Oxyhalide disinfection by-products in packaged drinking water and their associated risk

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Disinfection by-products (DBPs) are formed during treatment of water for drinking purposes. Among these oxyhalide DBPs, viz. bromate, chlorate and chlorite are potentially harmful to humans. In the present study packaged drinking water samples of various commercial brands, available in Mumbai, were analysed for bromide, bromate, chlorite and chlorate ions using ion chromatography. The average concentration levels of bromide, bromate, chlorite and chlorate in packaged drinking water were 28.4, 10.7, 7.1 and 20.8 µg/l respectively. Bromate in 27% samples was found to be higher than the World Health Organization (WHO) guideline value of 10 µg/l, whereas chlorite and chlorate levels were well within the guideline value recommended by WHO. A good correlation between bromide and bromate ($r = 0.85$) indicated formation of bromate from bromide present in drinking water. Health risks (carcinogenic risk and hazard quotient) associated with ingestion of bromate, chlorite and chlorate present in packaged drinking water are presented and discussed.

Keywords: Bromate, chromatography, disinfection by-products, packaged drinking water, water treatment

DISINFECTION is a part of the treatment process in which pathogenic organisms are eliminated by chemical (chlorination/ozonation treatment) and physical (UV irradiation) agents. During this process disinfection by-products (DBPs) are formed when the disinfectants used react with halides and/or the natural organic matter present in raw water. The inorganic 'oxyhalide' group of DBPs include bromate, chlorite and chlorate. These oxyhalide DBPs are known to cause significant health risks, even at low (µg/l) levels in drinking water. Bromate (BrO_3^-) is identified as a potential carcinogen and World Health Organization (WHO) recommended a guideline value of 10 µg/l for bromate in drinking water[1]. Chlorite and chlorate have shown to cause hemolytic anaemia in the laboratory studies on animals[2]. WHO has stipulated a guideline value of 700 µg/l for both chlorite and chlorate in drinking water.

Packaged drinking water is a popular alternative to water from public drinking water supply in India. Many brands of packaged drinking water are available in the country and its market is growing rapidly. The contaminant levels in packaged drinking water in India are regulated by the Bureau of Indian Standards (BIS), which gives an Indian Standards Institute (ISI) mark for meeting the regulatory limits. In general, raw water used for production of packaged drinking water is derived from groundwater using bore wells and from public drinking water supply system. BIS suggested treatment methodology for packaged drinking water includes pressure sand filtration, activated carbon filter, reverse osmosis, ozonation and UV treatment[3]. BIS has recommended standards for packaged drinking water in its document IS 14543. The recommended standards include concentration-based limits for physical (colour, taste, turbidity, etc.), chemical (trace and toxic elements, ions, etc.) and radioactive (alpha and beta emitters) parameters. However, for bromate, chlorite and chlorate standards are not recommended by BIS.

In view of the above, a study has been carried out with an objective to determine bromate, chlorite and chlorate in packaged drinking water in different commercial brands available in Mumbai. This study would be useful for regulatory agencies to recommend standards for the above oxyhalide DBPs that are present in drinking water. Also, an attempt has been made to estimate carcinogenic and non-carcinogenic (hazard quotient) risks associated with ingestion of these oxyhalide DBPs.

Bromate is formed during disinfection of water containing bromide with ozone. When bromide containing water is treated with ozone, bromide is oxidized to hypobromous acid (HOBr) and dissociation of acid results in the formation of hypobromite (OBr^-). Hypobromite ion further reacts with ozone to form bromate. This reaction is favoured at higher pH values.

$$Br^- + O_3 + H_2O \rightarrow HOBr + O_2 + OH^-, \quad (1)$$

$$HOBr + H_2O \rightarrow H_3O + OBr^-, \quad (2)$$

$$OBr^- + 2O_3 \rightarrow BrO_3^- + 2O_2. \quad (3)$$

Bromate is also formed by photochemical oxidation of bromide ions in chlorinated water[4]. Also, when water containing bromide and residual chlorine undergoes UV processing bromate is formed[5]. Under certain conditions bromate is also formed in concentrated hypochlorite solutions used for disinfection of drinking water[6]. Formation of bromate in water depends upon a variety of water quality and operational parameters such as bromide concentration, pH, concentration of dissolved organic matter, contact time, dosage of disinfectant, etc. Bromate once formed in aqueous solution is highly stable at room temperature, does not volatilize and will not be removed by boiling[7].

Chlorite and chlorate are formed during ozonation of chlorine present in water[8]. Residual chlorine reacts with water to form hypochlorous acid (HOCl). HOCl further

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*For correspondence. (e-mail: ggp@barc.gov.in) dissociates into hydrogen and hypochlorite ions. Ozone reacts with hypochlorite ion producing chlorite which is quickly oxidized to chlorate[9]

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^-, \quad (4)$$

$$HOCl \rightarrow H^+ + OCl^-, \quad (5)$$

$$O_3 + OCl^- \rightarrow O_2 + ClO_2^-, \quad (6)$$

$$O_3 + ClO_2^- \rightarrow O_2 + ClO_3^-. \quad (7)$$

Chlorate is also formed during storage of sodium hypochlorite solution[10]. Similar to bromate formation of chlorite and chlorate also depends on parameters like pH, organic matter, concentration of residual chlorine, contact time, dosage of disinfectant, etc. Chlorate once formed is stable in water and generally cannot be removed by common treatment methods.

Eighteen different commercial brands of 500 ml packaged drinking water were purchased from the local markets of Mumbai for analysis. The treatment method used by various brands was noted from the labels affixed on the bottles. pH and total dissolved solids (TDS) were measured on the same day of collection. The collected samples were filtered through cellulose acetate filters of 0.22 μm pore size and stored in a refrigerator at 4°C in precleaned glass vials for analysis of oxyhalides using ion chromatography. Sample analysis was carried out within a week of collection.

pH and TDS of packaged drinking water samples were measured using Eutech pH and conductivity meter (model: PC 510) respectively. Calibration of pH meter was carried out using standard buffers of pH 4, 7 and 9, and calibration for TDS measurements was carried out using standard KCl solution of different concentrations.

Bromide and oxyhalide DBPs in packaged drinking water samples were analysed using ion chromatograph (Metrohm make, 733 IC Separation Centre) with conductivity detector. Metrosep A Supp 5 (250) column was used as stationary phase, along with a suitable precolumn. Also, 3.2 mmol sodium carbonate and 1.0 mmol sodium bicarbonate mixture was used as mobile phase for analysis. Flow rate of mobile phase was optimized to 0.6 ml/min for clear separation of the ions. The background conductivity after chemical suppression was 14 μS/cm. Sample injection volume was optimized to 100 μl loop size after a number of trials with 20 μl, 100 μl and 500 μl loops. In 20 μl loop, sample volume was not sufficient for detection of lower ppb (μg/l) levels without pre-concentration. With 500 μl loop, though the detection limit had improved, sample carry over was observed in successive injections. The 100 μl loop provided detection limit in ppb range and did not show any sample carry-over effect. A typical chromatogram of a sample analysed is presented in Figure 1.

Standard was injected seven times repeatedly to determine the standard deviation. By multiplying with 3.14 times the lowest standard, detection limit for each ion was determined[11]. The detection limits obtained for different ions are presented in Table 1. Precision of the method was evaluated by five successive determinations of mix standards and the relative standard deviation (RSD) for the ions was in the range 2–3%. Recovery of ions was checked by spiking the drinking water sample with standards and the recovery ranged from 90% to 98% (Table 1).

Range and average values of pH, TDS, bromide and oxyhalide DBPs measured in packaged drinking water samples are presented in Table 2. The pH values ranged from 6.58 to 7.10 with a mean value of 6.9, while TDS values ranged from 28.6 to 146 mg/l with a mean value of 68.1 mg/l. The pH and TDS levels of all the samples are well within the limits specified by BIS for packaged drinking water[12]. Bromide concentration ranged from 6 to 73 μg/l with an average of 28.4 μg/l. Bromate levels varied from below detection limit to 43 μg/l with an average of 10.7 μg/l. Chlorite ranged from below detection limit to 18 μg/l with an average of 7.1 μg/l and chlorate ranged from 5 to 50 μg/l with an average concentration of 20.8 μg/l. Figure 2 presents the concentration of measured ions in different brands of packaged drinking water collected in Mumbai. As shown in the figure wide variations are observed in the concentration of ions for different brands of drinking water. Among the different brands analysed, water quality of some were found to be exceeding the WHO limit for bromate. In one brand (BW2) bromate level of 43 μg/l was observed, with chlorate and chlorite levels of 28 and 9 μg/l respectively. However, in some brands water quality found to be good with values well below the recommended limits and one brand (BW17) had bromate below detectable limit and chlorite and chlorate levels were 6 and 15 μg/l respectively. In many samples chlorite levels were below the detectable limit as it is the intermediate product formed which gets converted to chlorate during ozonation[8]. Some brands use activated carbon filtration (ACF) as one of the treatment methods for removal of trace contaminants. In these brands (BW4, BW7) very low level of bromate was observed indicating adsorption by activated carbon. The use of activated carbon is one of the suggested methods for reduction of bromate in drinking water[13,14]. The variations in the levels of DBPs and bromide in drinking water samples could be due to different sources of raw water used and different treatment methodologies adopted for purification. Bromate level in 27% (five nos) of drinking water samples exceeded the WHO recommended guideline value[1] of 10 μg/l. Whereas chlorite and chlorate levels in drinking water samples were well below the WHO recommended value[1] of 700 μg/l. The observed chlorite and chlorate in samples indicated the presence of chlorine in raw water. Chlorine (in the form of residual

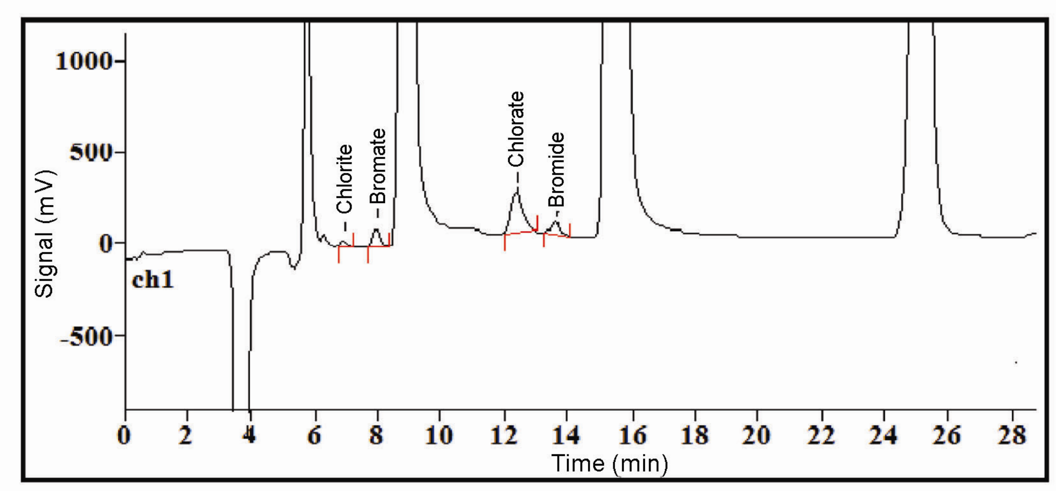

Figure 1. A typical chromatogram of drinking water sample.

Table 1. Detection limits and recovery of bromide, bromate, chlorite and chlorate

Analyte	Detection limit (μg/l)	Recovery (%)
Bromide	0.5	90
Bromate	0.7	94
Chlorite	0.7	98
Chlorate	0.7	92

Table 2. Range and average values of pH, TDS, bromide, bromate, chlorite and chlorate levels in packaged drinking dater

Parameters	Range	Average	Limits
pH	6.58–7.10	6.9	6.5–8.5 (ref. 12)
TDS (mg/l)	28.6–146	68.1	500 (ref. 12)
Bromide (μg/l)	6–73	28.4	–
Bromate (μg/l)	<0.7–43	10.7	10 (ref. 1)
Chlorite (μg/l)	<0.7–18	7.1	700 (ref. 1)
Chlorate (μg/l)	5–50	20.8	700 (ref. 1)

chlorine) is generally maintained in water supplied by the municipal authority to take care of microbial contamination, if any, during distribution. When this water is used by packaged drinking water plants, chlorine is converted to chlorite and chlorate during disinfection by ozonation/UV treatment. However, bromide is naturally present in water and exists as salts with sodium, potassium and other cations. Concentration of bromide in freshwater typically ranges[15] from trace amounts to 0.5 mg/l. Bromide in raw water is converted to bromate during disinfection by ozonation/UV treatment. Since formation of bromate depends on bromide concentration in water, a correlation study has been carried out between bromide and bromate. Figure 3 presents the correlation plot between bromide and bromate. As shown in the figure the two ions are reasonably well correlated with Pearson correlation coefficient of 0.85. Since formation of bromate also depends on a variety of other parameters (such as pH of water, dosage of disinfectant, contact time in addition to bromide concentration), a better correlation is not expected[16].

Table 3 presents the range and average values of bromate, chlorite and chlorate in drinking water samples measured by other researchers along with the values measured during the present study. As indicated in Table 3, chlorite and chlorate levels in the present study fall within the range reported in the literature. Similarly, bromate levels are also within the range observed by other researchers; particularly, the levels are comparable with the study carried out in Mumbai[6].

Based on the observations of the present study and by comparing with the literature data, bromate in few samples was found to be above the recommended limit, whereas chlorite and chlorate were well below the limits. In the absence of regulations for these DBPs, no specific treatment is being carried out by the manufacturers for removal of bromate. In order to regulate the bromate levels in water either the precursor, i.e. bromide should be removed prior to ozonation, or a suitable treatment must be used for removal of bromate. Bromide in water can be removed by employing treatments such as precipitation, ion exchange and membrane filtration[13]. Bromate formed during ozonation can be removed by ACF[13,14] and other treatment methods. As discussed earlier, in the brands that used ACF for removal of trace contaminants, bromate levels were found to be low. Therefore, using ACF treatment after ozonation bromate levels can be controlled in drinking water.

Risk assessment studies are useful to understand the health and environmental impacts associated with exposure to environmental contaminants. The United States Environmental Protection Agency (USEPA) has proposed guidelines for estimation of risk from exposure to contaminants through different pathways (inhalation, ingestion, etc.). In general, risk is estimated in terms of carcinogenic and non-carcinogenic risk.

Carcinogenic risk is estimated using the following equation

$$R_c = I_c \times \text{CSF}, \tag{8}$$

where R_c is carcinogenic risk, I_c is chronic daily intake of carcinogenic substance (mg/kg/d) and CSF is cancer slope factor of carcinogen ((mg/kg/d)$^{-1}$).

Non-carcinogenic risk is estimated in terms of hazard quotient (HQ) using the following equation

$$\text{HQ} = \frac{I_n}{\text{RfD}}, \tag{9}$$

where I_n is chronic daily intake of non-carcinogenic substance (mg/kg/d) and RfD is reference dose (mg/kg/d). For any compound hazard quotient value less than one is considered as safe, as RfD is the threshold dose above which adverse effect is observed[17].

The CSF and RfD values for various chemicals is available in the Integrated Risk Information System (IRIS) database developed by USEPA. Chronic daily intake for carcinogens (I_c) and noncarcinogens (I_n) is estimated using the equation

$$\text{Intake } (I_c \text{ or } I_n) = \frac{C_i \times \text{IR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}}, \tag{10}$$

where C_i is the contaminant concentration (mg/l); IR the ingestion rate of water (l/day), EF the exposure frequency (days/yr), ED the exposure duration (yrs), BW the average body weight (kg) and AT is the averaging time (days).

Averaging time will depend on the type of constituent being evaluated. For chronic effects associated with noncarcinogens intake is averaged over exposure duration (i.e. AT = ED), whereas for carcinogens intake is averaged over lifetime, to be consistent with the approach used to develop cancer slope factors[17].

Since bromate is considered as a probable human carcinogen (category B2)[18], carcinogenic risk was estimated for bromate and non-carcinogenic risk was estimated for bromate, chlorite and chlorate. Table 4 presents the values of different parameters used for estimation of

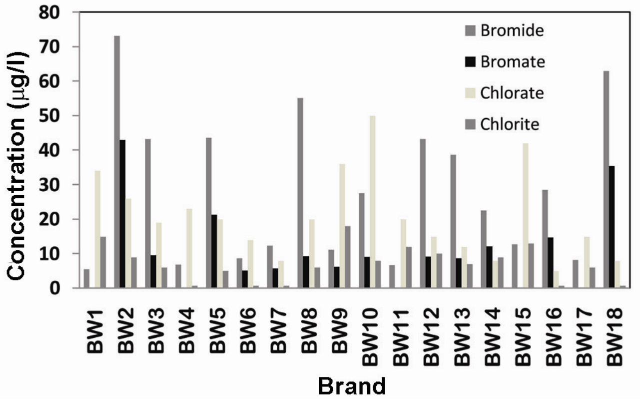

Figure 2. Concentration of bromide, bromate, chlorate and chlorite in different brands of packaged drinking water.

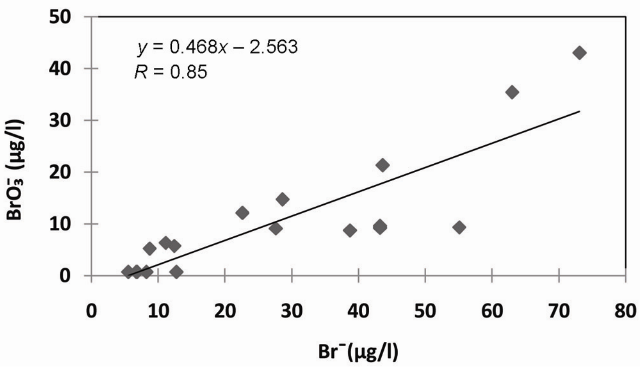

Figure 3. Correlation plot between bromide and bromate in packaged drinking water.

Table 3. Comparison of concentration levels of bromate, chlorite and chlorate in drinking water

Bromate (µg/l)	Chlorate (µg/l)	Chlorite (µg/l)
3.5–3.9 (ref. 21)	<7 (ref. 21)	30.6–22.5 (ref. 21)
<0.1–76 (ref. 20)	<0.1–5.8 (ref. 20)	–
<0.56–169 (ref. 22)	–	–
6–65 (ref. 6)	–	–
<3–178 (ref. 23)	–	–
0.32–2.58 (ref. 16)	–	–
5.34 (ref. 24) (maximum)	177 (ref. 24) (maximum)	–
–	130 (ref. 25) (winter) 210 (ref. 25) (summer)	360 (ref. 25) (winter) 480 (ref. 25) (summer)
<0.7–43*	5–50*	<0.7–18*

*Present study.

Table 4. Parameters and their values used for calculation of carcinogenic risk and hazard quotient

Parameters	Value used for risk calculation
Bromate (mg/l)	10.7×10^{-3}
Chlorate (mg/l)	20.8×10^{-3}
Chlorite (mg/l)	7.1×10^{-3}
Ingestion rate (l/day)	2 (ref. 26)
Body weight for adult male (kg)	70 (ref. 17)
Exposure frequency (days/yr)	350 (ref. 26)
Exposure duration (yrs)	30 (ref. 26)
Averaging time for carcinogenic risk (days)	350 × 70 (ref. 17)
Averaging time for hazard quotient (days)	350 × 30 (ref. 17)
Cancer slope factor for bromate (mg/kg/d)$^{-1}$	0.7 (ref. 27)
Reference dose (mg/kg/d)	
Bromate	4.0×10^{-3} (ref. 27)
Chlorite	3.0×10^{-2} (ref. 28)
Chlorate	3.0×10^{-2} (ref. 29)

Table 5. Statistical parameters of disinfection by-products (DBPs) in packaged drinking water and their associated risk different percentiles due to ingestion of bromate, chlorite and chlorate

DBPs	Mean	Median	Minimum	Maximum	5th percentile	25th percentile	75th percentile	99th percentile
Bromate (μg/l)	10.7	8.9	0.7	43	0.7	1.8	11.5	41.7
Chlorite (μg/l)	7.1	6.5	0.7	18	0.7	1.8	9.8	17.5
Chlorate (μg/l)	20.8	19.5	5	50	7.6	12.5	25.3	48.6
Carcinogenic risk for bromate	9.17×10^{-5}	7.62×10^{-5}	6.0×10^{-6}	3.68×10^{-4}	6.0×10^{-6}	1.54×10^{-5}	9.85×10^{-5}	3.57×10^{-4}
Hazard quotient for bromate	7.64×10^{-2}	6.36×10^{-2}	5.0×10^{-3}	3.07×10^{-1}	5.0×10^{-3}	1.29×10^{-2}	8.21×10^{-2}	2.98×10^{-1}
Hazard quotient for chlorite	6.73×10^{-3}	6.19×10^{-3}	6.67×10^{-4}	1.71×10^{-2}	6.67×10^{-4}	1.71×10^{-3}	9.33×10^{-3}	1.67×10^{-2}
Hazard quotient for chlorate	1.98×10^{-2}	1.86×10^{-2}	4.75×10^{-3}	4.76×10^{-2}	7.24×10^{-3}	1.19×10^{-2}	2.41×10^{-2}	4.63×10^{-2}

noncarcinogenic and carcinogenic risk. For the present study exposure duration of 30 years was considered and risk was estimated for adult male.

The estimated basic statistical parameters of bromate, chlorite and chlorate in packaged drinking water along with their percentile values are presented in Table 5. The estimated carcinogenic risk and hazard quotient due to ingestion of drinking water containing oxyhalide DBPs are also presented in Table 5. The risk is calculated at different percentile values covering a wide range from a minimum to the most conservative estimate. As shown in the table, the individual excess cancer risk due to ingestion of bromate present in packaged drinking water was observed to be in the range 6.0×10^{-6}–3.68×10^{-4} with a mean value of 9.17×10^{-5}. In the worst-case scenario (99th percentile), the excess cancer risk was about 3.57×10^{-4}. The range of risk levels is observed to be significant as USEPA advises consideration of more conservative risk levels[18] of the order of 10^{-5} or 10^{-6}. Considering 10^{-5} as an acceptable risk, the risk estimate for the worst-case scenario (99th percentile) is 35 times higher than the acceptable risk. The hazard quotient for bromate is in the range 5.0×10^{-3}–3.07×10^{-1}, which is less than unity indicating bromate in bottled drinking water is within the safe limits from chemical toxicity point of view. The hazard quotient values of chlorite and chlorate are in the range 6.67×10^{-4}–1.71×10^{-2} and 4.75×10^{-3}–4.76×10^{-2} respectively, which are much less than unity. These values clearly indicate the chlorite and chlorate levels are within the safe limits even for the worst-case scenario.

The present study highlights the need for determination of DBPs present in drinking water that are harmful and also creates awareness about recommendation of regulatory limits for these DBPs. Several agencies world over[1,19] have recommended regulatory limits for bromate, chlorite and chlorate in drinking water and studies carried out by various researchers highlight the importance of monitoring and regulation of these DBPs in drinking water[20]. The risk estimates of the present study further establish the need for the setting up of regulatory limits for DBPs, particularly for bromate.

In the present study oxyhalide, DBPs were analysed in packaged drinking water samples by optimizing a method using ion chromatography. The results showed that bromate, chlorite and chlorate were detected in a number of samples and bromate in 27% of samples analysed was more than the guideline value recommended by WHO. Correlation study indicated formation of bromate from bromide present in water. The study also included health-risk assessment due to ingestion of packaged drinking water containing bromate, chlorite and chlorate. The estimated lifetime excess cancer risk (average) due to ingestion of bromate through drinking water was 9.17×10^{-5}, whereas the hazard quotient was much less than unity. The present study is useful for Indian regulatory agencies to decide on recommending regulatory limits for oxyhalide DBPs, viz. bromate, chlorite and chlorate in packaged drinking water.

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Post-disaster assessment of impact of cyclone *Lehar* in South Andaman Island

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Disasters are the events which devastate human lives, properties and natural ecosystems; cyclones are frequent events in tropical countries which have adverse impacts on coastal areas. The present study evaluates the impact of cyclone *Lehar* post-disaster in the South Andaman Island using geographic information system (GIS) and remote sensing techniques. Cyclone *Lehar* originated in the Andaman Sea and had a major impact on the South Andaman Island. Digital elevation model was used to create elevation and slope maps of the study area. These maps were used to study the impacts of floods, landslides, storm surges and runoff. Land-use and land-cover features were mapped and overlaid with this model for preparing the vulnerability map for various outcomes of the cyclone. Preliminary impact assessments were made in these identified vulnerable areas and also throughout the study area. Finally the results were interpreted with vulnerability map prepared using the GIS technique which shows that most of the affected areas are correlated with the vulnerability map. Cyclone *Lehar* had adverse impacts on natural ecosystems such as forests, mangroves and sandy beaches. It also damaged man-made features such as settlements, infrastructure, agricultural fields and plantations. This study proves spatial technologies are the indispensible tools for post-disaster planning and impact assessment.

Keywords: Cyclones, impact assessment, post-disaster planning, spatial technologies.

THE Earth is experiencing frequent natural disasters, and their incidence and intensity seems to be increasing in recent years, particularly cyclones and floods which often cause significant loss of life, large-scale socio-economic impacts and environmental damage[1,2]. Two cyclones originated in the Andaman Sea during October–November 2013 and devastated bay islands and the east coast of India. Disaster-causing factors of tropical cyclones including those related to strong wind, rainstorms, floods and storm surges are the prerequisites and driving forces of tropical cyclones[2]. In addition, global warming intensifies the cyclone activity leading to severe loss and damages[3,4] to public properties and the environment. Risk can be measured either as loss of life, injuries, loss of property, livelihoods or other economic activities or

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