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Investigation of the presence of halogenated bi-products formation in the drinking water network in Tulkarm District / Palestine

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Abstract: The problem of pollutants in drinking water networks is neglected in many places all over the developing countries. This problem is normally caused by either direct pollution source such as organic carbon, or from the maceration pollutants of network materials. The heavy metals in the network facilities and the DOC from the wastewater leakage on the formation of toxic by-product in the water network and the rate of halogenated hydrocarbons formation in the drinking water network was studied. Results showed that water has the same constituents of pollutants similar to that before its being stored for relatively long periods. The results showed also that the trend of halogenated hydrocarbons formation is correlated, but not restricted, to the availability of total organic carbons. The amount of CCl_2Br and $CClBr_2$ were the highest, which indicate that most of the halogens are originated from natural sources. The Strontium values where the most dominant in all sampling points followed by Barium and Boron, which are the most abundant trace metals normally found in the groundwater in Tulkarm area

***Keywords*: ** Drinking water, Halogenated Hydrocarbons, Toxic metals, Tulkarm, Total Organic Carbons.

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Introduction

Organic Carbon and halogenated bi-product

A halogenated hydrocarbons (e.g. Trihalomethanes and halothane) are hydrocarbons in which one or more hydrogen atoms is replaced with a halogen atom such as chlorine or Bromine. Trihalomethanes THMs are disinfection by-products (DPDs) of post-chlorinated water. They result from reaction between chlorine and/or bromine and natural organic matter (NOM) (Marhaba and Washington, 1998; Chen and Weisel, 1998; Morris And Baum, 1998; Nikolaou and Lekkas, 2001)

THM formation depends on various parameters such as pH, temperature, bromide, natural organic matter (NOM), and exposure to ammonium and chlorine (Najm et al, 1994). When critical levels of chlorine or bromine are reached in the presence of chemical oxygen demand (COD) or total organic carbon (TOC), THM and chloramines are formed (Gallard and von Gunten, 2002). Chlorine reacts with inorganics (e.g. bromide) during disinfection forming hypobromous acid that rapidly reacts with THM organic and substantially increases THM molar yields . Brominated THMs occur at higher concentrations when water with high bromide concentration is chlorinated (Najm et al, 1994). Moreover, high Br^-/DOC and Br^-/Cl_2 favour the formation of brominated THM (THM-Br) over chlorinated THM (THM-Cl) (Huan et al, 2010). Therefore, the ratio of [Br/Cl] regulates the composition and concentration of THM (Nobakawa and Sanukida, 2001).

The US Environmental Protection Agency (EPA) has promulgated regulations for THMs because of harmful and adverse health effects caused by these halogenated by-products on animals and humans. They have been known to cause several types of cancers, liver and kidney damages, as well as reproductive disorders in humans (Arora et al, 1997; Hsu et al, 2001; USEPA, 2001; Bull et al, 2011; Narotsky et al, 2011). These carcinogenic and mutagenic by-products formed in water pose a serious health risk to end-users (Marei et al, 2010), thus drinking water regulations suggest a maximum total concentration of 80 ppb of THM (Nobakawa and Sanukida, 2001). According to World Helath Organisation report in 1993 the maximum acceptable concentration of THMs in drinking water is 0.100 mg/L (100 µg/L), while the maximum acceptable concentration of Bromodichloromethane (BDCM) in drinking water is 0.016 mg/L (16 µg/L). While the maximum limit allowed by the WHO for the DOC in dronkiung water is 5 mg/L.

Polluted water resources in the Occupied Palestinian Territories is a major challenge facing domestic water suppliers. About 12 MCM wastewater drained through Wadi Zomar from Nablus city and other Palestinian cities and villages to the west, as well as some Israeli settlements.

Many previous studies upraise the effect of septic tanks commonly used in Tulkarm area and Wadi Zomar on the Groundwater quality in Tulkarm area. (Khayat et al, 2012; Abu Khalaf et al, 2013). One of the common source for increasing the Dissolved Organic Carbon (DOC) (2-16 mg/L), and high nitrate values (15-82 mg/L) is resulted mainly from the sewage leakage.

Study Area

Tulkarm is located in the north western part of the West Bank (**Figure 1**). The total area of Tulkarm district is about 246 km². Its ts current population is estimated at 167,000 people, representing about 12% of the total population of the West Bank (PCBS, 2010). The number of people living in the rural areas exceeds 55% of the total population. The population density in Tulkarm area is about 679 persons/km² (ARIJ,1996).
The city receives most of its drinking water after stored in 3 main water resrvoirs that are connected to the water distributed network, some of those reservoirs are more than 30 years old and located on an elevated place in the town. In recent years, the city suffered from water shortage(PWA, 2011).

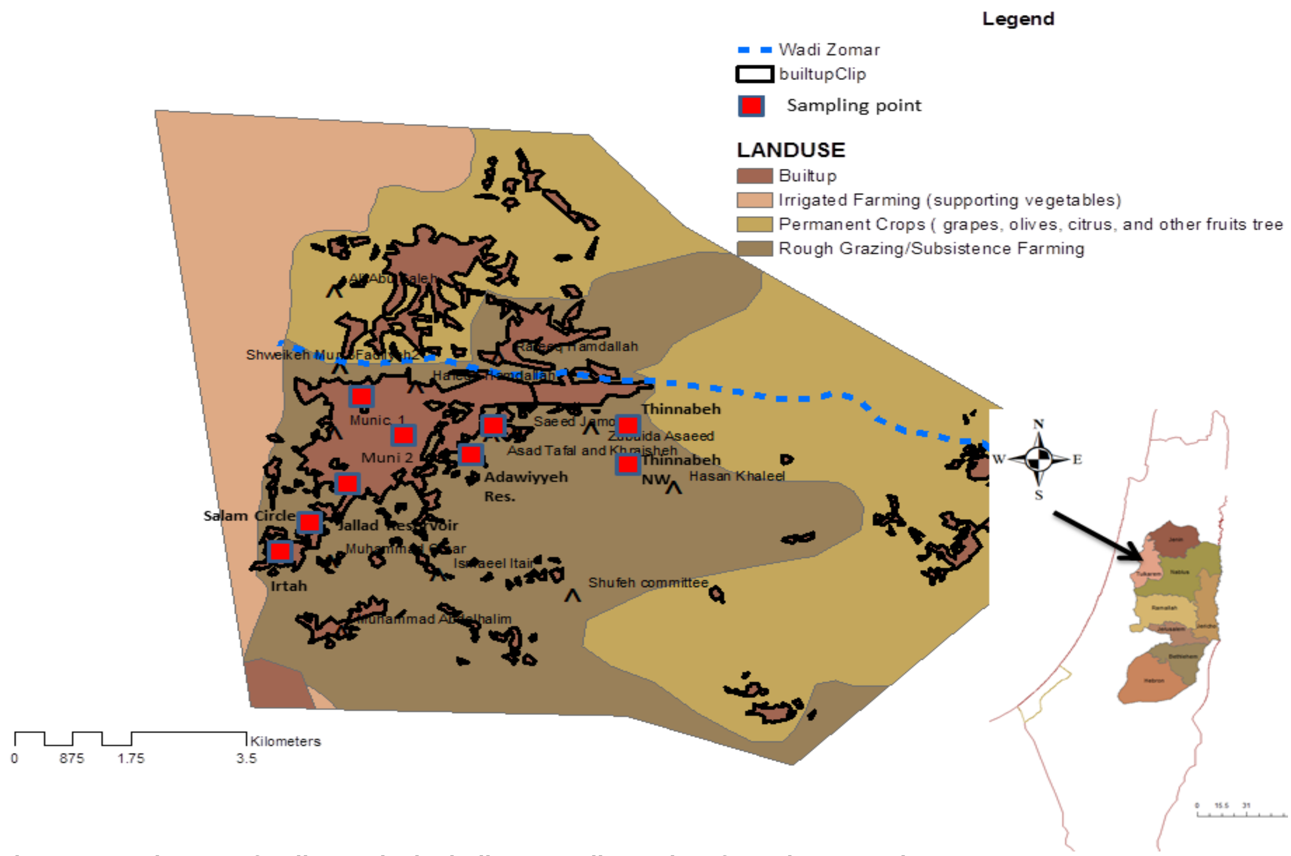

Figure 1: Study area of Tulkarm city including sampling points from the network.

Despite that, the UNDP project rehabilitated the old tank and built a new tank, the city still suffers from many water problems... Water is pumped from the two main water wells on the western side of Tulkarm to the main reservoirs where it stored, and then flows to the surrounding houses, serving around 25,000 people . New piping lines were re-constructed to ensure effecient and safe water service system in the city.
During 2002, the underground water pipe lines were destroyed by the invasion of Israelis to the city causing a severe water pollution as a result of mixing fresh water with wastewater (PWA,2011).

The aim of this work was to determine the impact of DOC from the wastewater leakage on water network contents and assess the rate for halogenated hydrocarbons formation in the drinking water network.

Material and methods

During the end of winter season 2012/2013, water samples from different locations along the drinking water network in Tulkarm city were collected. These locations include: the main pumping wells that supply water for the whole network (Well nr. 1 and Well nr.2) (**Figure 1**), reservoirs in Adawiyeh, Thinnabeh and and Aljallad different network points near houses.

Two samples were taken from 8 sampling point in glass bottles, one for Dissolved Organic Carbon (DOC), and the other for the Tri-halomethanes (THMs).

For the purpose of sampling THMs and DOC in water, according to the standard method 5710 B (APHA, 2005). In order to preserve samples, dark glass bottles which were washed and rinsed with distilled and dried in an oven at 500ºC for 5 hours water were used,. Caps with Teflon liners, which also were washed and abundantly rinsed with distilled water were also used. For the trace and heavy metals, few drops of concentrated HCl to lower the pH were added and the samples were wraped with aluminum foil. The organic carbon were measured using 680°C Combustion Catalytic Oxidation Method, while the halogenated hydrocarbons were measured using headspace gas chromatograph. All analysis were done in CTM-Manresa Technology Centre in Spain.
(Sugimura and Suzuki, 1988)

Results and Discussion

Formation of halogenated hydrocarbons

The measured values of halogenated hydrocarbons formed in the water network and source wells in the study area are shown in **Table 1**.

Except in the reservoirs where drinking water stored for several days which range from 4-7 days, most of the sampling points showed conservative behaviors between the Total Organic Carbon (TOC) values and the NO_3 values. These behaviors indicate that the water have the same constituents of pollutants, before it is stored in the storage reservoirs for relatively long period of time. This period is enough to keep the water in direct contact with the reservoir old pollutants and organics which accumulate and adhered on the walls and bottom. This finally leads to additional TOC contents that can mix with drinking water. In this context, the TOC value from Al-Jallad and Adawiyeh reservoirs with capacity of 1200 CM and 1500 CM respectively), which are the oldest reservoirs in Tulkarm area, emphasize this hypothesis, where it is supposed to contain the highest amount of organic pollutants due to the time factor (**Figure 2**). The higher values of nitrate suggest that the presence of septic tanks that leaked to the water resources.

Table 1: Values of halogenated hydrocarbons formation in µg/l from the sampling points in the study area.

Reference	CCl3	CCl2Br	CClBr2	CBr3
PTU 1	< 5	< 5	< 5	< 5
PTU 2	< 5	< 5	< 5	< 5
PTU 3	47.7	49.6	51	45.7
PTU 4	< 5	< 5	< 5	< 5
PTU 5	< 5	< 5	< 5	< 5
PTU 6	< 5	< 5	< 5	< 5
PTU 7	35	39	37	31
PTU 8	< 5	< 5	< 5	< 5

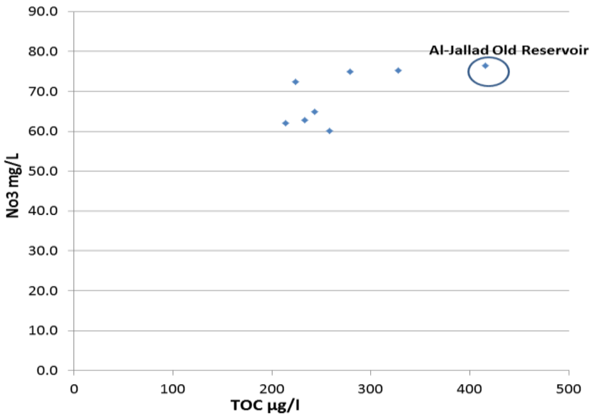

Figure 2: The Total Organic Carbons (TOC) Vs. the Nitrate contents from sampling points.

Interestingly, lower organic pollutants were found Thinnabeh reservoir which might be due to the relatively new renovation of this reservoir (less than 2 years). The relation between the electrical conductivity (EC) and the TOC (**Figure 3**). While most of the samples showed a constant electrical conductivity with TOC, Al-Jallad old reservoir showed a high EC values which is related to the high amount of accumulated pollutant inside the reservoir. The variation of the TOC amount in these samples is subjected to the conditions of different distribution system facilities, or pumping conditions. Moreover, the constant EC suggest the contribution of organics which are present in the water reservoirs rather any other type of inorganic pollution.

On the other hand, Thinabeh reservoir showed low EC value and pollutant values, this might be related to the recency of the building and the closeness of the reservoir to the pumping station.

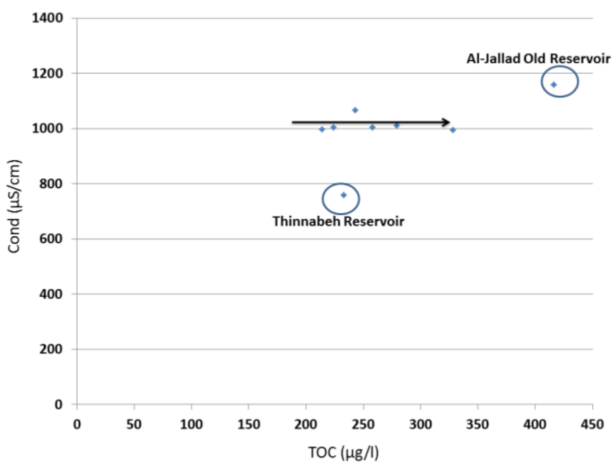

Figure 3: The Total Organic Carbons (TOC) Vs. Electrical Conductivity (EC).

Figure 4 shows that the trends of halogenated hydrocarbons production are correlated, but not restricted, to the availability of total organic carbons. In two sampling points the amount of halogenated hydrocarbons where relatively high, this is also might be related to the conditions of storage, and the media where water path through. However, the amount of natural and added chloride also plays a role in this regard. Moreover, between all halogenated carbon forms the amount of CCl_2Br and $CClBr_2$ were the highest, which indicate that most of the halogens are originated from natural sources. In this context, the amount of free chlorine may play a role in the process of halogenated hydrocarbons formation, is not expected to occur at the end of winter season where the dilution of water takes place. Moreover, the municipality add small amount of chlorine which is expected to have low potential in reaction that leads to low formation of THMS in the network .

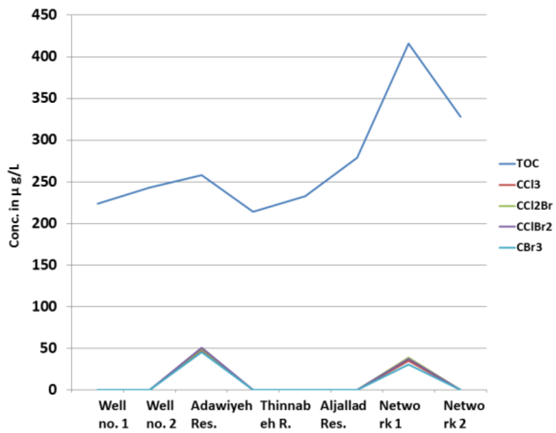

Figure 4: The trend of halogenated hydrocarbons hydro-carbons products formations.

Heavy metals in water resources and network.

Several studies showed that trace metals are abundant in groundwater from Tulkarm area (Khayat et al, 2013). In spite of that the samples were collected after a reasonable time of flush out, samples from public networks showed relatively high values of Zinc, Nickel, and Cupper (**Table 2, Figure 5**). This might be due to the fact that parts of the network facilities are old (over 20 years old) and need to be replaced in many places in the city.

The natural constituents of the other metals behave in the water were the same as the water from the wells and reservoirs. Strontium was the most dominant in all sampling points with values ranging between 400 to 500 µg/l followed by Barium and Boron with values varied between 160-190 µg/l and 40-95 µg/l respectively.

Table 2: Heavy metals concentrations (μg/l) in water samples collected from different local points in Tulakarm city.

Reference	B	Al	Mn	Fe	Co	Ni	Cu	Zn	Se	Sr	Cd	Ba	Gd	Pb
PTU 1	80	< 5.6	< 1.1	< 5	< 0.1	1	1	< 5	12	482	< 0.1	186	< 0.1	< 0.6
PTU 2	95	< 5.6	< 1.1	< 5	< 0.1	2	3	< 5	13	504	< 0.1	189	< 0.1	< 0.6
PTU 3	85	< 5.6	< 1.1	< 5	< 0.1	2	3	6	11	473	< 0.1	186	< 0.1	< 0.6
PTU 4	84	< 5.6	< 1.1	< 5	< 0.1	1	6	13	13	473	< 0.1	187	< 0.1	< 0.6
PTU 5	43	< 5.6	< 1.1	< 5	< 0.1	0	3	7	13	266	< 0.1	160	< 0.1	< 0.6
PTU 6	82	< 5.6	< 1.1	< 5	< 0.1	4	7	42	12	493	< 0.1	186	< 0.1	1
PTU 7	72	< 5.6	< 1.1	< 5	< 0.1	2	4	33	12	412	< 0.1	154	< 0.1	< 0.6
PTU 8	79	< 5.6	< 1.1	< 5	< 0.11	1	26	47	11	484	<0.11	185	< 0.11	1

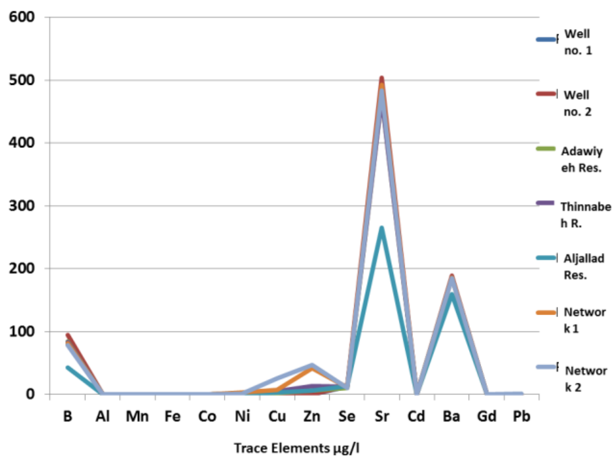

Figure 5: The amount of trace metals in the water resources and water samples from the network.

Recommendations

From the research results, it can be concluded that:

- Some of the network facilities, boosters, and storage tanks are relatively old and need to be replaced, this can be noticed through the relatively high amount of TOC, THMs, and heavy metals, in some sampling points.

- Further attention about the consequences of chlorination process must be studied from temporal term.

- The water storage reservoirs must be cleaned, and a sufficient amount of continuous pumping must be ensured to avoid long water persistence in the storage reservoirs.

- Further monitoring of groundwater pumped from wells in the district must be done, in order to avoid any sudden increase in the amount of organics especially at the end of winter season where the thunder storm occurs. In this case, simple Pre-treatment for the groundwater might be required before pumping the water to the network.

- Other detailed studies are still needed in order to assess the potential of THMs formations, and related factors that paly roles or accelerate the formation of THM spatially and temporally.

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