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AINA IZZATI MUHAMMAD RASHID (ORCID 0000-0002-1068-566X)[1, 2]

ROSIAH ROHANI (ORCID 0000-0002-4391-4043)[1, 3]

OTHMAN JAAFAR (ORCID 0000-0002-0820-6260)[2]

NUR SHAZWANI MUHAMMAD (ORCID 0000-0002-2432-4822)[2]

CHLORINE DECAY AND FORMATION OF THM IN MALAYSIA'S WATER DISTRIBUTION SYSTEM

Chlorine is a popular disinfectant used in Malaysia in the treatment process of drinking water supply because of its effectiveness. The concentration of chlorine deteriorates upon travelling in the system due to its reaction with organic matter to produce carcinogenic substances known as disinfection by-products (DBP) such as trihalomethanes (THM). This study was conducted to investigate chlorine decay and THM formation in a state's drinking water distribution system in Malaysia specifically across a 24.9 km distance. EPANET 2.0 Software program was used to perform hydraulics and water quality analysis using the extended period simulation (EPS) for 24 hours demand pattern. A simulation of the water distribution system was performed to identify the formation of THM and its relationship between chlorine and total organic carbon (TOC). The value of THM was maintained at a lower level at the water treatment plant (WTP) than at the endpoint of the distribution system. At the endpoint, which was at the targeted industrial area, the level of THM was found to increase and the obtained data showed that its formation occurred along the investigated distribution system. THM formation manifested as the natural organic matter (NOM) presence along the pipe continuously reacted with chlorine which was dosed in the distribution system.

[1]Department of Chemical and Process Engineering, Faculty of Engineering and Built Environment, University Kebangsaan Malaysia, UKM, Bangi 43600, Selangor, Malaysia.

[2]Department of Civil Engineering, Faculty of Engineering and Built Environment, University Kebangsaan Malaysia, UKM, Bangi 43600, Selangor, Malaysia.

[3]Research Centre for Sustainable Process Technology, Faculty of Engineering and Built Environment, University Kebangsaan Malaysia, UKM, Bangi 43600, Selangor, Malaysia, corresponding author, email address: rosiah@ukm.edu.my

1. INTRODUCTION

Access to safe drinking water is essential for health and is one of the components of an effective policy for health protection [1]. Water governance in Malaysia is considered successful in the sense that water is served to more than 95% of the population and the water tariffs are considered as one of the cheapest in the world [2]. To supply water of good quality, Malaysia's water authorities and operators are required to follow the regulations of drinking water quality outlined by the Ministry of Health (MOH) of Malaysia and the World Health Organisation (WHO). Water treatment chemicals are commonly used to improve the quality of drinking water but they can also be a source of hazards if they are dosed at a very high concentration [3].

In a water treatment process, the addition of chemicals that have both organic and inorganic nature including heavy metals and hazardous compounds is a concern in the drinking water from the health perspective as they are toxic to humans and may cause cancer [4]. Many harmful chemicals can contaminate water in the distribution system, which some of them can be sourced from the unwanted residues of chemicals used during the water treatment process, reacting materials, scales and deposits, and also chemicals entering from faults and breaks [3]. Water treatment should be optimised to prevent microbial growth, corrosion of pipe materials, and formation of deposits to maintain good water quality in the distribution system [5].

Water entering the distribution system must be consistently safe and ideal [5]. Thus, disinfection is the most crucial step in the treatment process and become the final barrier against bacteriological impurities in supplying safe and ideal drinking water [6, 7]. Chlorination is a popular disinfection method used in water treatment plants (WTP) around Malaysia with liquid chlorine being a common chemical disinfectant used in the process. Therefore, an adequate concentration of disinfectant is essential to be dosed in the water treatment system [5].

Reactions of chlorine with other substances in the water distribution system can cause deterioration of the treated water quality. Its effectiveness as a disinfectant depends on the dose amount required in the treatment plant and chlorine contact time in the distribution system [8]. Since the 1970s, researchers have observed the formation of chlorinated organic material in drinking water systems upon using chlorine as a disinfectant and hence recognised their carcinogenic potential [6, 9] in the formation of disinfection by-products (DBPs) such as trihalomethanes (THM).

Alternatively, operational monitoring can be done to evaluate the effectiveness of each control measure for a pipe distribution system including chlorine residual monitoring [10], which may provide a rapid indication of any problem encounters. The water distribution network is an essential element of the service provision which function is to deliver water to consumers [11]. A good water supply practice can be achieved by managing the water distribution concerning chlorination via maintaining sufficient residual chlorine and low DBPs concentrations in the whole system [7].

DBPs can be produced by the reaction between disinfectants with naturally present organic acids and/or other natural organic matters as well as halogenic ions in the water [5]. However, guidelines have been established at certain disinfectant concentrations which values below a certain level are acceptable [3]. These guideline values are derived for different chemical constituents present in drinking water [5]. The set guidelines for specific chemicals potentially exist in drinking water are very important in the water treatment plant processes, in a way to control the amount of chemical dosing incorporated during the process. However, risks associated with inadequate disinfection are already well assessed and the impacts are far greater than the potential risks due to the long term exposure of DBPs [3]. It is known that most disinfectants will produce DBPs during the disinfection process, which can be due to the natural organic matters (NOM) present, the type of disinfectant used, disinfectant dosage applied, pH, temperature and disinfectant contact time [4].

The formation of DBPs in drinking water is mainly caused by the reaction between organic compounds with chlorine [6, 12, 13]. It is believed that the DPBs presence in water can increase the risk of cancer in long term consumption and this has become a potential concern [14]. Although a wide range of DBPs' derivatives may be formed in water as a side reaction from the chlorination disinfection process, THM can be considered as a good indicator of the chlorination by-products from the reaction process [5]. THM has been identified as the first class and primary halogenated DBPs from chlorination and among the premier compound regulated in the drinking water standards [9, 14, 15].

The formation of THM in drinking water may not only be due to the reaction with organic matters, it may also happen because of extended contact time that could influence its formation. Apart from that, the THM formation also depends on the variable hydraulic properties and water demands for the specific water distribution system, as reported by Chowdry et al. [14]. They also reported that the effect of temperature poses a similar impact on the THM formation especially due to seasonal variability [14]. For instance, the winter season requires different chlorination doses thus a different trend of THM formation might occur compared to that of the summer season.

In Malaysia, as an equatorial country, the temperature is rendered as one of the parameters investigated in the relation to the formation of THM along the water distribution system. Due to that, a specific model is usually used to predict THM in the water system as the information is essentially needed in evaluating the concentration of chlorine and THM as well as in obtaining their relevant reaction rates that could influence the water quality [7]. This important information is required in measuring the formation rate of THM in water and consequently understanding the chlorination and its by-products related mechanism. A predictive DPB or THM formation model that considers the bulk and wall kinetic rates, which these rates may be varied and dependent on the distribution system used and also their environmental conditions [7, 14]. These parameters are very important in understanding the referred model and thus are required to be investigated.

The common reaction between chlorine and natural organic matter (NOM) produces DBPs of a different range of substances namely THMs, haloacetic acids (HAAs), haloacetonitriles (Hans), chloral hydrate, halopropanes, cyanogen halides, chloroporicin and other DBPs. Since THM is the main by-product of chlorination and also a major constituent of DBPs [16], its formation can represent the DBPs as a whole. THM can be formed right after the injection of chlorine in the water treatment plant, can be identified as early as in the clear water tank (CWT) and its detection could resume along with the distribution system. THM is believed could be continuously formed in the water distribution system as long as there is sufficient residue of disinfectant and reactive precursors available in the water [17]. In particular, the longer the reaction period between chlorine and NOM in the water, the greater amount of THM can be formed. Hence, an understanding of the chemical reaction between NOM and chlorine is important in a way to eliminate the THM occurrence [6].

The consumption of water that has been contaminated with THM may cause liver, kidney, or central nervous system disturbances as well as increase the risk of cancer, especially in a long-term period. Moreover, a higher concentration of chlorine and NOM in the distribution system may subsequently increase the production of DBP or THM in the distribution system. Of these DBPs, THMs are produced at the highest concentration when disinfectants and organic materials such as humic and fulvic acids are present [4]. Other factors that could affect the formation of THMs are including pH, temperature, contact time, a specific concentration of disinfectant and its residual concentration and also characterisation of NOM itself [4, 6]. As the level of THM is influenced by temperature, seasonal conditions will be significantly affecting the THM level, especially for seasonal countries. For instance, Sadiq et al. [18] have observed higher DBPs concentrations, particularly in the summer months. Meanwhile, in a country such as Malaysia with temperature variation between 28 and 32 °C annually at a higher degree, the formation level of THM may be varied and this must be observed and monitored for better control of water quality.

NOM is a complex matrix consisting of organic materials that are found in natural water. NOM is derived mainly from living or decayed vegetation and microbial decomposition processes [8]. Total Organic Carbon (TOC) is a well-known parameter indicating the content of carbon in organic form in any sample, which is used to measure different NOM fractions, as well as DBPs precursors [6, 8]. The TOC parameter reveals a detailed chemical composition or reactivity of NOM constituents with chlorine, which is commonly used as a disinfectant. Chloroform is the most common THM and is the main DBP in chlorinated drinking water [5], which detection in the water distribution system can prove the potential formation of THM [18].

Thus, identification of DBPs formation in water, as well as the development of its model, became one of the main aims of this work, as it is important to assess the presence of DBPs and their impact upon its formation. The DPBs reactions and the reaction kinetics in the distribution system have been investigated as it is also essential in this

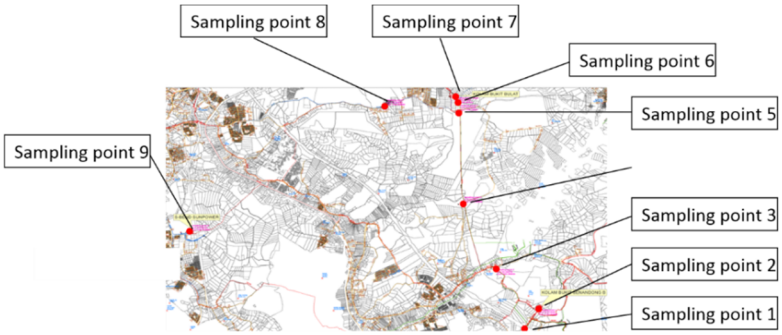
assessment. From this assessment, the developed DBPs model of THM constituent may expose the relationship between each parameter of water quality with THM formation during the water quality monitoring in the WTP operations and water distribution. The modelling of DBPs and THM could assist in decision-making processes in the drinking water industry and emphasise the need for examining the current knowledge concerning the issue. On the other hand, the predictive result from the model on the THM formation concentration in water could also be related to the water quality and operational parameters in the WTP and distribution system.

Therefore, in this study, the bulk and wall decay rates were investigated in a way to assess the rate of THM formation along the specific distribution line as well as identify the effects of chlorination and water demand patterns in the distribution route. Both the laboratory-scale and plant-scale water distribution samplings were used in the model simulation to predict the THM formation and validate the model. This work was conducted by undertaking relevant analyses for the water samples from a specific distribution system identified from local WTP located in a state in Malaysia. The water samples were analysed for their water quality, followed by performing the THM formation prediction modelling and simulation of the process by using EPANET 2.0 software program in comparison to the real measured THM. The obtained results were found to be useful for understanding the operational and water quality management, as well as in the evaluation of water treatment facilities, health risk assessment, and estimating the impacts of the THM standards and regulations.

2. MATERIALS AND METHODS

Field study and sampling. The sampling process was conducted along the water distribution system in a state in Malaysia, which was a dedicated line to distribute water from WTP (starting from the clear water tank (CWT)) to a specific industrial area. To have a clear understanding of the reduced chlorine concentration (based on the chlorine residual parameter) and the THM formation, nine (9) sampling points were identified along the distribution system. The water quality deterioration and THM formation were investigated by conducting the water sampling programme for more than six months at the stated nine sampling points along the 24.9 km route pipeline. The sampling frequencies and locations were selected to provide the greatest confidence that all parts of the systems operated within the target ranges [3]. Coordinates and locations of the sampling points are given in Fig. 1.

Full sampling was performed at least once a month from September 2016 to April 2017 to provide sufficient samples and data for the laboratory testing. More data was also collected to support the finding whereby additional test was included for five days in mid-November 2017, to support the THM formation pattern study in the distribution system. The samples were collected starting from the CWT until the specified industrial area at the same time daily.



Sampling point	Details	North (N)	East (E)	Sampling point	Details	N	E
1	Clear water tank Bertram WTP	2.295700	102.2996	6	Inlet SRO Bkt. Bulat	2.381356	102.2877
2	Inlet SRO Bkt. Senandong B2	2.313939	102.3107	7	Outlet SRO Bkt. Bulat	2.381257	102.2878
2	Outlet SRO Bkt. Senandong B2	2.326751	102.2990	8	S-bend at Taman Sri Manggis	2.378124	102.2687
4	Estate area 1	2.347110	102.2900	9	S-bend at AUO Sunpower Factory	2.338172	102.2148
5	Estate area 2	2.376429	102.2889				

Fig. 1. Locations and coordinates of the sampling points

Sampling and experimental methods and procedures. The sampling method used in this research followed the *Guide to Drinking Water Sample Collection*, 2nd Ed., by Environmental Protection Agency (EPA), 2016. The tests involved in this study were divided into two categories namely in-situ and laboratory tests. The in-situ tests were done based on the *Standard Method for the Examination of Water and Wastewater*, 22nd Ed., by the American Public Health Association, American Water Works Association (AWWA). The laboratory tests were separated according to different tests and methods required. Table 1 displays the methods used for in-situ and laboratory tests.

Table 1

In-situ and laboratory tests

Subject of the test	Method
Residual chlorine	4500-Cl G (DPD colourimetric method)
Total Organic Carbon (TOC)	HACH Direct (10129) test N tube vials d
Trihalomethanes (THM) measured by chloroform	HACH THM Plus (10132) water bath

The in-situ test was conducted for residual chlorine (a very active compound that easily evaporated in the air) by using the HACH spectrometer DR 890 colourimeter equipment. The laboratory tests for both TOC and THM were done by using the HACH

spectrometer DR 6000 series with different methods (Table 1). The testing was performed for duplicate samples and repetitive testing, to ensure the accuracy of results obtained as recommended by the guidelines from the Ministry of Health (MOH) of Malaysia. The samples for each parameter were taken from all the sampling points and kept in cleaned amber glass bottles. The sample collection was performed based on the Guide to Drinking Water Sample Collection by EPA.

Bulk and wall decay coefficients. The bulk and wall decay rates used for the EPANET simulation process (presented in the next subsection) were obtained from the samples test conducted at the laboratory from 21 to 24 h. The value was calculated based on the first-order reaction kinetics

$$C(t) = C_0 \mathrm{e}^{-k_b t} \tag{1}$$

where $C(t)$ and C_0 are actual and initial residual and initial chlorine concentration, respectively, mg/dm^3, k_b is the bulk decay rate constant, 1/h, and t is contact time in h.

The rate of water quality reactions at the pipe wall or near it can be considered as dependent on the concentration in the bulk flow. This rate was determined using an expression [19]

$$\frac{dC}{dt} = \frac{A}{V} K_w C^n \tag{2}$$

where K_w is the wall reaction rate coefficient (m/day), A/V is the surface area per unit volume within a pipe (equal to 4 divided by the pipe diameter).

Networks modelling. The prediction of the THM formation represented by chloroform in the treated water was developed by performing a modelling selected system using EPANET 2.0, free software usually used for determining the hydraulics and water quality.

In this study, the source of raw water was obtained from the river and then pumped to the WTP for undergoing a conventional treatment process. Next, the distribution system modelling was developed involving: i) a CWT connected to a pumping station that was used to provide water to the system, ii) two tanks with a respective estimated volume of 4000 m^3 and 5000 m^3, iii) 29 junctions, and iv) 17 pipes made of mild steel and ductile iron with diameter between 650 mm to 1000 mm. The ages of the pipes in the distribution system were identified between 5 and 30 years and these pipes were maintained accordingly following a proper schedule. The selected system was chosen for investigation in this study as it only has a single route for water delivery to a specific industrial area. The water demand from this dedicated industrial area was captured

around 12 000–14 000 m^3 treated water per day, as identified from the flow meter installed at the industrial premise.

The treated water was pumped from CWT to tank 1 and then it flowed by gravitational force along the distribution system. Water in the reservoir and tanks was assumed fully mixed as it entered the tank from an inlet pipe. The hydraulics properties of the distribution system such as water velocity and retention time were taken into consideration for the modelling and simulation processes. The water demand for the industrial area was taken into consideration upon calculating the flow and velocity at the distribution system as it was a dedicated line to distribute treated water to the industrial area. Meanwhile, the retention time was recognised as another important parameter that could influence the formation of THM in the distribution system. The retention time was obtained using the equation:

$$T(\theta) = \frac{AX}{Q} \tag{3}$$

where $T(\theta)$ is the water retention time, s, A is the pipe surface area, m^2, X is the distance from the outlet, m, and Q is the flow rate, m^3/s.

Numerical simulations were conducted considering various parameters and conditions of the distribution system. The Lagrangian approach was used for the system modelling which was more efficient for simulating chemical transport [19, 20]. Another consideration required for the simulations was extended period simulation (EPS), which was chosen in doing the simulation for 24 hours for each of the studied parameters. From the simulation, the obtained THM and TOC predicted results were finally compared with the actual and experimental results for validation. The most accurate and the best data obtained from the simulation and experimental work were determined, which these data are expected to pose the least error and within the range of data at the best fit line.

3. RESULTS AND DISCUSSION

By-products of chlorination in drinking water were discovered as early as 1974. Ever since, numerous studies to predict DBP formation in drinking water were conducted and presented in simulation models including using EPANET 2.0 software program [14]. It was observed that the organic compounds present in the water distribution system tend to react along the pipe wall or near it and also through the reactions in bulk water [7]. To investigate this, the main parameters relevant to chlorination namely TOC and THM were simulated in this study using the first-order decay kinetics for bulk decay and wall decay order kinetics. First-order decay simulation was chosen as it was repeatedly used by many researchers in predicting the water quality in the water distribution system or network [12, 21–23].

In this work, the water sampling test was performed to investigate the formation of THM in the distribution system specifically in one of the states located in Malaysia. Due to the equatorial and tropical climate of the country, its formation was considered active since it could highly influence the process. The dosage of chlorine was emphasised during the testing and simulation of THM formation in the distribution system, whereby it was also dependent on the raw water quality and volume of water produced from the WTP. As the drinking water quality deteriorates along the distribution system due to the reaction of the components in the water itself and/or between the water component and the pipe wall, its reaction with other chemical substances or organic matters will result in further decay in the water quality before it reaches the endpoint. This reaction happens in the distribution system either as bulk water reaction or bulk decay reaction, which will cause a reduction in the dissolved oxygen (DO) and increment of the bio-films formation after a certain period of contact with pipe surfaces [11]. Further reaction between the bulk fluid kinetics with the NOM and pipe wall can also cause oxidation reaction in the presence of corrosion and/or with the bio-films [24, 25].

The investigated water quality in this study via in-situ tests included pH, temperature, turbidity, and residual chlorine. Each different sample for TOC and THM was tested in the laboratory with a specified method and procedure. Since the temperature is considered a constant factor for water quality deterioration in the distribution system, and the daily temperature usually does not change much (constant) in Malaysia compared to other seasonal countries. Therefore, this temperature parameter in the water distribution system can be neglected if compared to the other available factors. Hence, the understanding of THM formation and its relationship with other water quality parameters such as chlorine (residual) concerning chlorine concentration is very important and this information can be obtained directly from the water sample analysis. Such findings will help in understanding the formation of THM in water by taking into consideration of the hydraulic properties and water distribution system management.

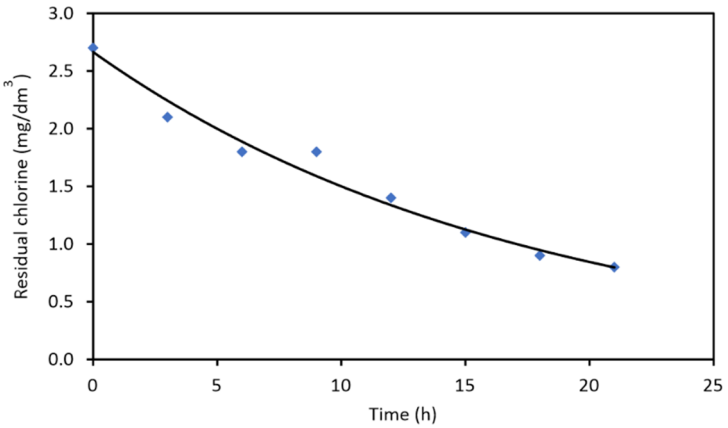


Fig. 2. Time dependence of residual chlorine concentration

To perform the EPANET simulation and model for the TOC and THM, both the bulk and wall decay rates were identified using equations (1) and (2), respectively. From the analysis using equation (1), the bulk decay rates were obtained in the range from –0.24 to –0.59 day^{-1} based on Fig. 2 and equation 1. The wall decay rate, K_w was determined by mass per area per time, or length per time. Based on the trials and error run for the wall decay rate using EPANET 2.0 software program, the results ranging between 0.0 and 0.3 m/day were obtained for K_w. The first order reaction rates were used to perform the simulation since it has a better suitability and adaptability with the system properties, parameter data and obtained results.

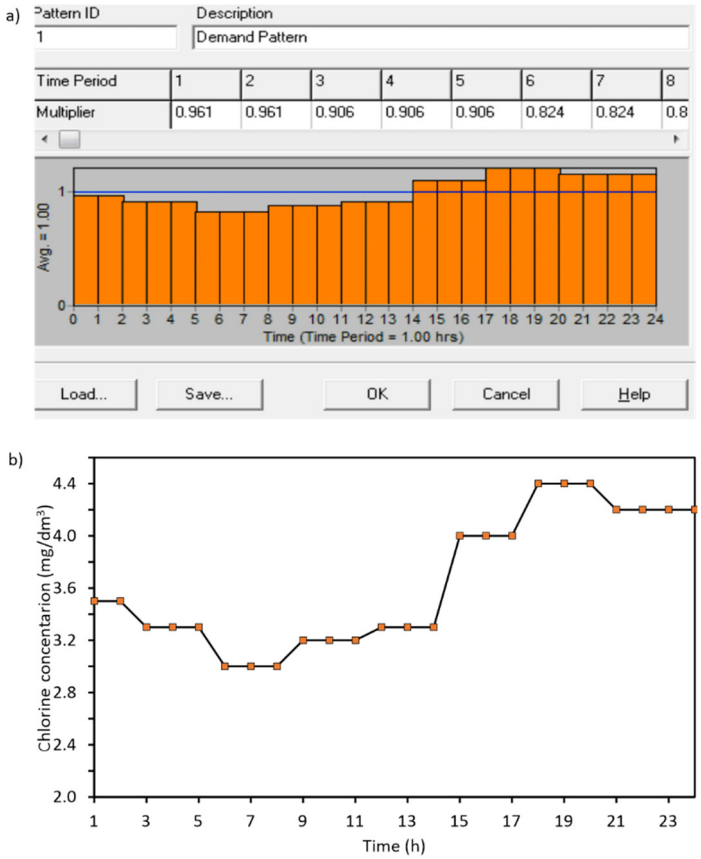


Fig. 3. Water demand pattern plotted by EPANET 2.0 which values were measured by using flowmeter (a), and chlorine average concentration daily demand pattern for October 2016 (b)

After the rates and other relevant parameters were identified, they were utilized to obtain the water and chlorine demand pattern for the investigated industrial area in a way to accumulate the predicted TOC data for the selected system. The simulation of

the TOC and THM values was conducted using the demand pattern for the same sampling period as the samples were collected from the experimental and sampling data. This simulation step was done to get a set of data, similar to the actual operation and reactions that occurred in the distribution system during the investigated period. By using the demand pattern results simulated and plotted in Fig. 3, the simulated results using the predicted data are introduced and presented in Figs. 4–7. It is acceptable that the same demand pattern results were used since both the TOC and THM samples were taken at the same time and thus their relationship with chlorine dose in the distribution system was identified as mentioned beforehand. Furthermore, the water demand pattern for the investigated industrial area can be considered as consistent as shown in Fig. 3. The demand pattern identified in 24 hours have the average value of nearly 1.0 (with the lowest at 0.796 and the highest at 1.375), indicating that the demand is at 1:1 ratio. This finding is obtained because the industrial area is known to own a dedicated distribution system from the WTP with no other additional water demands is presence along the distribution system. This result confirmed the demand pattern is at the fullest for this industrial area. Next, the experimental data of the real measured values were obtained for the formation rates of THM along the distribution system, which were collected from different nine sampling points (Fig. 4). For every water sampling points fixed at the distribution system, the data on the chlorine dosage and water demand pattern obtained were considered as adequate to be used in explaining the formation of THM along the system. The attained relationship between the parameters can be used to monitor and control the formation of THM in the future and further optimization step is to be conducted for the chlorine dosage in the water treatment process, in order to have a sufficient chlorination with the least by-products formation. From this practice, the regulatory agencies can easily use the model to identify the water quality in the water supply system as well as the model can be used in decision-making for selecting the best approaches for the water treatment processes.

3.1. TOTAL ORGANIC CARBON (TOC) MODELLING

Total organic carbon (TOC) detected in the water distribution system may come from any escaped organic matter within the water treatment process. It may be present at/in the reservoir and along the distribution system. This organic matter in the treated water could be the most important challenges to be overcome in protecting a water distribution system or network and also in supplying clean water to consumers [20]. The TOC parameter is found important to be managed and closely monitored in the distribution system in order to control the formation of THM along the distribution system.

In this work, the TOC values were identified upon performing the water sampling from nine (9) sampling points conducted within May 2016 to April 2017 and then testing of the water samples in the laboratory. The analysed TOC data from all the sampling points along the distribution system were tabulated in Table 2. The TOC data values

were presented using the mean, median and range of TOC collected at every sampling point. Generally, the range of TOC concentration was found between 0.10 to 2.70 mg/dm^3. The TOC concentrations showed a slight increase in September 2016 with a slightly higher TOC range compared to other months, which values between 1.90 and 2.80 mg/dm^3. The increase in the TOC concentration in September 2016 was probably due to the high content of organic matter in the raw water which posed a huge impact on the treated water quality, even though the organic matter was expected to be removed during the treatment process. This escaped organic matter can still slip through the WTP in a small quantity and enter the distribution system, thus causing an increase in the TOC value.

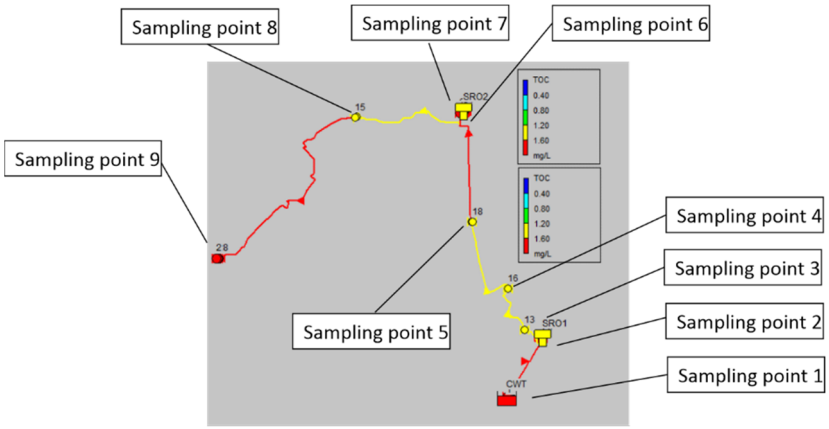


Fig. 4. Results of simulation using EPANET 2.0 to predict the TOC concentrations from the clear water tank (CWT) to the dedicated distribution system (location sampling points cf. Fig. 1)

Since the standard deviations for all data from different sampling points were found below 1.0, this indicates that the results of TOC concentration were predictably within the range of the obtained data. This standard deviation value was strongly affected by the TOC concentration observed in September. If the data in September is excluded from the calculation, the TOC value calculated from the remaining months could be within the range of 0.1 to 1.8 mg/dm^3. Anyway, with or without it, the overall TOC concentration in the system was still considered as under control as the mean and median TOC values were found below the maximum value allowed in the water quality standard, which regulated at 2.0 mg/dm^3.

The risk of having NOM present in the system not only allows it to react with chlorine to form THM but also helps microorganisms to grow in the pipe wall. The maximum level of THM allowed in the system is 1.0 mg/dm^3 as set by the MOH of Malaysia, in a way to prevent high THM formation and to control the growth of bacteria and/or microorganisms in the system. Therefore, proper management and monitoring of TOC or NOM values at the distribution system may not only sustain the water quality in terms

of the chemical parameters but also could strengthen the physical and biological parameters in the distributed drinking water.

Table 2

Values of TOC, mg/dm^3, obtained experimentally from each sampling point during the period from May 2016 to April 2017

Sampling month	Number of the sampling point								
	1	2	3	4	5	6	7	8	9
May 2016	1.00	1.00	1.10	1.00	0.90	0.90	0.90	1.00	0.70
July 2016	1.50	1.50	1.50	1.40	1.30	1.80	1.30	1.40	1.30
August 2016	0.30	0.40	0.40	0.20	0.20	0.50	0.20	0.20	0.20
September 2016	2.80	2.70	2.30	2.20	2.10	2.20	2.30	2.30	1.90
October 2016	1.60	1.70	1.70	1.40	1.40	1.30	1.20	1.40	1.20
November 2016	1.80	1.80	1.80	1.50	1.40	1.50	1.30	1.60	1.40
March 2017	0.20	0.30	0.30	0.20	0.20	0.10	0.10	0.20	0.10
April 2017	1.20	1.20	1.20	1.00	1.10	1.00	0.90	1.10	1.00
Mean	1.30	1.33	1.29	1.11	1.08	1.16	1.03	1.15	0.98
Median	1.35	1.35	1.35	1.20	1.20	1.15	1.05	1.25	1.10
Range	0.20–2.80	0.30–2.70	0.3–2.30	0.20–2.20	0.20–2.10	0.10–2.20	0.102.30	0.20–2.30	0.10–1.90

The formation of THM is advised to be frequently monitored as the high environmental temperature condition in Malaysia could encourage the THM to form faster in the water distribution system. It is also directly related to the amount of organic matter in the water source and finally entering the distribution system, whereby chlorine from the disinfection process could react with it, resulting in the THM formation and occurrence. The formation of THM could continuously occur as chlorine is consistently injected into the system due to the ever-high demand for treated water by the consumers. This reaction continues to occur if the organic matter and chlorine amount are sufficiently present. Therefore, a proper control towards chlorine dosing or monitoring of its concentration may not be sufficient and it should be equipped with strict control of the organic matter level in the water treatment process.

To validate this experimental finding, the predicted TOC value is deemed necessary to be obtained to help accurately monitor and manage the water quality in the distribution system, especially to observe and control the formation of THM. This simulated TOC value was obtained by using EPANET 2.0 software that was run under real input conditions and followed the same hydraulics properties (such as pipe roughness, pump curves, etc.) from the distribution system and the results are presented in Fig. 4. This figure shows the ranges of predicted TOC concentration along the system determined between May 2016 to April 2017 at 9 different sampling points as shown in Table 2. The TOC concentration simulation was performed using the same hydraulic properties identified from the distribution system and the bulk and wall decay rates obtained from

equations (1) and (2), to assess and compare the predicted and actual TOC data in the dedicated distribution system.

Based on Fig. 4, TOC value monitoring for the treated water prior to primary disinfection using chlorine has to be limited at 2.0 mg/dm^3 and below [10, 17] as the TOC is the key indicator for the formation of THM after the chlorination process. The technical report [17] has indicated that THM formation could occur at the level of TOC below or greater than 4.0 mg/dm^3. Meanwhile, the presence of NOM at some point may affect the TOC value and could lead to the reduction in the chlorine concentration demand [26] due to the THM formation. In addition, the oxidation of the soluble NOM that is present in the water could also favour the formation of different organohalogenic compounds [27].

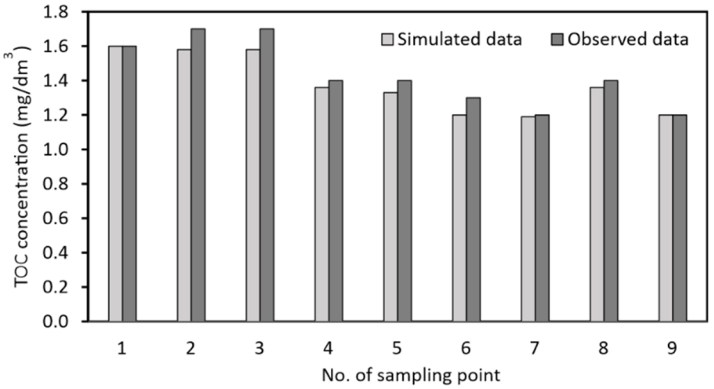


Fig. 5. Predicted and observed TOC concentrations at each sampling point (measured in October 2016); TOC maximum value allowed in drinking water is 2.0 mg/dm^3

Next, the predicted data (from simulation) and the measured data (from water sampling and lab testing) of the TOC were compared for each sampling point in the sampling month of October 2016 (chosen as the values were in the middle of the whole sampling exercise). From the result shown in Fig. 5, it was found that the difference between the highest and the lowest TOC concentration for all nine (9) sampling points in both the simulated and observed data was obtained at 0.5 mg/dm^3 only. All TOC concentration results for both the simulated and observed data were found below 2.0 mg/dm^3, which have fully complied with the TOC requirement by the WHO and EPA.

The accuracy of the modelling and simulation for both the predicted and the observed data was supported by the value of Root Mean Square Error (RMSE) of 0.307 for all the obtained data. The RMSE value between 0.2 and 0.5 indicated that the model can relatively predict the data accurately. The use of the RMSE value has been reported could measure the actual predictive capability of the developed model upon using a set of data [6].

3.2. TRIHALOMETHANES (THM) MODELLING

The concentration of chloroform as the main substance in THM should be less than 200 μg/dm^3 as regulated in the guidelines from the MOH of Malaysia. This limit is lower than the maximum value set by the World Health Organization (WHO) of 300 μg/dm^3. THM build up in the water distribution system can result in a higher THM concentration due to its accumulation in the system and the value could exceed the allowable limit as it may continuously form upon simultaneous reaction between the dosed chlorine and the organic matter presence. Organic matter such as lignin rooted from the decaying vegetative matter owes reactive properties to react with oxidants of chlorine in disinfected water that could lead to high THM formation [17]. THM represented by chloroform values identified from the water samples collected from 9 sampling points between September 2016 to April 2017 are shown in Table 3. The values of all THM data is also presented in the form of mean, median and range of THM for the 9 sampling points. From the result, it was found that the observed data for THM were quite high with some values were recorded above the maximum allowable drinking water quality standard for chloroform.

Table 3

Chloroform concentrations as a main substance of THM in μg/dm^3 measured at each sampling point at the sampling period

Sampling month	Number of the sampling point								
	1	2	3	4	5	6	7	8	9
September 2016	127	130	129	118	128	116	90	95	95
October 2016	210	169	208	225	242	249	165	138	151
November 2016	383	474	430	474	355	176	282	259	241
March 2017	35	35	39	45	33	40	20	18	25
April 2017	194	193	231	210	170	198	129	125	139
Mean	190	200	207	214	186	156	137	127	130
Median	194	169	208	210	170	176	129	125	139
Range	35–383	35–474	39–430	45–474	33–355	40–249	20–282	18–259	25–241

The data for THM referred from chloroform reading recorded in November 2016 were found at the highest than the other observed months with values ranging between 176 and 474 μg/dm^3. The value obtained was more than doubled than the maximum level allowed by MOH of Malaysia and WHO for the chloroform standards of 200 and 300 μg/dm^3, respectively. If the data in November 2016 is eliminated due to its abnormal overshoot values than the rest of the months, the range of all THM data recorded from the system only felt between 25 μg/dm^3 and 249 μg/dm^3. This new range of THM only slightly exceeded Malaysia's standard by 49 μg/ dm^3. However, this obtained range value of THM for the investigated distribution system was considered relatively acceptable and below the WHO standard. These observed values must be monitored and

minimised in the future to avoid any more THM violations and maintain the water quality following the referred guidelines. Precautionary actions also need to be taken to eliminate the THM formation in drinking water by optimizing the chlorine dosing management and minimizing the organic matter content from the treatment process.

Next, the simulation of THM value was conducted from all the sampling points to obtain the predicted THM values for different water and chlorine demand. The dosing of chlorine at a certain concentration level from the WTP (Fig. 3) on the chlorine demand pattern might be the reason for the increase in THM formation. This chlorine dosing can be properly controlled to limit the formation of THM in water, by adding a consistent ratio of chlorine dosing amount with a constant volume of treated water in the system. In this work, the first-order coefficients and rates were used in simulating the THM, to validate the model with the real observed value from the system. The results of THM simulation are shown in Fig. 6, where the network showed different ranges of THM detected along the distribution system. The variation in the predicted values indicated the occurrence of the potential relevant chemical reactions in the system. This value obtained from the developed model may greatly assist the water operators to manage and predict the formation of THM as early as at the plant level. The THM prediction is affirmed to highly dependent on the chlorine concentration and TOC level present in the system. This data could be synchronized with the demand pattern of water that was found to depend on the water consumption in the industrial area and this value changes at every hour (Fig. 3).

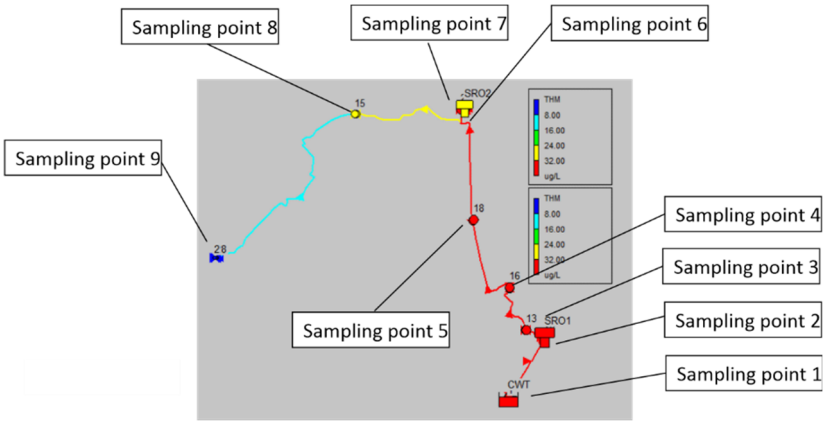


Fig. 6. Results of simulation using EPANET 2.0 to predict THM concentrations from the clear water tank (CWT) to the distribution system (locations sampling points cf. Fig. 1)

The hourly change demand pattern will surely affect the predicted TOC and THM values. Thus, the same demand pattern could also be used to explain the amount of NOM present in the system that could result in the formation of THM. From Figure 6,

the formation of THM is considered as high in the reservoir due to the long contact time between the chlorine and the NOM. The occurrence of organic matter in a higher quantity than usual in the system especially in the reservoir will cause it to react with chlorine to form THM; the longer the contact time, the more the formation of THM. Based on the result presented in Fig. 6, the THM formation in the selected system was considered as low. The formation of THM depended on the concentration of TOC and chlorine in the distribution system. Therefore, it will be continuously occurring with the continuous injection of chlorine into the system. As chlorine reacts with NOM to produce THM, a controlled dosage of chlorine into the system is crucial and must be monitored closely.

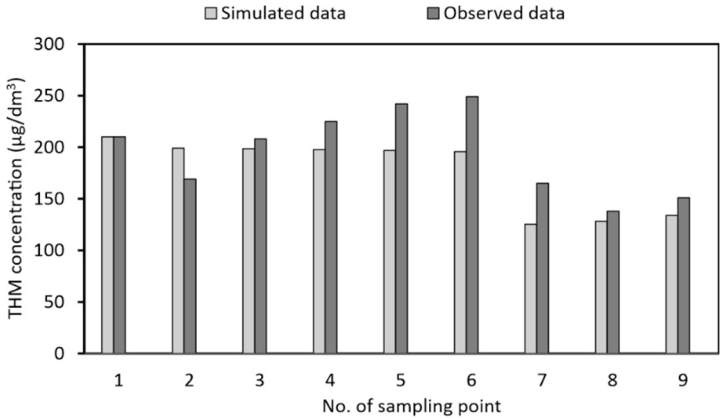


Fig. 7. Predicted and the observed THM concentrations for each sampling point
THM maximum value allowed in drinking water is 2 µg/dm^3

Improved system management and close monitoring of water quality are importantly needed to minimise the impact of THM formation in drinking water supplies [10]. Figure 7 shows the data of the predicted and the observed data of THM for the selected date of October 2016 for each sampling point. From the analysed data, the difference between THM value out of the total number of data obtained at different sampling point was only observed at 25.69 µg/dm^3. Meanwhile, the small error value for the difference in the reading yielded by EPANET 2.0 (Fig. 7) indicated the accuracy of the model upon mapping with the real system hydraulic properties, as well as with other necessary settings required for the simulation.

On the other hand, the formation of THM could depend on the temperature, which is consistently high in summer (for seasonal countries) at 25 °C and higher [12, 13, 27]. Thus for the formation of THM in Malaysia, with an average temperature of around 30 °C and above, the THM concentration could be exceptionally high in volume and the formation rate could be potentially higher if no proper control is implemented. The high value of pH can also increase the THM level in the treated water [6], thus it is essential to manage and control water quality from the water treatment plant itself before it is

being distributed. However, the formation of THM at the endpoint was found to remain manageable in this study whereby all the data obtained were mostly below the standard limit as set by the WHO and MOH of Malaysia.

Although the observed data for sampling points 1, and 3–6 were slightly higher than the maximum value allowed in the drinking water quality standards, the predicted data for the sampling points from the simulation were below the maximum value allowed except for sampling point 1 (Fig. 7). At sampling point 1, the predicted and observed value were almost equal, which might indicate the relationship between the NOM or TOC values with the chlorine dosing at the CWT from the WTP. Chlorine could actively react with the organic matter in the water to form THM. Even though the TOC level was relatively below the maximum value allowed (Table 2), the formation of THM could still occur if the amount of organic matter and chlorine were sufficient to initiate the reaction.

3.3. RELATIONSHIP BETWEEN RESIDUAL CHLORINE, TOC, AND THM

The minimum residual chlorine to be present in the water that reaches the consumers is at 0.2 mg/dm^3 and the maximum at 5.0 mg/dm^3 as set by the Ministry of Health (MOH) of Malaysia. To achieve the minimum level of residual chlorine at the end of the system (0.2 mg/dm^3 or higher), the water operators require a higher concentration of chlorine to be injected into the distribution system. However, as the water flows through the distribution system, the concentration of chlorine decays. THM formation monitoring should be considered along the distribution system at various locations regarding the population size or treated water usage volume. In this studied case, the sampling locations should be set best at between 8 and 10 points, as suggested in the technical report [17]. In Fig. 8, the levels of residual chlorine, TOC and THM are shown. The relationships between them can be observed. The level of the generated THM can be varied depending on the reactions of TOC with chlorine in the distribution system. Such formation continuously happened as long as residual chlorine and the precursors were available at the site [13], namely based on the TOC or organic matter availability in the system. Sampling points 1 to 5 showed the low level of chlorine and TOC indicating that the reaction and formation of THM have already taken place.

Sampling points 6 to 9 showed the formation of THM still ongoing with chlorine as the controlled parameter. The reaction may also depend on the water retention time, which allows the formation of THM to increase over time. The longer period of contact time between chlorine and the organic matter or TOC, the higher the potential of THM formation. In this study, the flow in the distribution was found consistent as the system was based on the main pipe or system to deliver a large volume of water required by the user or industrial area (see demand pattern in Fig. 3).

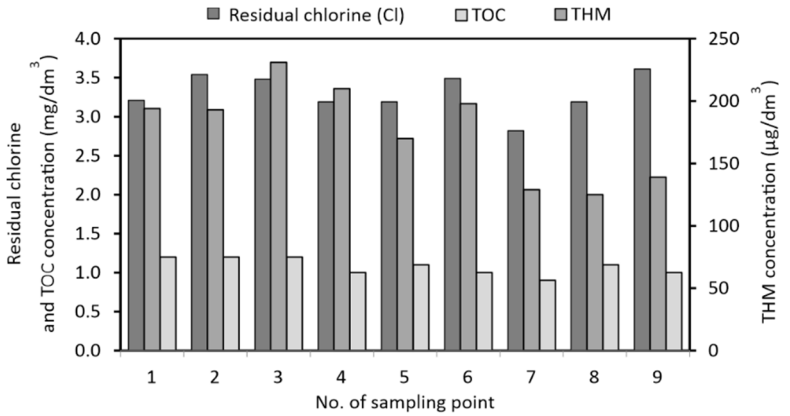


Fig. 8. Observed residual chlorine concentration, TOC and THM for each sampling point

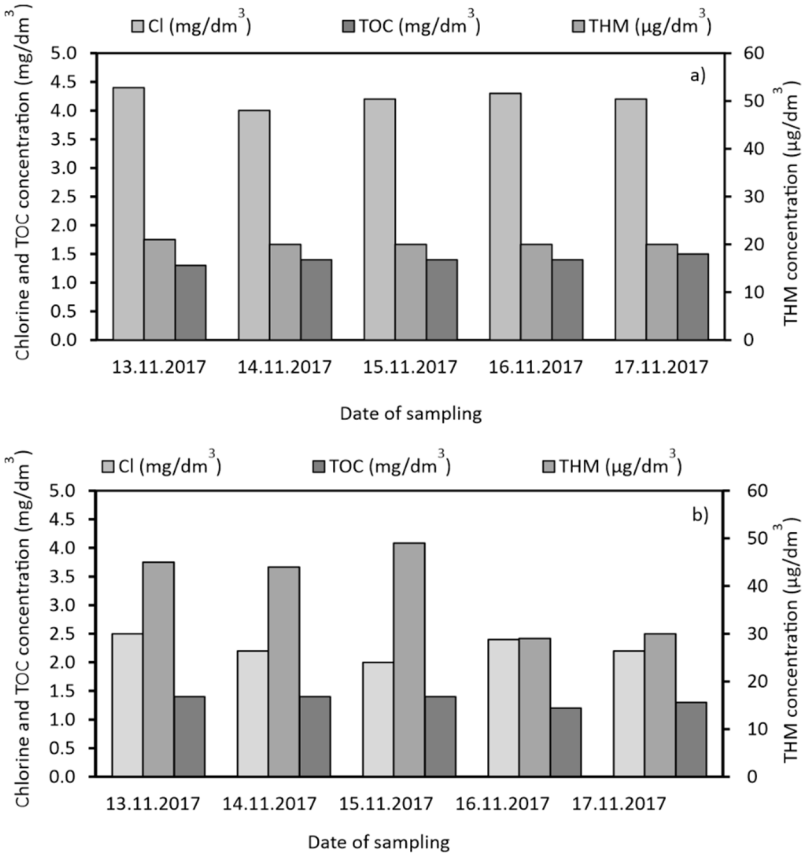


Fig. 9. Observed residual chlorine concentration, TOC and THM for the continuous sampling at start- and endpoints of a selected distribution system: a) sampling point 1, clear water tank, b) sampling point 9, industrial area

Figure 8 also shows two sampling points, i.e., points 3 and 4, that had THM levels higher than the level of chlorine. This indicated that the formation of THM occurred in the system as chlorine was continuously injected into the system. Even though chlorine concentration was high, the level of THM formation can be considered still low along the distribution system. The formation of THM was at the minimum level despite the level of TOC slightly decreased, confirming that the reactions remained to happen.

The possibility of THM to be produced is always there even though the chlorination is controlled at the WTP level. This is proven from the results presented in Fig. 9a that shows the level of THM recorded from all the sampling points which are under control at the WTP level. At the same dates of sampling in the month of October 2016, the formation of THM at the industrial area was found higher than at the WTP, with the reaction between chlorine and organic matter along distribution system (Fig. 9b).

During all the investigated period, the levels of THM at the CWT were below 25 $\mu g/dm^3$ while the concentrations of chlorine were within the range allowed, i.e., below 5.0 mg/dm^3. At the same period, the level of THM was shown higher for the dedicated industrial area. Therefore, the reactions between chlorine and organic matter are believed to occur in the system. The level of THM formation observed has increased by 20–100% along the distribution system with a range of 5–25 $\mu g/dm^3$. The reaction between chlorine and organic matter as TOC was actively present along the system but remained controllable. This was shown by the level of THM, which was measured to be below the maximum level allowed in the drinking water quality standard as per the MOH of Malaysia and WHO.

Overall, it is important to control and remove DBPs such as THM as these substances carry mutagenic and cancerogenic potential [28], which are the first initiated from the WTP. The removal of THM can be done by eliminating its forming potential reaction and indicator which is the organic matter and TOC during the water treatment process. The level of THM in the form of chloroform along the studied distribution system was below the guidelines' maximum value of 200 $\mu g/dm^3$ THM formation depended on factors such as pH, temperature, contact time, disinfectant concentration and characteristics of organic matter in the distribution system [6, 12, 13, 29]. To have better control and management towards its formation in the context of Malaysia, other parameters that can affect the disinfection using chlorine, temperature (despite it being constantly high all year) and hydraulic properties of the distribution system, which must be taken into consideration as well.

4. CONCLUSION

The overview of the specified water distribution system assessed by using EPANET 2.0 has revealed that the use of such software is found essential for water operators to deliver safe drinking water to consumers. The process also requires the use of an adequate

and properly managed system, with proper monitoring and effective planning and management. This study found that the investigated distribution system was well maintained and optimally functioning to deliver good water quality to consumers. Proper dosing of chlorine and a controlled presence of organic matter in the distribution system with consideration of hydraulic properties will lead to good management towards controlling THM formation. Although the level of THM at the WTP was low, the value usually could increase at the endpoint, and this indicated that the reaction between NOM and chlorine actively happened along the distribution system. The formation of THM is higher in tropical countries such as Malaysia due to its constantly high temperature all year. Therefore, an accurate model and simulation of the system that has been developed may give an advantage to the water operators in ensuring proper THM formation monitoring and chlorine dosage management. It is also important for the operators to apply the best management and monitoring practices in sustaining the drinking water quality along the distribution system to ensure human health is protected against harm, and most importantly in supplying good quality treated water to consumers.

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