The selected industries in Gaza were the jeans washing industry, detergents industry, textile industry and textile washing industry. The field surveys showed that these industries are light with no treatment facilities. Their effluents are being discharged to municipal sewage system and the existing treatment plants are capable of absorbing the industrial effluents with no significant impact on treatment bioprocesses. These industries were using old technologies and they represented the largest consumer of chlorine (Cl₂) and are expected to be the dominant source of toxic organochlorine compounds discharged directly into waterways. Large quantities of toxic organochlorine byproducts and thousands of other

substances were being discharged into the municipal sewage system from small chemical and pharmaceutical, agricultural industries and hospitals. Many organochlorines resist natural breakdown processes, so they build up over time in the sludge, and this may explain the high AOX ratio in the sludge of Gaza.

2.4. Sampling

Local personnel trained by the author conducted the sampling campaign in December 2006. The sampling locations were correlated with the wastewater treatment process. The average depth of wastewater at the sampling sites was 30 cm. Eight to ten wastewater grab samples were taken from each of 12 locations within the treatment plant (Table 1). The grab samples from each location were combined to form a composite sample. Both pH and dissolved oxygen (DO) were measured on site while the chloride (Cl⁻) was measured in the laboratory. Finally, 250 ml of the mixture was taken in an acid-washed bottle and transferred to the laboratory, where it was filtered in an acid-washed filter holder through 0.45 µm pore size Sartorius membrane filters; the first few milliliters were used for rinsing, then discarded, and the remaining filtrate was transferred to clean acid-washed polyethylene bottles and acidified with concentrated nitric acid (Ultrapur, Merck, v/v) to pH < 2. The samples were transported to Germany for AOX analysis.

Sludge samples were collected in polyethylene containers from the same locations as wastewater samples when applicable. Two sludge samples that had been drying for three months were collected from the drying areas and five samples were collected from the sludge amended soils in the surrounding areas. Four wastewater and four sludge samples were collected from each of the industries previously described (Table 1). After collection, sludge samples were freeze-dried; then they were ground and homogenized in an agate mortar and sieved through a mesh of 63 µm pore size.

Table 1
Major parameters of Gaza wastewater treatment plant (2003)

Parameter	Unit	Influent	Anaerobic pond no 3	Trickling filter	Aerobic pond	Settling channel	Polishing pond
pH		7.5	6.77	6.81	7.64	–	7.79
Temp.	°C	25.9	25.5	25.4	25.5	–	24.9
Sett. Solid	ml l ⁻¹	7	2.5	1.5	5	–	–
TS	ml l ⁻¹	2184	2092	2040	2040	1900	1924
TDS	ml l ⁻¹	1751	1748	1842	1820	1820	1867
TSS	ml l ⁻¹	433	344	198	220	80	57
TVSS	ml l ⁻¹	340	236	122	148	52	28
NO ₃ -N	ml l ⁻¹	0	0	0	5.5	2.5	1.5
NH ₃ -N	ml l ⁻¹	32	43	48	46	40	43
N-KJD	ml l ⁻¹	64	77	94	64	–	56
Cl ⁻	ml l ⁻¹	550	600	600	600	580	560
PO ₄ ³⁻	ml l ⁻¹	6	9	9.3	9.6	11	11
BOD ₅	ml l ⁻¹	390	300	280	150	70	35
COD	ml l ⁻¹	806	680	496	431	208	102

Source: Adapted from Gaza Municipality.

2.5. Determination of AOX

AOX was determined in wastewater using a Euroglas Organic Halogen Analyzery-Netherlands according to the German standard methods DIN 9562 H14, sludge and sediment (Group H) (DIN, 2006). In short, 100 ml of the wastewater sample was placed in an Erlenmeyer flask. Fifty mg of activated carbon and 10 ml acidified nitrate solution were added to the flask and the suspension was shaken for 1 h. Milli-Q water was used as blank samples. The suspension was filtered through a polycarbonate filter and rinsed with an acidic nitrate solution. The filter with the filter cake were then combusted in an oxygen stream at 1000 °C. The resulting hydrohalides were transferred to a titration cell where they were titrated microcoulometrically. The sludge samples were analyzed according to the DIN 38414 S 18 (DIN, 1990). For quality control, duplicated samples were analyzed with three readings for each and finally the average value was recorded.

3. Results and discussion

Table 1 summarizes major parameters of the wastewater treatment plant's performance in 2003. Shomar et al. (2004, 2005) provides additional detail on more than 32 parameters for the same treatment plant. Other supporting parameters related to the AOX production such as chemi-

cal oxygen demand (COD) are also shown in Table 1. Table 2 shows the results of AOX in both wastewater and sludge, as well as the DO, pH and Cl for each sampling location. Comparison between AOX in wastewater and sludge for the same treatment locations mentioned in Table 2 is shown in Fig. 2.

There was no significant variation in the values of pH and Cl in the treatment units. DO shows some variation especially in the aerobic and anaerobic facilities. For statistical readings the Pearson correlation coefficient (r) was applied to measure the strength and direction of a linear relationship between the four variables (Table 2) and the numbers in parentheses represent the p -values or significance. The findings showed that there is a negative correlation ($r = -0.658$) between AOX in wastewater and sludge (at 10% level). The other correlations are not significant at 10% level. Correlation coefficient $r = 0.499$ ($p = 0.142$) was found between AOX in wastewater and pH. The AOX in wastewater and DO showed $r = 0.548$ ($p = 0.101$); while between AOX in wastewater and Cl, $r = -0.221$ ($p = 0.540$). The COD and AOX in wastewater showed $r = 0.375$ ($p = 0.285$). Finally, the Pearson correlation coefficient between AOX in wastewater and sludge was $r = -0.658$ ($p = 0.076$).

The Cl concentration in groundwater and wastewater of Gaza reached the average of 1000 mg l⁻¹ (Shomar, 2006) and it was found that the high Cl concentration in wastewater adversely affected the degradation rate of the paints

Table 2
AOX concentrations in wastewater and sludge of Gaza (October 2004 and December 2006)

No.	Wastewater treatment plant	pH	DO (mgO ₂ l ⁻¹)	Cl (ml l ⁻¹)	COD (mgO ₂ l ⁻¹)	Wastewater AOX (μg l ⁻¹) 2006	Sludge AOX (mg kg ⁻¹) 2006
1	Inlet chamber	7.91	0.78	913	1382	85	–
2	Sedimentation tank (in)	6.68	0.14	641	1020	71	213
3	Sedimentation tank (out)	6.97	0.13	504	980	123	39
4	Anaerobic lagoon (1)	7.04	1.0	566	690	86	271
5	Anaerobic lagoon (2)	6.9	0.39	613	650	117	280
6	Sludge bed	–	–	–	–	–	356
7	Aerobic lagoon (in)	7.83	4.03	593	340	458	24
8	Aerobic lagoon (out)	7.79	2.97	600	135	132	340
9	Settling channel	6.59	0.46	620	190	34	–
10	Sludge holding pond	–	–	–	–	121	105
11	Secondary sedimentation tank (in)	–	–	–	–	51	–
12	Secondary sedimentation tank (out)	7.73	0.39	600	109	402	100
13	Effluent polishing pond	7.97	0.31	686	102	106	379
14	Dried sludge three months old	–	–	–	–	–	129
15	Dried sludge three months old	–	–	–	–	–	121
<i>Industrial effluents</i>							
16	Jeans washing industry	–	–	–	–	38 486	513
17	Detergents industry	9.55	7.69	3449	–	42 874	255
18	Textile industry	8.44	6.33	470	–	84	105
19	Textile washing industry	6.89	0.94	1370	–	346	160
<i>Soil samples</i>							
20	Soil irrigated by WW	–	–	–	–	–	17
21	Soil beside dry sludge	–	–	–	–	–	6
22	Soil beside plant effluent	–	–	–	–	–	32
23	Soil behind drying lagoons	–	–	–	–	–	29
24	Soil beside plant entrance	–	–	–	–	–	17

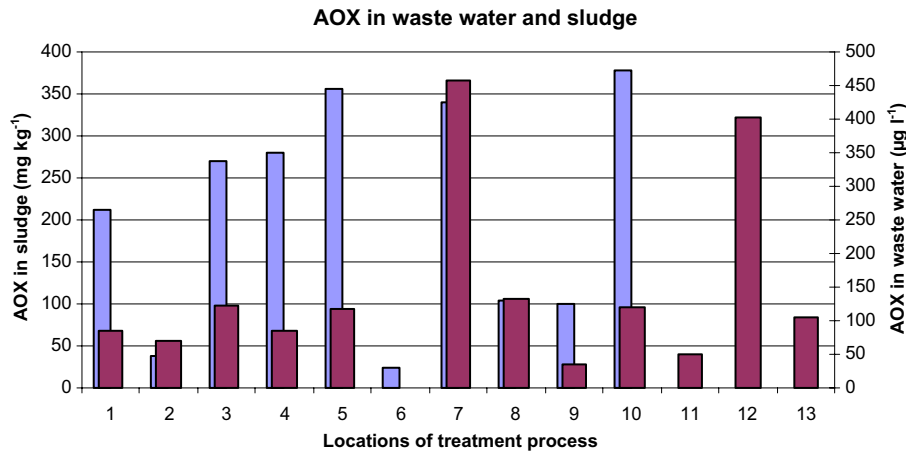


Fig. 2. Comparison between AOX in wastewater and sludge for the same treatment locations.

and dyes (Yang et al., 2005). Low chloride concentrations in wastewater corresponded to low concentrations of AOX, and the high chloride concentrations correlated with high AOX (Emmanuel et al., 2004). The results show that there is no significant variation in the values of pH and Cl in the treatment units. DO shows some variation especially in the aerobic and anaerobic facilities. The production of AOX is strongly connected to the presence of oxygen and specifically to the oxidizing agent. Organic carbon in wastewater represents the substrate of AOX production in the presence of oxidizing agent and Cl⁻. The high concentration of Cl⁻ in wastewater influent of Gaza (913 mg l⁻¹) enhances the production of AOX; keeping in mind that groundwater of Gaza—which is the only source of water—is salty. Baycan et al. (in press) found that the Cl concentration in the wastewater and the chemical structure of the substances are more important than the pH of the wastewater for AOX production. This may imply that OH-radicals oxidize some chloride ions to form chlorine, which further reacts with organic compounds so that AOX *de novo* is produced; eventually, these AOX compounds are destroyed. On the contrary, several studies stated that the pH plays the important role on the decomposition of halogenated compounds (Thang and Hung, 1996; Lu et al., 1997). The wastewater influent is alkaline (pH = 8) which becomes acidic in the anaerobic conditions (pH < 7). It is believed that the AOX concentrations in the anaerobic lagoon decreased to 86 µg l⁻¹ as a result of the destruction of carbon halogen bonds by the attack of sulfide (S²⁻) and hydrosulfide ions (HS⁻). However, the results of this study show that the pH of the Gaza wastewater has no significant impact on the AOX production. As the treatment plant is partially functional and dependent mainly on natural treatment and partially on non-functional biological treatment. It is believed that the absence of correlation between DO, pH and chloride concentration and the AOX in both sludge and wastewater is due to the absence of sludge treatment facilities. Sludge is exposed to

the sun and then accumulated and transferred to dumping sites. The AOX concentration in the three-month-old sludge was 130 mg kg⁻¹.

The political and socio-economic situation in Gaza has affected the water sector including the performance of the wastewater treatment plants (Bohannon, 2006). Industrial activity has decreased by 70% in 2006 compared to the previous four years. Consequently, the AOX in the industrial effluent (Table 2) decreased from 150 000 to 40 000 µg l⁻¹ over the same time period for the same industries. Moreover, sludge from these industries showed 1300 mg kg⁻¹ AOX for the past four years and only 400 mg kg⁻¹ for the year 2006. Generally, the AOX in the influent wastewater to the treatment plant decreased from 320 to 85 µg l⁻¹ for the same period.

4. Conclusions

1. Regardless of the AOX concentrations in the wastewater influent and the treatment efficiency, the AOX in the produced sludge is formed predominantly inside the wastewater plant.
2. There is no correlation between the AOX in wastewater and sludge. Moreover, there is no correlation between the AOX in wastewater/sludge and pH, DO and Cl in wastewater.
3. The AOX in the influent/effluent wastewater and subsequently produced sludge decreased dramatically in 2006 compared to the previous years most likely due to declining industrial production as a result of the political and socio-economic situation in the Gaza Strip.
4. The AOX concentration in sludge exposed to the sun for three months was reduced to two thirds of its original concentration. To reduce potential environmental and health hazards, it is recommended that sludge be exposed to the sun for several months before using it in agriculture.

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