



Thesis Approval

Total Organic Carbon and Formation Potential of Trihalomethanes In
Spring Water in Jericho Area
Jericho, Palestine

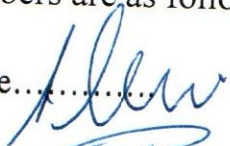


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Abstract

Many of the problems associated with water quality control are due to the presence of organic matter and nitrogen compounds from natural source or from waste water discharge. Chlorine is still the most commonly used disinfectant in many water treatment plants all over the world. However, studies conducted demonstrated that the chlorine reacts with the natural organic matter (NOM) in source waters, and leads to the formation of variety of chlorinated organic compounds, disinfection by products (DBPs). Among these, the most common are the trihalomethanes (THMs) which exhibit potentially carcinogenic activity.

This study was conducted in order to determine the concentrations of both TOC and TN during one hydrological season; during long term monitoring system can be established to trace the variable changes in the concentrations along the winter seasons in the area. In order, collectively, to find the best solutions for controlling the formation of natural hazardous by-products in the drinking water, mainly the THMs.

Samples were collected from available springs in Jericho during August 2009 and April 2010. Raw water from Ein-Sultan, Ein-Dyouk, Nuaima and Al Quilt canal were collected. Multi N/C 2000 analyzer from Analytik Jena AG was used for TOC (NPOC) and TN analysis.

Other analysis was focused on the determination of THMs in the urban net work, in order to study the formation of trihalomethanes THMs in drinking water of the study area. For this effort, sample collection period was divided into two stages. The first stage of sample collection took place during the month of October 2009, which is representative of summer month. The second stage of sample collection was undertaken during the month of January 2010, which is representative of winter. Liquid-liquid Extraction and Gas chromatography with electron capture detection method and Head Space with Gas Chromatography-Electron Capture Method were used for THMs analysis. The average Non-Purgable Organic Carbon (NPOC) in four springs and Al Quilt canal was 4.1 mg/L with concentrations ranged between 0.2 and 18.4 mg/L. The average minimum and maximum values of TN in the examined spring fluctuated between 2.8 mg/L in summer at Al Quilt canal and 7.2 mg/L in winter at Sultan spring.

Total trihalomethanes (TTHMs) in tap water were below the MCL of WHO and USEPA. The average values of TTHMs for Jericho and Aqbat Jabber camp during sampling periods summer and winter was (3.1 $\mu\text{g/L}$), (16.8 $\mu\text{g/L}$) and (7.1 $\mu\text{g/L}$), (6.9 $\mu\text{g/L}$), respectively.

Results of residual chlorine shows that average total and free chlorine measured from Jericho and Aqbat Jabber camp during summer and winter periods were (0.15-0.15), (0.29-0.25) and (0.23-0.22), (0.20-0.19) mg/L respectively. These values are less than the recommended values by the WHO required for efficient drinking water distribution. Such low concentration shows that the chlorine dose added to the net work through a process of disinfection is not sufficient for effective disinfection processes. High temperature conditions in Jericho area may properly promote the accelerated depletion of residual chlorine which mitigates THM formation.

The results for the THMFP in Ein Sultan and Al Quilt canal, before and after sand filtration show that, the total trihalomethanes exceeded the WHO guideline (100 $\mu\text{g/L}$) and the maximum contamination level of USEPA (80 $\mu\text{g/L}$) at different time period, according to incubation temperature for each sample. Results have shown that THM formation increase with increasing time and higher chlorine dose and both these parameters will increase the THM concentrations. Reaction time is one of the most important factors affecting their formation because investigation of the influence of reaction time on THMs formation is critical in order to determine the final concentrations to which people could be exposed.

The results of this study will be valuable to the Palestinian Water Authority (PWA) in defining the chlorination process efficiency and THMs regulation. Findings are also beneficial for the Palestinian Ministry of Health.

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Chapter One

Introduction

1.1 Background

Ground water quality could be influenced by many factors, including: climate, topography, aquifer lithology, surface water recharge and human activities (Wu et al., 2005). Total organic carbon (TOC) and total nitrogen (TN) measurements are used as indirect water quality parameters to determine the water quality of water sources used for drinking water.

TOC is considered an indicator of natural organic matter (NOM) such as: humic acid, fulvic acid and amines, which combine with chlorine added to the water for disinfection purposes of drinking water and leads to the formation of many by products potentially harmful to human health (Reckhow et al, 1990). Among all the chlorination by-products, trihalomethanes (THMs), which exhibit potentially carcinogenic activity (Tokmak et al, 2004). Classical trihalomethanes consist of chloroform (CHCl_3), dichlorobromomethane (CHCl_2Br), dibromochloromethane (CHBr_2Cl) and bromoform (CHBr_3). They are formed through the reactions of hypochlorous acid (HOCl) with natural organic matter (NOM) in presence or absence of bromide (Gallard and Guten, 2002).

Many studies have been conducted in the past three decades on the health effects of Disinfection By-Products (DBPs) in drinking water supplies (Morris et al, 1992; Tokmak et al, 2004). Although THMs are suspected probable carcinogenic and their concentration in water should be monitored. Surrogate parameters such as TOC, UV (A) $_{254\text{nm}}$, and THMFP are used to predict or monitored them. In 1999 TOC was selected by (US.EPA) as a surrogate measurement to control (DBPs) in drinking water, in order to protect consumers from potentially carcinogenic by-product of disinfection processes of drinking water. In order to minimize the formation of such harmful (DBPs), the parameters affecting their formation should be understood. THM formation has been shown to be a function of many water quality parameters, including TOC, pH of chlorination, temperature, and contact time, Bromide and chlorine dose.

About 5 to 10% of carbon in natural water occurs in organic forms (Volk et al, 2002). The combined total of these forms is known as (TOC), which is a regulated water quality parameter. TOC varies from around 1 to 10 mg/L as carbon in surface waters (Mcphee,

1992). Ground water typically has less than 1 mg/L TOC (Dunalska et al, 2004), although ground water residing in sediments with high organic content has more. Humic substances comprise up to half the TOC and are thought to be polymeric molecules formed from condensation reactions of plant lignin, polysaccharides and/or proteins, although direct determinations of molecular forms have been illusive (Loudon et al, 2003). These substances are most commonly observed in ground water. Non-humic substances are typically lower molecular weight and simpler compounds derived from various sources.

TOC in water can originate from various sources. The fraction of organic matter that derives from biota growing in the water bodies is classified as internal natural organic matter and includes humic acid, fulvic acid and amines. While the external organic matter which enter the water stream from watersheds, degradation of terrestrial vegetation and soil leaching during run off, these include detergents, pesticides, fertilizers, herbicides, industrial chemicals are example of synthetic source (Dezuane, 1997 ; Volk et al, 2002). TOC occurs in the form of dissolved organic carbon (DOC), particulate organic carbon (POC) and volatile organic carbon (VOC). The dominant form of organic carbon in the inland waters is the dissolves from (Dunalska et al, 2004). High levels of TOC in water are of concern because they play an important role in many biogeochemical processes. TOC also strongly affects the transport of contaminants, such as: heavy metals from soil to surface water, changes water pH, create complexes of phosphorous compounds, metal cation and toxic substances (Loudon et al, 2003; Dunalska, et al, 2004).

TN is the total content of nitrogen species of organic nitrogen, ammonia, nitrate (NO_3^-) and nitrite (NO_2^-) present in water or waste water. TN data is used when nitrate is not available, so it is assumed to be a surrogate measurement for nitrate. Nitrate is a problem as a contaminant in drinking water, primarily from ground water and wells due to harmful biological effects. Toxicity of nitrates is demonstrated by methemoglobinemia for infants (Comly, 1987; Dezuane, 1997). Previous study has demonstrated that organic nitrogen (organic-N) compounds are precursors of some nitrogen- containing DBPs like haloacetonitrils (HANs) in chlorination. Active chlorine atoms can also be transferred from monochloroamine to organic-N compounds in aqueous solutions (Snyder and Margerum, 1982; Isaac and Morris, 1993). The transfers lead to the formation of organic chloramines and then DBPs.

The principle sources of nitrogen input include: soil mineralization, fertilizer, legumes, pasture animal manure and municipal and industrial point sources (Batzika et al, 2007). Research has demonstrated the significant contribution of agriculture to rising concentrations of nitrate in ground water, especially in urban area (Kingsbury, 2008).

1.2 Problem Statement

Safe and good quality drinking water is the basis for good human health. Water provides essential elements but, when polluted it may become the source of an undesirable substance dangerous to human health. Many of the problems associated with water quality control are due to the presence of organic matter and nitrogen compounds from natural sources or from waste water discharge (Tebbutt, 1998). In Jericho, spring water is the major source for drinking and agricultural use. Ein-Sultan Spring is the main source for drinking water in Jericho. High concentrations of TOC were measured in spring water (Sultan and Dyouk) (Khayat et al, 2008). With chlorination, it is possible to form THM. Many factors play roles in the process of THM formation, mainly the high temperature, as well as the dose of chlorine which applied as disinfectant to the spring water to prevent the growth of microorganisms in the drinking water distribution system, thus protection the public from water borne disease. Chlorination, which is a widely used disinfection process, in 1974, was first reported by Rook to cause the formation of (THMs). The presence of chlorination disinfection by-product CDBP, in the drinking water is of concern from a public health aspect, as they are suspected to be carcinogen (Morris et al 1992; Tokmak et al, 2004). Also, drinking water from a spring in an agricultural area in Jericho shows evidence of pollution with nitrate, and this pollution correlates with intensive farming practices and man-made applications. Babies and infants living around the agricultural area who drink water from spring water are the most vulnerable to health risks from nitrate. Another spring group which is important as a drinking source is Wadi Al-Quilt Spring group, water from this group drain water to Aqbat Jabber refugee camp. Wadi Al-Quilt canal, drain water from Wadi Al-Quilt spring group through a 12 km open canal is used for drinking purposes, after using sand filtration. The recharge area of Jericho Springs extends westwards into an urban area where human activities are presence. Flood water and treated waste water which originate from Al Bireh municipality could be a source for TOC and TN in spring water.

1.3 Objective

The main objective is to determine the TOC and TN during one hydrological year; during long term monitoring system can be established to trace the variable changes in the concentrations along the winter seasons in the area.

Specific Objectives

1. To measure the TOC and TN in spring water and trace the factors which control its variable concentrations during a hydrological year.
2. To measure the free chlorine in the water net at different locations
3. To measure the THMs concentration in distribution system.
4. To determine the THMs formation potential (THMFP) of spring water in Jericho area.

1.4 Hypothesis

We developed four hypotheses to serve the objective. These are:

1. Water from springs gives variable levels of TOC and TN along the hydrological year, when THM is formed.
2. The concentrations of THMs formed in the water network vary, depending on the concentration of TOC in the discharged spring water.
3. Chlorination doses in parallel time of application play an important role in the formation of THM in drinking water in various concentrations.

1.5 Study Area

Jericho district is part of the Jordan Rift Valley, which is situated 250 m below sea level and is the lowest living town on the earth. Located on the eastern boundary of the West Bank, it extends from Fasayel in the north and from the eastern slopes of the Jerusalem and Ramallah mountains in the west to the Jordan River in the east (HWE, 2007) (Figure 1.1). The area within the municipality borders extends over 45 km² with an estimated population of 43,620 (PCBS, 2006). While it has a desert climate, Jericho is considered an oasis since it is watered by many springs. This characteristic makes it an important agricultural area, especially for fruits and vegetables. (Arij, 1995).

ملخص الدراسة

إن العديد من المشاكل التي لها علاقة بضبط جودة المياه مرتبط بوجود المواد العضوية والمركبات النيتروجينية في مصادر مياه الشرب والتي تأتي إما من مصادر طبيعية أو من تسرب مياه الصرف الصحي. على الرغم من إجراء العديد من الدراسات والتي اثبتت تفاعل الكلور مع المواد العضوية الطبيعية في مياه المصدر مما يؤدي إلي تشكيل مجموعة متنوعة من المركبات العضوية الكلورة، ومن بين هذه المركبات وأكثرها شيوعا مجموعة مركبات ثلاثي هالوجين الميثان (الهالوميثانات) والتي اثبتت الدراسات احتمالية تسببها بالسرطان.

تعتبر عملية التعقيم بالكلور الأكثر شيوعا في محطات معالجة المياه في جميع أنحاء العالم. في منطقة أريحا، تعتبر مياه الينابيع المصدر الرئيس لمياه الشرب، نبع عين السلطان المصدر الرئيس لمياه الشرب لمركز مدينة أريحا بينما مياه قناة واد القلط تعتبر مصدر الشرب لمخيم عقبة جبر للاجئين. حيث يتم تجميع المياه في تلك القناة المفتوحة بطول 12 كم من مجموعة ينابيع واد القلط وتزويدها للمخيم بعد استخدام تقنية فلتر الرمل. حيث أن منطقة التغذية ليناابيع منطقة أريحا تمتد غربا حيث تكثر الأنشطة البشرية.

تم إجراء هذه الدراسة من اجل تحديد تركيز كل من المركبات العضوية والمركبات النيتروجينية خلال موسم مطري واحد، لدراسة العوامل التي تؤثر في تركيزها خلال فصل الشتاء من اجل إيجاد حلول مستقبلية لمنع تشكيل المركبات غير المرغوب فيها الناتجة من عملية الكلورة.

تم جمع العينات من ينابيع منطقة أريحا : عين السلطان، عين النويعمة، عين الديوك وقناة واد القلط، خلال الفترة بين شهري أغسطس 2009 و ابريل 2010 وتم التركيز على فحص آخر وهو قياس الهالوميثانات في شبكة مياه الشرب لمدينة أريحا ومخيم عقبة جبر، حيث تم تقسيم عملية جمع العينات إلى مرحلتين ، المرحلة الأولى خلال شهر أكتوبر والتي مثلت فترة الصيف والمرحلة الثانية خلال شهر كانون الثاني مثلت فترة الشتاء. وقد أظهرت نتائج البحث أن معدل المركبات العضوية 4.1 ملغم/لتر، متراوحة بين 0.2 ملغم/لتر و 18.4 ملغم/لتر. وقد دل أعلى ارتفاع في تركيز المركبات العضوية في نبعي عين السلطان وعين الديوك في الشتاء على ارتباط التركيز بموسم المطر. وفي هذه الدراسة سجلت اقل وأعلى قيم للمركبات النيتروجينية بين 2.8 ملغم/لتر خلال الصيف في قناة القلط 7.2 ملغم/لتر خلال الشتاء في نبع عين السلطان.

وأوضحت النتائج أن الهالوميثانات تكونت بمعدل اقل من القيم المعتمدة من منظمة الصحة العالمية ووكالة حماية البيئة الأمريكية. وكان متوسط قيم الهالوميثانات لمنطقتي أريحا وعقبة جبر خلال

فترتي الصيف والشتاء بين (3.1 ميكروغرام/لتر)، (16.8 ميكروغرام/لتر) و (7.1 ميكروغرام/لتر)، (6.9 ميكروغرام/لتر)، على التوالي.

وبينت نتائج فحص الكلور المتبقي في شبكتي أريحا ومخيم عقبة جبر، أن متوسط الكلور الإجمالي والحر خلال فترتي الصيف والشتاء (0.15 - 0.15 ملغم/لتر)، (0.29 - 0.25 ملغم/لتر) و (-0.19 - 0.20 ملغم/لتر)، (0.22 - 0.23 ملغم/لتر)، على التوالي. هذه القيم اقل من القيم التي أوصت بها منظمة الصحة العالمية لكفاءة عملية تعقيم المياه الصالحة للشرب. القيم المنخفضة السابقة تفسران كميات الكلور المضافة خلال عملية التعقيم غير مضافة وفقا لاحتياج الماء الخام، وان ظروف الحرارة المرتفعة بالمنطقة تسارع من استنزاف الكلور المتبقي والتي تؤثر في عملية تشكيل الهالوميثانات في مياه الشبكة.

كانت نتائج فحص إمكانية تكون الهالوميثانات على مدار الأسبوع من كلورة المياه تجاوزت الحد المسموح به والمعتمد من منظمة الصحة العالمية (100 ميكروغرام/لتر) و (80 ميكروغرام/لتر) لوكالة حفظ البيئة الأمريكية في فترات زمنية مختلفة اعتمادا على درجة حرارة الحاضنة لكل عينة، فمثلا تجاوزت كمية الهالوميثانات الحد الأعلى المسموح به من هذه المواد في مياه نبع السلطان بعد 72 ساعة على درجة حرارة 19 س° وبعد 24 ساعة على درجتَي حرارة 25-38 س°، أما بالنسبة لمياه قناة القلط فقد تجاوزت الحد المسموح به بعد 6 ساعات على درجتَي حرارة 19-25 س°، وبعد ساعتين على درجة حرارة 38 س°. وهذا يفسر أن درجة الحرارة عامل مهم يؤثر بزيادة التركيز للهالوميثانات.

ستكون نتائج هذه الدراسة ذات قيمة لسلطة المياه الفلسطينية لتحديد كفاءة عملية الكلورة وتنظيم كمية الهالوميثانات، والنتائج أيضا مفيدة لوزارة الصحة الفلسطينية.

Chapter Six

Conclusion and Recommendation

6.1 Main Conclusions

The following conclusions can be derived from the results of the above studies:-

Spring water quality in Jericho area varies with time and distance from replenishment source. This is due to some other contribution of pollutants along the wadies path mainly, the waste water discharge from the settlements.

The recharge intensity plays an important role in controlling the chemical composition of spring water and the concentration of both TOC and TN.

According to chemical analysis, sources of pollutant of Jericho spring water might be:

- Runoff of agrochemicals. (Khayat et al, 2008)
- Discharge of waste water from some Israeli settlements.
- Leaching from stone quarries and the municipal and other industrial waste water that discharging from the eastern side of the city of Al Bireh (Khayat et al, 2008)
- Bedouins living at the down stream dumping their waste water into the stream. (Khayat et al, 2008)
- Absence of efficient treatment plants and control of waste water in the West Bank (vacuum tanker W.W drains freely through natural streams to adjacent wadies (ARIJ, 1995).

Most springs in the study area are moderately contaminated by NO_3^- from agriculture and waste water, ranging from about 6 to 40.9 mg/L. The highest concentrations were recorded at Sultan spring (40.9 mg/L) and Dyouk spring (38.7mg/L). These concentrations approached but did not exceed the WHO (MCL) of 50 mg/L.

Water from spring gives variable level of TOC and TN, along the hydrological year, when THM formed.

High levels of TOC and TN in the spring water during the sampling period were closely related to rainfall/runoff events. Also, the TOC levels were highly variable and were found to change from season to season as well as from spring to spring.

Water chlorination is the only treatment measure that takes place before distribution water. During this study, it was reported that the free residual chlorine values is usually measured at levels less than the recommended values required for efficient drinking water disinfection by WHO. The average concentrations for residual free chlorine during the sampling periods (summer and winter), in Jericho were (0.09 and 0.19 mg/L) respectively, while that for Aqbat jabber camp were (0.18 and 0.09 mg/L) respectively. Such low concentrations show that the chlorine dose added to the net work through a process of disinfection is not sufficient effective disinfection process. High temperature conditions in Jericho area may properly promote the accelerated depletion of residual chlorine which mitigates THM formation.

The concentrations of THMs formed in the water network vary depending on the concentration of TOC in the discharged spring water, and chlorine dose plays an important role in this process with increasing the contact time.

The study showed that the THM levels in the tap water of Jericho and Aqbat Jabber camp are at levels below the USEPA limit of 80 $\mu\text{g/L}$, as well as the WHO limit of 100 $\mu\text{g/L}$. In this study, from most samples analyzed, CHBr_3 was found to be the major contributor to the total THMs.

The experimental data of THMFP test performed on Ein Sultan spring and Al Quilt canal water show that:

Higher TOC content of Al Quilt canal water than Sultan spring water resulted in higher chlorine demand and, therefore, higher THM formation potential.

Chloroform was the dominant species observed after the chlorination of each sample in the experiment.

As temperature increased, more THMs formed. Also, THM values measured at high temperature incubation were always higher than the ones measured at low temperature incubation from all of the samples.

A result of chemical analysis for Al Quilt canal, before and after sand filtration, shows a small change in water quality after this process. It is clear that sand filter purifications process is not effective to purify this water from pollutants which are present in Al Quilt canal. TOC and TN are still high in this water.

6.2 Recommendations

Special attention should be drawn to the values of nitrate in the groundwater, especially in Sultan and Dyouk springs should be monitored periodically, as the two springs are considered as the main sources from drinking water in the surrounded area. The presence of relatively high nitrate value in the springs >40 mg/L should be considered as a serious warning about more high values from the surrounding pollutants from the upper wadis in the future; thus, the main drinking water resources in this semi-arid area is still under threaten.

Chlorination process needs more attention for the optimization of chlorine dose and residual concentrations that should be maintained in the distribution system according to the contact time and other chemical and physical properties of drinking water. Also, the chlorine demand for effective and safe disinfection should be estimated according to the source water quality and the organic material content.

The formation potential of THMs of Jericho spring water used for drinking water should be studied for longer times (at least for all months in the year) in order to better understand the seasonal changes and its effects on THM formation.

Water utilities should direct their efforts at reducing the levels of organics during drinking water treatment. Reduction in organic carbon levels result in long term improvements of water quality (lower THMs levels), better stability of disinfectants, no taste and odor customer complaints, and no bacterial and other infectious agent regrowth in the distribution system.

Sand filters efficiency must be measured around the year to provide good quality water for Aqbat Jabber camp.