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ORIGINAL PAPER

Speciation and geospatial analysis of disinfection byproducts in urban drinking water

A. Siddique · S. Saied · E. Zahir · Z. A. Nayyer · N. A. Zaigham

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Abstract Trihalomethanes (THMs) are the most abundant disinfection byproducts (DBPs) of the chlorination disinfection. THMs speciation and their geospatial distribution were examined in 58 locations throughout the water distribution network of Karachi city. THMs ($CHCl_3$, $CHCl_2Br$, $CHClBr_2$ and $CHBr_3$) and physico-chemical parameters (pH, TDS, DO, Residual chlorine, temperature and TOC) were determined. $CHCl_3$ was the major THM found in all water samples of 58 locations, which accounted for 91.69 % of the total THMS followed by $CHCl_2Br$ (5.69 %), $CHClBr_2$ (1.78 %) and $CHBr_3$ (0.85 %). Total THMs level exceed the maximum contamination level of WHO and USEPA at some locations. Varying nature of correlation from high to low was found within THMs and among the physico-chemical parameters. GIS linked geospatial analysis revealed the association of THMs level with demographical and geological based variations from east to west of Karachi city. Continuous monitoring program and legislation for the contaminant levels were suggested to avoid adverse public health impact of THMs in drinking water supplies.

Keywords Chlorination · Chloroform · Geographical information systems (GIS) · Natural organic matters (NOMs) · Trihalomethanes (THMs)

Introduction

Chlorine is the most commonly used disinfection agent in drinking water systems throughout the world due to its economical and disinfection efficiency among the other disinfectants. A growing concern has emerged among the public health communities in the recent past about the health threat due to disinfection residues and their byproducts. Use of disinfection byproducts (DBPs) reduces the risk of pathogenic contamination in the drinking water treatment process but may pose a serious threat due to the adverse health effects of the byproducts formed during the process. During chlorination a complex mixture of chlorine byproducts is formed and more than 600 different types of DBPs have been identified (Becher 1999; Richardson et al. 2007). Among chlorination DBPs major group of chemicals include trihalomethanes (THMs), haloacetic acids (HAAs), haloacetonitriles (HANs), haloketones (HAKs), chloral hydrate (CH) and chloropicrin (CP) (Golfinopoulos et al. 2003; Golfinopoulos and Nikolaoub 2004; Kampioti and Stephanou 1999; Nikolaou et al. 1999). Free chlorine reacts with the natural organic matters (NOMs) mainly humic acid (HA) and fulvic acid (FA) in the drinking water streams to generate DBPs.

THMs has been one of the most concerned DBPs among the scientific community and the regulating agencies because they have been recognized as potentially

A. Siddique (✉) · Z. A. Nayyer · N. A. Zaigham
Unit for Ain Zubaida and Groundwater Research,
King Abdulaziz University, Jeddah, Saudi Arabia
e-mail: chm97@hotmail.com

A. Siddique · S. Saied · E. Zahir
Department of Chemistry, University of Karachi,
Karachi 75270, Pakistan

A. Siddique
ars Power Design Ltd, 4891 Dundas Street W,
Toronto, ON M9A1B2, Canada

Z. A. Nayyer
Department of Applied Physics,
University of Karachi, Karachi 75270, Pakistan

hazardous and are the most prevalent on a weight basis byproducts of chlorination (Bull et al. 1995). THMs include chloroform ($CHCl_3$), dichlorobromomethane ($CHCl_2Br$), chlorobromomethane ($CHClBr_2$) and bromoform ($CHBr_3$). The speciation of the DBPs in the natural waters influenced by several factors but the presence or absence of bromide ion greatly affect the ratio of brominated species in the mixture of DBPs and a shift towards brominated species is expected in the presence of bromide ion, which might result in a more adverse influence on the public health (Liua et al. 2011; Singer 1994). The occurrence of THMs in treated chlorinated water significantly affected because of the quality of the water i.e. presence of NOMs, HA and FA and operational parameters i.e. chlorine dose, temperature, pH and contact time (Christman et al. 1990; Abdullaha et al. 2003; Li et al. 2011). Since the first major studies on presence of THMs in chlorinated water by Beller et al. (1974) and Rook (1974) the scientific community and regulating authorities closely monitor the source and community supply of the treated water for THMs as a safeguard to the public health. United State EPA (USEPA) defined a limit of 80 ppb for TTHMs, European Union (EU) standard of TTHMs of 100 ppb, World Health Organization (WHO) defined guideline values of 100 ppb for Bromoform, 100 ppb for Dibromochloromethane, 60 ppb for bromodichloromethane, 200 ppb for chloroform, some of the individual countries also regulated THMs in the drinking water supplies since it is a matter of great concern with public health stand point of view (USEPA 1998a; EU 1998; NHMRC and NRMMC 2004; Health Canada 2010; WHO 2011).

Karachi being the largest industrial hub and populous city of Pakistan with over 18 million population is served by 640 million gallons per day (MGD) of water from Indus and Hub rivers. Disinfection of about 440 MGD water is carried out by means of chlorination in seven water filter plants (WFP) at five locations. Karachi Water Board according to the documents maintains 2.0 ppm of free chlorine at reservoir outlet as compared to WHO value of 0.25–0.50 ppm of free chlorine in the distribution system (KWSB 2011). The major supply of the water (about 75 %) to Karachi is through Indus River in addition to Hub River (about 15–20 %) and groundwater resources in the city. The distribution network consists of open canals, conduits, siphons, multi-stage pumping and filtration.

Much of the concern towards THMs in water is directed because of the categorization of chloroform as known carcinogen and chloroform is the component of THM which is present at highest levels. In addition to chloroform, bromodichloromethane has also been classified as potential carcinogen (Pieterse 1988; Krasner et al. 1994). Therefore, the speciation of the trihalomethane mixture due to its health effects has a great importance to be assessed and evaluated in the drinking water supplies. Geographical information system (GIS) has been widely accepted as an efficient tool for the evaluation and visualization for the geospatial distribution of contaminants in air, water and soil, their management and mitigation (Tabesh et al. 2010; Zerrouqi et al. 2008; Karamouz et al. 2006; Jerrett et al. 2001; Briggs et al. 1997). GIS can play a vital role for analysis and in formulating the quick mitigation plans for high-risk environments as envisaged in the present study. This study has been carried out during 2007–2008 in Karachi metropolitan city (Pakistan) to delineate the speciation and spatial behaviour of the THMs mixture distribution in the drinking water supply of Karachi city (Pakistan).

Materials and methods

Drinking water samples were collected from different parts of the city (Fig. 1) represents the distribution network of city water supply. Source water samples were collected from Kanjher Lake, Hub dam, and from water reservoir in each of the WFP in the city. Samples were collected in headspace–free borosilicate amber glass bottles with Teflon joint screw cap. Each sampling vial was carefully filled just to overflow, without trapping air bubbles inside. Before sampling, sodium thiosulfate (10 %) was added to each container (1.7 mL) to quench further formation of THMs (APHA 1992; Rodriguez and Sérodesb 2001). Samples were collected in duplicates at each sampling point. The samples were transferred to the laboratory for further analyses under dark and refrigeration conditions. Separate samples of the same sampling points were also collected for the physico-chemical analyses [temperature, pH, conductivity, total dissolve solids (TDS), dissolved oxygen (D.O), residual chlorine, total organic carbon (TOC)]. Temperature, pH, conductivity, TDS, DO were analysed in the field with the help of handylab multi 12 m (Schott instrument, Germany), and Free residual chlorine was also measured in the field using the DPD titrimetric method with a field DR-700 colorimeter (Hach USA). Once collected, the samples were stored in the dark at 4 °C and carried to the laboratory for analytical procedures.

In the laboratory, TOC was analyzed in sulfuric acid preserved water samples by Shimadzu TOC analyzer 5000 (Shimadzu Corp. Japan). For the analysis of THMs, a modification of EPA Method 551.1 was applied for the assessment of THMs. The method included liquid–liquid extraction with methyl tert-butyl ether (MTBE) (USEPA 1998a, b; Nikolaou et al. 2002; Golfinopoulos et al. 2003). 6 g of anhydrous sodium sulfate as drying agent and 2 ml of MTBE were added to 35 mL of chlorination byproducts

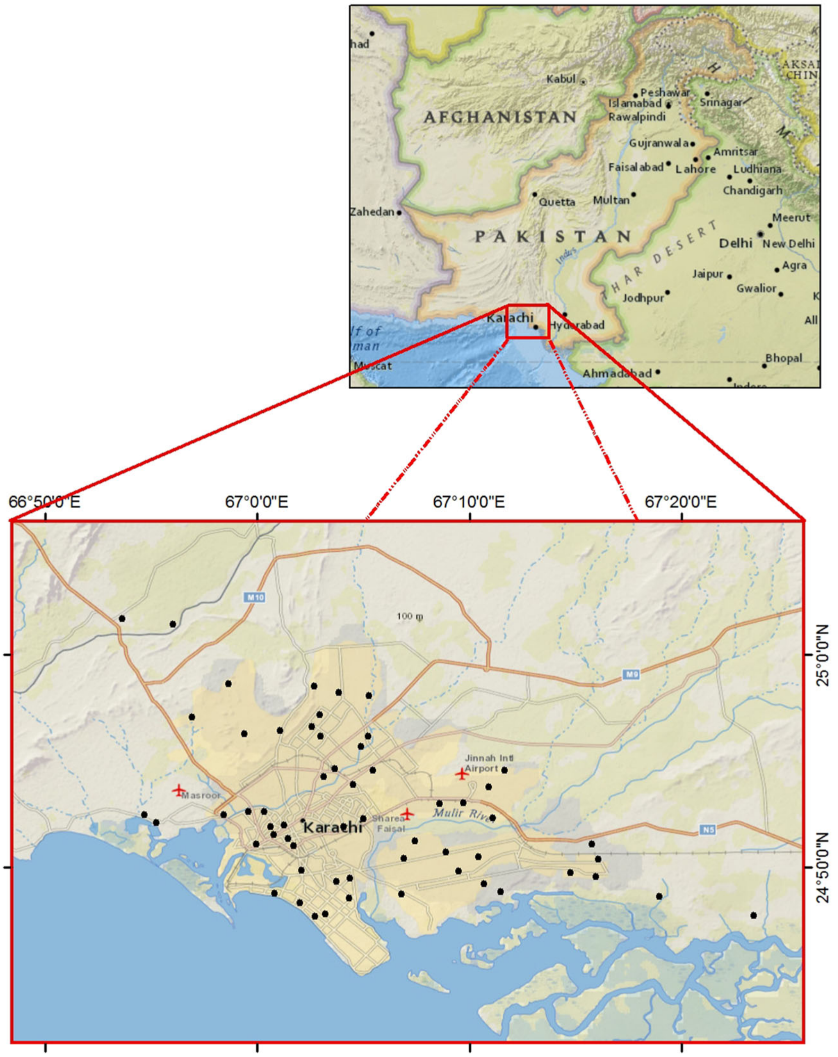

Fig. 1 Index map showing sampling locations

Table 1 Concentration of THMs and physico-chemical parameters for the distribution network of Karachi

	$CHCl_3$ (μg/L)	$CHCl_2Br$ (μg/L)	$CHClBr_2$ (μg/L)	$CHBr_3$ (μg/L)	Total THM (μg/L)	Temp °C	pH	TDS (mg/L)	D.O (mg/L)	Residual chlorine (mg/L)	TOC (mg/L)
Min	32.53	2.02	0.63	0.30	36.04	27.4	7.2	100	4.50	0.05	0.45
Max	167.32	6.22	3.46	0.42	176.24	32.3	8.1	386	9.00	0.42	11.69
Mean	68.80	3.24	1.75	0.36	73.51	31.4	7.4	259	5.92	0.13	3.61
Median	55.77	2.89	1.56	0.38	60.63	30.8	7.3	255	5.75	0.11	3.36
SD	34.28	1.18	0.72	0.05	35.38	2.2	0.24	74	0.75	0.07	2.45

solution in a 40 mL glass vial capped with PTFE-faced silica septum. The vial was sealed and shaken by hand for 1 min and left undisturbed for 2 min for phase separation. The upper 1 μg organic layer was injected into gas chromatograph (GC). Analysis was carried out on DB 1 (30 m, 0.32 mm i.d., 0.25 μm film thickness) chromatographic column and an HP 5890 GC was used. Analysis conditions: injection on column volume 0.5 μL; oven temperature 50 °C for 5 min, 50–150 °C at 6 °C/min; carrier gas Helium 1.3 mL/min, make-up gas N2 60 mL/min; detector temperature 300 °C. For the confirmation of the results DB-1301 (60 m, 0.25 mm i.d., 0.25 μm film thickness) capillary column was used under the same GC conditions that were used for DB 1 capillary column. The method was evaluated at five different concentration levels covering the range from 0.5 to 10 μg/L. Field blanks were used to determine any background contamination. Method blanks and spiked blanks were analyzed and final results were

Table 2 Characteristics of source water quality of Karachi from Keenjhar Lake and Hub dam

	$CHCl_3$ (µg/L)	$CHCl_2Br$ (µg/L)	$CHClBr_2$ (µg/L)	$CHBr_3$ (µg/L)	Total THM (µg/L)	Temp ^0C	pH	TDS (mg/L)	D.O (mg/L)	Residual chlorine (mg/L)	TOC (mg/L)
Min	N/A	N/A	N/A	N/A	N/A	29.4	7.5	227.00	6.70	N/A	2.29
Max	N/A	N/A	N/A	N/A	N/A	37.4	8.1	415.00	8.10	N/A	8.34
Mean	N/A	N/A	N/A	N/A	N/A	31.1	7.7	299.25	7.24	N/A	6.00
Median	N/A	N/A	N/A	N/A	N/A	29.9	7.7	268.00	7.00	N/A	6.32
SD	N/A	N/A	N/A	N/A	N/A	3.2	0.18	73.63	0.56	N/A	2.24

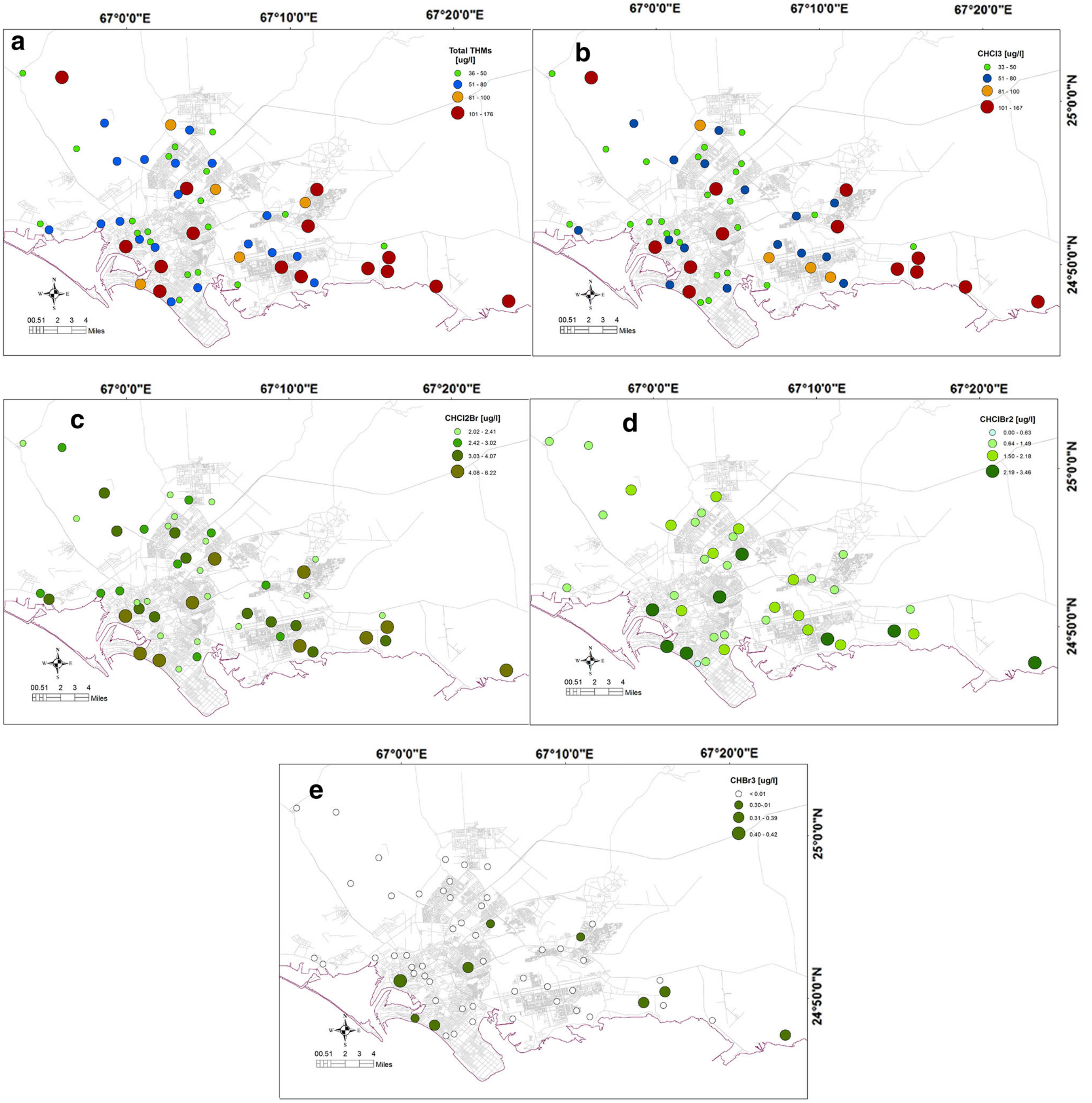

Fig. 2 Distribution of THMs in the water samples of Karachi city distribution network: **a** Total THMs, **b** $CHCl_3$, **c** $CHCl_2Br$, **d** $CHClBr_{2,}$ **e** $CHBr_3$

shown after subtracting any contribution of contamination from the laboratory. The recoveries obtained for $CHCl_3$, $CHCl_2Br$, $CHClBr_2$ and $CHBr_3$ were 78.1–113.2 % (RSD 3.9 %), 86.6–109.2 % (RSD 3.1 %), 80.3–121 % (RSD 4.9 %), 89.2–113.7 % (RSD 3.4 %). The detection limits ranged between 0.01 and 1.0 µg/L.

Results and discussion

Tables 1 and 2 list the minimum, maximum, mean, median and standard deviation values of THMs, temperature, pH, TDS, dissolve oxygen (DO), residual chlorine and total organic contents (TOC) in the water samples from the distribution network and raw source waters (Keenjhar Lake and Hub Dam reservoir) of Karachi. The statistical mean and median values were calculated to demonstrate the central tendency of each parameter in the observed samples. The temperature varies from 27.4 to 32.3 °C with a mean value of 31.4 °C for the distribution network and 29.4–37.4 °C for the source water samples. pH is varied between 7.5 and 8.1 in source water and no significant difference was observed with the pH of distribution network water samples, i.e. 7.2–8.1. Total dissolve solids (TDS) and DO in the source water were higher compared to the minimum, maximum and mean concentrations of the distribution network samples. The concentration of residual chlorine ranged from 0.05 to 0.42 mg/L in the water samples from different parts of Karachi city distribution network whereas no residual chlorine was found in the source waters from Keenjhar Lake and Hub Dam reservoir. The presence of residual chlorine in the distribution network samples could be explained by the excess chlorination dosage in the finish treated water by the water board (KWSB 2011). The TOC levels range between 0.45 and 11.69 mg/L in distribution network with 3.61 and 3.36 mg/L as mean and median level whereas the source water TOC level as surrogate of a THMs precursor was observed from 2.29 to 8.34 mg/L and 6.0, 6.32 and 2.24 mg/L were estimated mean, median and standard deviation values.

Distribution levels of THMs (Total, $CHCl_3$, $CHCl_2Br$, $CHClBr_2$ and $CHBr_3$) in the water samples of Karachi city are shown in Fig. 2. Figure 2a, b represents the total THMs and $CHCl_3$ concentrations and the values exceeding EPA (80 µg/L) and WHO (100 µg/L) guidelines were also shown as different classes. The concentrations of total THMs in water samples from different parts of Karachi city distribution network were found between 36.04 and 176.24 µg/L (Table 1). The mean concentration of 73.51 ± 35.38 µg/L standard deviation and 60.63 µg/L median values represent central tendency of the total THMs less than the USEPA 80 µg/L standard and WHO 100 µg/L guideline value. However, at places total THM concentration exceed the USEPA and WHO values. No THMs were observed in the raw source water sample. THMs speciation revealed that the $CHCl_3$ is the major contributor among the total THM species which contribute 91.69 %, $CHCl_2Br$, $CHClBr_2$ and $CHBr_3$ contribute 5.69, 1.78 and 0.85 %, respectively (Fig. 3) in the distribution network of Karachi city water supply.

The Pearson correlation among the THM species and other water quality parameters were shown in Table 3. A strong correlation of residual chlorine to $CHCl_3$ ($r = 0.656$) and Total THM ($r = 0.647$), and pH to $CHBr_3$ (0.46) was observed in the present study. The available chlorine and presence of precursor(s) favored the formation of THMs (Krasner 1999; USEPA 2006). In the case of pH and THMs formation, only $CHBr_3$ shows relatively significant correlation among the other THMs. In contrast to positive correlation among TOC and THMs in published literature (Uyak et al. 2005) there was negative or weak correlation ($r = -0.278$) was observed in TOC and total THM formation in the present study. Several factors can account for such effects of TOC and total THMs. TOC

Table 3 The Pearson's correlation matrices of water quality parameters and THMs in water samples

	$CHCl_3$	$CHCl_2Br$	$CHClBr_2$	CHBr3	Total THM	pH	TDS	D.O	Residual Chlorine
$CHCl_2Br$	0.532**								
$CHClBr_2$	0.524**	0.970*							
$CHBr_3$	0.521**	0.899*	0.998*						
Total THM	0.891*	0.573**	0.569**	0.549**					
pH	-0.107	-0.046	0.073	0.460	-0.103				
TDS	0.237	-0.088	-0.015	-0.635*	0.232	-0.059			
D.O	0.052	-0.063	-0.133	-0.859*	0.046	-0.033	0.082		
Residual Chlorine	0.656*	0.141	0.167	0.138	0.647*	0.021	0.155	0.093	
TOC	-0.284	-0.031	-0.03	-0.298	-0.278	-0.045	-0.075	-0.131	-0.177

* Significant at the 0.01 level

** Significant at the 0.05 level

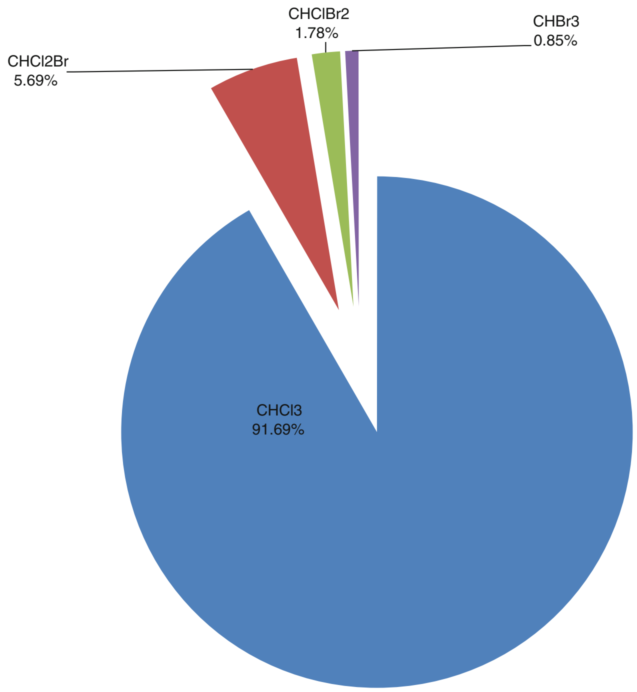

Fig. 3 Mean concentration speciation (%) of the THM in Karachi city water supply network

represents mass organic compounds without any differentiation between precursor and non-precursors to the THM formation (USEPA 2001; Wei et al. 2010).

Geospatial analysis of the THMs in the water distribution network of Karachi was analyzed using ArcGIS 9.3. The Kriging procedure estimates the semivariogram to model the spatial structure of the pollutant, i.e. THMs and predicts the values at unmeasured sites using the fitted model for the spatial behaviour. The THMs concentrations were interpolated with Ordinary Kriging method and presented in Fig. 4 except for $CHBr_3$ due to its limited occurrences and insufficient data for interpolation. The raster maps showed clear distribution of THMs in space. The variations of total THMs levels showed a general trend of decreasing levels of total THMs from east to west, i.e. from main supply of Karachi distribution network towards the receiving ends, probably because the decomposition rate exceeds the formation rate of THMs. The saline water intrusion in the coastal areas can affect the formation of THMs in the presence of residual chlorine (Simpson and Hayes 1998). Higher bromide and iodide content and bromide to TOC and chlorine ratio due to the intrusion of seawater along the coastal areas would increase the THMs

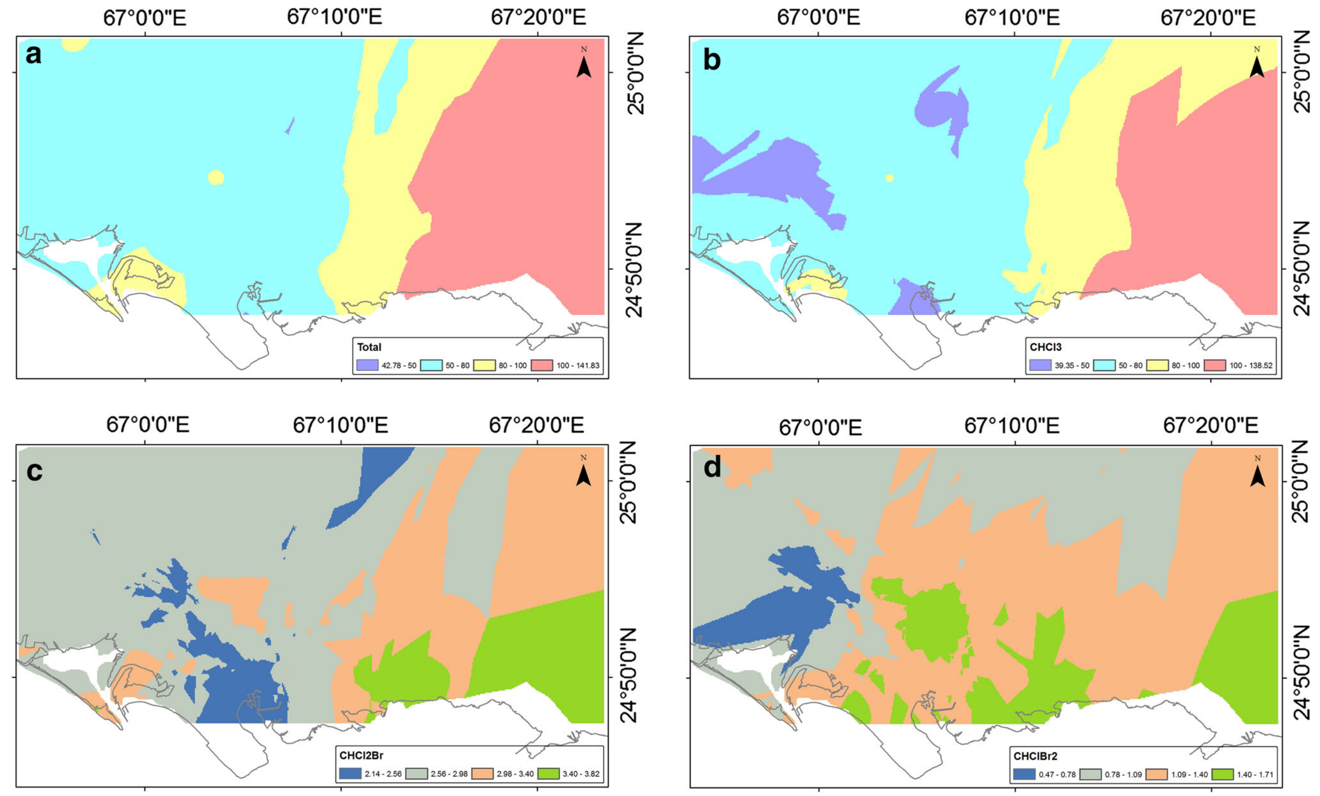

Fig. 4 Geospatial distribution in the water samples of Karachi city: **a** Total THMs, **b** $CHCl_3$, **c** $CHCl_2Br$, **d** $CHClBr_2$

except for bromoform (Zhai et al. 2010). The iodinated THMs are more toxic and their formation is controlled by the ratio of chlorine, bromide and availability of ammonia. Similarly the speciation of chloro and bromo THMs mainly depend on chlorine to bromide ratio and increased availability of chlorine at a given bromide concentration increases the chloro-THMs. (Chang et al. 2001). The presence of higher bromide and elevated pH also leads to higher TTHMs formation (Wang and Huang 2006). Presence of THMs variations at places can also be dependent on the treatment plant operational parameters, natural organic matter removal, chlorine dosage and contact time. It was further revealed that the areas in high demand of water supply and populous show low to moderate concentrations of THMs compared to areas served relatively scattered population. Availability of the contact time between chlorine and NOMs also play an important role in the formation of THMs and the increasing trend in the east or vice versa decreasing trend towards westward could also explain the decreasing availability of chlorine and precursors to form higher concentrations of THMs. $CHCl_3$ being the major and leading component of the total THMs in the observed samples followed the pattern of total THMs. The levels of brominated species $CHCl_2Br$, $CHBr_2Cl$ and $CHBr_3$ were varied insignificantly and observed in a narrow window of concentrations.

Conclusion

A survey of disinfection by-products occurrences in urban Karachi city was conducted throughout the distribution network which serves more than 18 million people. $CHCl_3$ was found to be the major THMs in the drinking water of all the 58 locations followed by $CHCl_2B$, $CHClBr_2$ and $CHBr_3$ respectively. At some locations, Total THMs and $CHCl_3$ levels exceed the WHO (100 µg/L) and USEPA (80 µg/L) prescribed threshold values. THMs show varying nature from high, moderate and low correlations with the observed bulk parameters and themselves. A negative correlation of TOC and total THMs was reported in the present study that may reflect the varying nature of organic compounds comprising precursor and non-precursors of THMs. This research is being continued to provide more insights into the formation of THMs in the complex nature of urban water supplies. The results of the present study have shown that a visible number of THMs are present in the water distribution system of Karachi and due to their significance from public health point of view their continuous monitoring is deemed necessary and establishment of a rational legitimate contaminant level by the concerned authorities is highly recommended.

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