

# Guideline Development History of Trihalomethanes in Drinking Water – A Review in Indian Perspective

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## Abstract

Disinfection by chlorine is a crucial step for the inactivation of microbial pathogens from the drinking water. Throughout this practice, natural organic matter (NOM) presence in water reacts with disinfectant, result in the formation of cancer-causing Trihalomethane (THMs) compounds. The study focuses on the occurrence THMs level in India, and compressive review of its guideline development history by several agencies, and comparison of the same with India regulatory compliance. The Indian interest in THMs was increased in early 1996-97, where the diverse concentration range of THMs (6.03 to 594 µg/l) was reported to date. In most studies, the concentration level of  $\text{CHCl}_3$  in India was observed to be very high. Canada was the first country to develop the guideline value for THMs in 1978, followed by the united state environment protection agency (USEPA) (1979) and the World Health Organization (WHO) (1984). In 2004, the Indian authority Bureau of Indian Standards (BIS) set the first regulatory guideline for all THMs compounds separately. The regulatory compliance of THMs by BIS was found stringent than WHO but more lenient than USEPA. New Zealand and Italy set the highest (400 µg/L) and lowest (30 µg/L) guideline values for THMs worldwide. The present work could prove to be very useful for national or international management bodies to manage and control the level of THMs in drinking water supplies to ensure public health safety.

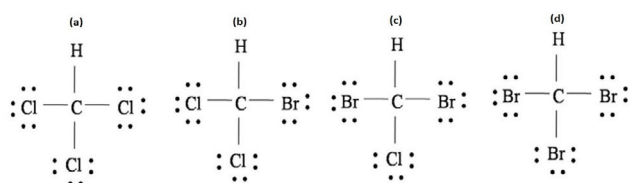
**Keywords:** Trihalomethanes, India, Formation mechanism, Guideline development history

## Introduction

As of now, India is at risk of an acute water crisis. According to a report of “India’s worst water crisis in history -2019”, by the end of 2020, the groundwater of 21 major cities in India will run out, which may affect approx. 0.1 billion inhabitants [1]. Lately, India also dropped down to 2nd last position in the safe drinking water index out of 123 countries across the world [2]. During the year 2010-13, 12,901 deaths were reported due to the unsafe drinking water, with the maximum number in Uttar Pradesh (3382) followed by West Bengal (1778), Andhra Pradesh (1359), and Odisha (730) [2]. Disinfection by chlorine is an extensively used technique for the deactivation of disease-causing microorganisms and to make the water safe for drinking [3,4]. However, the issue associated with the chlorination can not be ignored as it reacts with natural organic matter (NOM) and result in the formation of various disinfection by-products (DBPs),

especially the Trihalomethane (THMs) [2-4]. These THMs compounds include chloroform ( $\text{CHCl}_3$ ), dibromochloromethane ( $\text{CHClBr}_2$ ), bromodichloromethane ( $\text{CHCl}_2\text{Br}$ ), and bromoform ( $\text{CHBr}_3$ ), increased risk of carcinogenicity and mutagenicity as Class B carcinogens [5]. The chemical structure of these four THMs compound are shown in Fig.1. The formation of THMs in chlorinated drinking water was first acknowledged by Rook (1974) [6] and Bellar et al. (1974) [7]. Later in early 1996, the evidence of its occurrence was also investigated in Indian drinking water by many researchers [8,9]. In last 3 to 4 decades, research on THMs received huge attention due to the occurrence of its unacceptable concentration in drinking water and having potential risk to reproductive disorders and many cancer [3,4,10]. The concentration range of THMs in Indian drinking water was varied from 231-511 µg/l [11-13]. Similarly, a diverse range value of the same was also observed in the water supplies system of many countries like Pakistan (575-595 µg/l), Japan (378 µg/l), Canada (137.8–141 µg/l), Turkey (96 -102 µg/l) and China (92.77 µg/l) [10,14-17]. Due to widespread occurrence and carcinogenicity, it was essential to promulgate the regulatory standards of THMs for drinking water to minimize the potential health risk. Many regulatory authorities like the World health organization (WHO), the united state environment protection agency (USEPA), European Union (EU), and countries like China, Australia, Canada, Japan, etc. were established guideline value for THMs according to the suitability of their region.

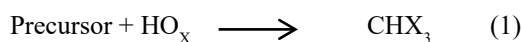
The study aims at to review the development history of THMs standards by various regulatory agencies and compared it with the regulatory compliance of the Bureau of Indian Standard (BIS). In addition, it also covered a brief review of the formation mechanism and the occurrence of THMs level in Indian drinking water. This type of study has not complied yet with particular reference to the Indian perspective. The present work could prove to be very useful for national or international management bodies to manage and control the level of THMs in drinking water supplies to ensure public health safety.



**Figure 1:** Chemical structure (a) Chloroform (b) Bromodichloromethane (c) Dibromochloromethane (d) Bromoform [18].

### Mechanism of THMs Formation

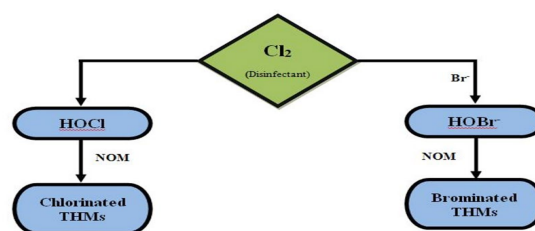
In the literature, only limited information is available to reveal the mystery of THMs formation during the chlorination process. According to the Chlorine residual testing fact sheet of the Centers for Disease Control and Prevention, when the chlorine added to the water, the various transformation occurred (Figure 2). This aqueous chlorine reacts with the NOM precursor such as humic and fulvic acid in raw water and results in the formation of THMs [12]. During this formation process, multi-step reactions occur; in the first steps, organochlorine intermediates are produced, and then in the second stage, it is converted into THMs [11]. The formation of THMs is generalized by the following equation (1):



Where X may be chlorine or bromide, and  $\text{CHX}_3$  may be regarded as a general formula for THMs.

Eq. (1) clearly illustrated that THMs are the class of chemical compounds mainly derived from methane ( $\text{CH}_4$ ), where three of the four hydrogen atoms have been replaced by halogens. The chloroform was found to be the principal compound in chlorinated drinking water [10]. However, the water containing bromides, the concentrations of  $\text{CHCl}_3$  decrease with the formation of brominated THMs [7]. In addition, bromide is rapidly oxidized by the free chlorine into hypobromous acid ( $\text{HOBr}$ ), which react with NOM precursors and results in the formation of brominated and mixed chlorobromo by-products ( $\text{CHBr}_3$ ,  $\text{CHBrCl}_2$ , and  $\text{CHClBr}_2$ ) [2,7].

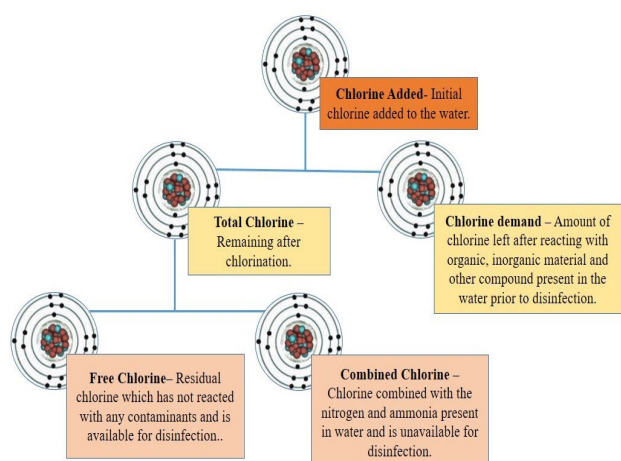
The basic concept of THMs formation pathway is illustrated in Fig. 3. The THMs formation is also greatly influenced by some water quality parameters like temperature, pH, concentration of NOM, and contact time [19,20].



**Figure 3:** Basic concept of THMs formation pathway

### Status of THMs in Indian drinking water

The evidence of the first existence of THMs in drinking water of various major cities (Agra, Ahmedabad, Bombay, Calcutta, Delhi, Goa, Guna, Kanpur Madras, and Nagpur) in India was reported by Thacker et al. (1996) [8]. The concentration was not mentioned; however, they confirmed the occurrence of THMs in the drinking of these cities. After this investigation, India also received huge attention to the research of THMs, where a diverse concentration range was observed in various parts of the country by many researchers (Table.1). In 1997, Srikanth monitored THMs in the municipal drinking water of Hyderabad city and set the range value of the  $\text{CHCl}_3$  (0.0 to 86.5  $\mu\text{g/l}$ ) [21]. Later, by the year, the elevated concentration range of  $\text{CHCl}_3$  was observed in the chlorinated drinking water of cities like Delhi (311-377  $\mu\text{g/l}$ ), Kolkata (466  $\mu\text{g/l}$ ), Dhanbad (503  $\mu\text{g/l}$ ), Bokaro (594), Varanasi (380.9  $\mu\text{g/l}$ ), Raipur (324.3  $\mu\text{g/l}$ ), and Bhubaneswar (319.7  $\mu\text{g/l}$ ) [3,21-24]. The  $\text{CHCl}_3$  was also only the compound identified at all locations and reported to be the most dominant THMs compound in chlorinated drinking water [3,18]. The concentration of  $\text{CHCl}_2\text{Br}$  and  $\text{CHClBr}_2$  was fairly good for all the locations except in Kalpakkam. In another study by an unexpected range value of  $\text{CHBr}_3$  (9.78– 1854.90  $\mu\text{g/l}$ ) was monitored in the water sample of the various thermal power station (Bokaro, Chandrapura, and Durgapur) [25]. The variation in the concentration range of these THMs compounds is greatly influenced by the fluctuation of operational parameters (pH, residual chlorine, temperature) and seasonal and geographical distribution of the location [22,23].



**Figure 2:** Chlorine addition flowchart (CDCP 2012)

**Table1:** Status of THMs compound in Indian drinking water

Sl. No	City/Location	Concentration range of THMs (µg/l)				Reference
		CHCl <sub>3</sub>	CHCl <sub>2</sub> Br	CHClBr <sub>2</sub>	CHBr <sub>3</sub>	
1	Agra, Ahmedabad, Bombay, Calcutta, Delhi, Goa, Guna, Kanpur Madras and Nagpur	Presence	Presence	Presence	Presence	Thacker et al. (1996)
2	Hyderabad	0.0 to 86.5	---	---	---	Srikanth (1997)
3	Mumbai	29.1 to 231.26	87.36	0.63 to 50.2	46.78	Thacker et al. (2002)
4	Gantok	36.50	8.70	7.70	5.10	Sharma and Goel (2007) [26]
5	Delhi	311 to 377	113.3	---	---	Hasan et al. (2010)
6	Bokaro (Thermal Power Station)	232.00 41.70	41.70	41.70	868.00	
7	Chandrapura (Thermal Power Station, Bokaro)	203.00	32.10	105.00	428.00	Basu et al. (2011)
8	Durgapur (Thermal Power Station)	222.00	97.70	19.20	19.20	
9	Lucknow	13.84 to 74.12	4.71 to 62.69	1.10 to 8.46	1.06 to 5.23	Singh et al. (2012)
0	Kalpakkam	98	185	201	---	Rajamohan et al. (2012) [27]
11	Gwalior	6.03	16.16	16.16	0.13	Nisha et al. (2013) [28]
12	Kanpur	77.6 to 259.64	---	---	---	Mishra and Dixit (2013)
13	Kolkata	466	12	2	---	
14	Durgapur	255	8	11	---	Minashree Kumari (2014)
15	Ranchi	236	14	31	---	Minashree Kumari (2014)
16	Dhanbad	503	4	2		Minashree Kumari (2014)
17	Bokaro Steel City	594	---	---	---	Mishra et al. (2014)
18	Dhanbad, Raniganj, Barrackpore, and Ranchi	231 to 484	---	---	---	Kumari and Gupta (2015)
19	Varanasi	380.9	18.3	15.5	---	
20	Dhanbad	360.2	16.9	12.3	---	
21	Raipur	324.3	21.7	14.2	---	Mahato and Gupta (2020)
22	Bhubaneswar	319.7	20.3	8.5	---	
23	Barrackpore (Kolkata)	353.1	18.8	12.1	---	

### THMs guideline development history

Disinfection of drinking water by using chlorine was first practiced in Chicago (United States) in the year 1908, which spread quickly worldwide [3]. Later, in 1974 the discovery of THMs alarmed the world to concern about its guideline value to minimize the possible adverse health effect.

### USEPA

In the five years after THMs discovery (1979), USEPA established the permissible limit for total THMs (TTHMs) (100 µg/L) under the safe drinking water act [29]. Later, in 1998 under the Stage 1 DBPs Rule, USEPA lowered its permissible limit to 80 µg/L [30]. Although, the implementation of Stage 2 DBPs rule in March 2006 maintained the same guideline value as of Stage 1. (USEPA 2006). According to 2012, Edition of the Drinking Water Standards and Health Advisories of USEPA, a maximum contaminant level (MCL) of 80 µg/L was set for all the individual THMs compounds, and no change was made to its latest edition of 2018 (USEPA 2012; USEPA 2018). Table.2 depicted the THMs guideline development history of USEPA.

**Table 2:** THMs guideline History of USEPA

Sl.No	THMs rule	Maximum Contamination Level (MCL) µg/L					References
		TTHMs	CHCl <sub>3</sub>	CHCl <sub>2</sub> Br	CHClBr <sub>2</sub>	CHBr <sub>3</sub>	
1	THMs Rule (1979)	100	---	---	---	---	Simpson and Hayes (1998)
2	Stage 1 DBPs rule (1998)	80	---	---	---	---	Tak et al. (2020)
3	Stage 2 DBPs rule (1998)	80	---	---	---	---	USEPA (2006)
4	Drinking Water Standards and Health Advisories (USEPA, 2012-18)	---	80	80	80	80	USEPA (2012-18)

### WHO

In the first edition of Guidelines for Drinking-water Quality by WHO in 1984, no guideline values for THMs other than CHCl<sub>3</sub> was proposed [31]. Though the chloroform was the most commonly encountered member of THMs group, so a health-based guideline value of 300 µg/L was recommended for it [22]. Later, in the second edition published in 1993, WHO lowered the permissible limit of CHCl<sub>3</sub> (200 µg/L) and also established separate guidelines for all THMs compounds (Table. 3).

The same guideline value of all four THMs compound was brought forward to the third edition (1998-2004). In the latest edition, released in 2011, WHO recommended following the guideline of the first edition only for  $\text{CHCl}_3$ , whereas the guideline value remained the same for other compounds as of the third edition. The complied guideline value of all the edition proposed by WHO is illustrated in Table.3.

**Table 3:** THMs guideline history of W.H.O

Sl.No	Edition	Guideline value ( $\mu\text{g/L}$ )					References
		TTHMs	$\text{CHCl}_3$	$\text{CHCl}_2\text{Br}$	$\text{CHClBr}_2$	$\text{CHBr}_3$	
1	1st edition (1984)	---	300	---	---	---	WHO (2006)
2	2nd edition (1993)	---	200	60	100	100	WHO (2006)
3	3rd edition (1998-2004)	---	200	60	100	100	WHO (2006)
4	Latest edition (2011)	---	300	60	100	100	WHO (2011)

## India

Indian interest in THMs had increased in early 1996-1997 when Thacker et al. (1996) confirmed THMs in chlorinated drinking water [8]. However, the guideline value of THMs in India was promulgated late in the year 2004 by BIS. This year, Indian authorities set the individual permissible for all THMs compounds similar to the guideline value of the 3rd edition of WHO (1998-2004) (Table.4). During the second revision of the draft Indian standard drinking water specification (IS 10500) in 2009, BIS established a single guideline value of  $100 \mu\text{g/L}$  for all four THMs compounds (IS 10500 2009) [33,34]. Further in 2012, in the second revision of final Indian standard drinking water specification, these guideline value was again revised and suggested to remain same as per IS 10500 (2004) (Table.4).

**Table 4:** THMs guideline history of BIS

Sl No	Edition	Guideline value ( $\mu\text{g/L}$ )					References
		TTHMs	$\text{CHCl}_3$	$\text{CHCl}_2\text{Br}$	$\text{CHClBr}_2$	$\text{CHBr}_3$	
1	IS 10500, (2004)	---	200	60	100	100	IS 10500 (2004)
2	IS 10500, (2009)	---	100	100	100	100	IS 10500 (2009)
3	BIS, 2012	---	200	60	100	100	BIS 2012

## In Other countries

Canada became the first country to set the guideline value of total THMs (TTHMs) ( $350 \mu\text{g/L}$ ) in 1978 [31]. This guideline was revised in 1996 and reduced the value to  $100 \mu\text{g/L}$ . Again in the year 2006, the revision of Guideline for Canadian Drinking-water Quality re-affirmed the value of TTHMs at  $100 \mu\text{g/L}$ . Also, it proposed an individual guideline value for  $\text{CHCl}_2\text{Br}$  ( $16 \mu\text{g/L}$ ) [31]. In the second revision of the guideline for Drinking Water Quality 1985, China proposed its standards for  $\text{CHCl}_3$  ( $60 \mu\text{g/L}$ ) [35]. Further, the standards were revised in 2006 and set separate guideline values for all THMs compounds (Table.5).

For the first time in 1995, the European Union (EU) recommended the permissible limit only for two THMs compound viz.  $\text{CHCl}_3$  ( $40 \mu\text{g/L}$ ) and  $\text{CHCl}_2\text{Br}$  ( $15 \mu\text{g/L}$ ) [36]. Later in 1998, EU established a single guideline value only for TTHMs which was brought forward to 2007 and in 2014 (EU 2014) (Table.4) [37]. The standard for THMs in Australia was first drawn in 1996 under the Australia Drinking Water Guideline (ADWG). A guideline value of  $250 \mu\text{g/L}$  was proposed for TTHMs [31]. The ADWG 1996 was revised in 2004 and then in 2011, but no change was made in the guideline of THMs (Table.5). Many countries and regions across the world follow the THMs guideline value of either WHO or USEPA. However, countries like the UK, Taiwan, New Zealand, South Africa, Japan, Italy, and Korea promulgated their THMs standards for drinking water to ensure public health safety (Table.5).

**Table 5:** THMs guideline value in various countries

Sl No	Country/ Organization	Guideline value ( $\mu\text{g/L}$ )					References
		TTHMs	$\text{CHCl}_3$	$\text{CHCl}_2\text{Br}$	$\text{CHClBr}_2$	$\text{CHBr}_3$	
1	CANADA (2012)	100	16	---	---	---	Hrudey and Charrois (2012)
2	CHINA (2014)	---	60	60	100	100	Wang et al. (2015)
3	EU (2014)	100	---	---	---	---	EU (2014)
4	AUSTRALIA (2013)	250	---	---	---	---	Hrudey and Charrois (2012)
5	JAPAN (2009-10)	100	60	30	100	90	HASANI et al. (2010); Sharma et al. (2009) [38,39]
6	UK (2010)	100	---	---	---	---	HASANI et al. (2010)
7	Korea (2010)	100	---	---	---	---	HASANI et al. (2010)
8	Tiwan (2010)	100	---	---	---	---	HASANI et al. (2010)
9	New Zealand (2009)	---	400	60	150	100	Sharma et al. (2009)[38]
10	Italy (2005-2017)	30	---	---	---	---	Villanueva et al. (2017)
11	South Africa (2015)	---	300	60	100	100	SANS (2015) [41]



## Conclusion

From the review of the various study, it is concluded that Indian water produced a high concentration of THMs upon chlorination in most cases. An unexpected value of  $\text{CHBr}_3$  (1850  $\mu\text{g/L}$ ) was also seen in the thermal power plant's drinking water. In most studies, the concentration of  $\text{CHCl}_3$  was observed beyond the guideline value of BIS, WHO, and USEPA. The promulgated standards of THMs in India are stringent than WHO but more lenient than USEPA. New Zealand and Italy set the highest (400  $\mu\text{g/L}$ ) and lowest (30  $\mu\text{g/L}$ ) guideline values for THMs worldwide. At this time, India needs to modify the existing water treatment technology with an advanced system to meet the safe drinking water supplies concerning THMs [41-50].

## Authors' Contributions

Jaydev Kumar Mahato (First author): Investigation and Writing - original manuscript. Sunil Kumar Gupta (Corresponding author): Conceptualization and Supervision.

**Conflicts of Interest** The authors declare no conflicts of interest.

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