



Occurrence and Health Risk Assessment of Phthalates in Municipal Drinking Water Supply of a Central Indian City

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Abstract

In this study, the occurrence of phthalates in the municipal water supply of Nagpur City, India, was studied for the first time. The study aimed to provide insights into the extent of phthalate contamination and identify potential sources of contamination in the city's tap water. We analyzed fifteen phthalates and the total concentration ($\sum_{15}\text{phthalates}$) ranged from 0.27 to 76.36 $\mu\text{g L}^{-1}$. Prominent phthalates identified were di-*n*-butyl phthalate (DBP), di-isobutyl phthalate (DIBP), benzyl butyl phthalate (BBP), di (2-ethylhexyl) phthalate (DEHP), di-*n*-octyl phthalate (DNOP), and di-nonyl phthalate (DNP). Out of the fifteen phthalates analyzed, DEHP showed the highest concentration in all the samples with the median concentration of 2.27 $\mu\text{g L}^{-1}$, 1.39 $\mu\text{g L}^{-1}$, 1.83 $\mu\text{g L}^{-1}$, 2.02 $\mu\text{g L}^{-1}$, respectively in Butibori, Gandhibaaag, Civil Lines, and Kalmeshwar areas of the city. In 30% of the tap water samples, DEHP was found higher than the EPA maximum contaminant level of 6 $\mu\text{g L}^{-1}$. The average daily intake (ADI) of phthalates via consumption of tap water was higher for adults (median: 0.25 $\mu\text{g kg}^{-1} \text{day}^{-1}$) compared to children (median: 0.07 $\mu\text{g kg}^{-1} \text{day}^{-1}$). The hazard index (HI) calculated for both adults and children was below the threshold level, indicating no significant health risks from chronic toxic risk. However, the maximum carcinogenic risk (CR) for adults (8.44×10^{-3}) and children (7.73×10^{-3}) was higher than the threshold level. Knowledge of the sources and distribution of phthalate contamination in municipal drinking water is crucial for effective contamination control and management strategies.

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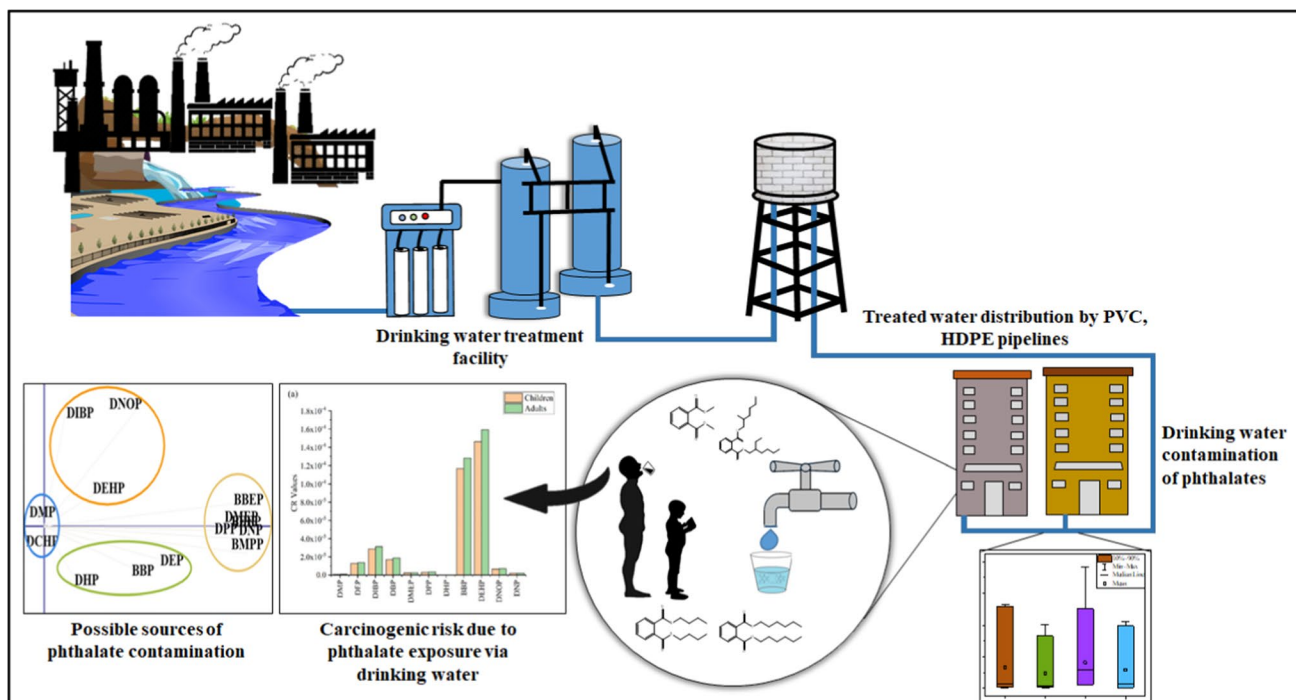
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Graphical Abstract



Access to clean and safe drinking water is essential for human life to maintain good health and well-being (Brusseau et al. 2019). Several developing countries with growing population and industrialization are facing the challenge of ensuring access to safe drinking water (Moe and Rheingans et al. 2006). Rapid urbanization, inadequate waste management infrastructure, and laxity of environmental regulations have contributed to the proliferation of contaminants in water supplies (Stoler et al. 2017). In India, tap water is the primary source of drinking water for a major portion of the population (Chakraborti et al. 2011). However, in recent years concerns have been raised regarding the presence of various contaminants in drinking water, especially organic contaminants such as per- and poly-fluoroalkyl substances (PFAS) (Teymoorian et al., 2023; Mak et al., 2009), bisphenol A (BPA) (Lim et al. 2023), polycyclic aromatic hydrocarbons (PAHs) (Liu et al. 2021), phthalates (Chenchen Wang et al. 2021a, b, c), organophosphate esters (Gbadamosi et al., 2021; Ding et al., 2015) and few inorganic contaminants such as cadmium, arsenic, and lead (Abedi Sarvestani and Aghasi et al. 2019). These chemicals are part of numerous industrial and consumer products, that eventually end up in the environmental matrices; hence, they are detected extensively in different water resources worldwide (Morin-Crini et al., 2022; Tijani et al., 2016). The presence of these chemicals in water above the threshold limit, either alone

or in the mixture, is likely to pose a risk to human health. Amongst the different contaminants reported, phthalates in drinking water have been a major concern, due to their extensive utilization in consumer products such as plastics, water bottles, plastic utensils, plumbing materials, polyvinyl chloride (PVC) pipes, food packaging, cosmetics, medical devices, and personal care products (Abtahi et al. 2019; Chen et al. 2022).

Phthalates are a class of semi-volatile organic compounds, commonly used as plasticizers in PVC, polypropylene (PP), and polyethylene (PE) products to enhance flexibility and durability (Cao et al. 2021; Pang et al. 2021). Phthalates are high-production volume chemicals that find various industrial applications such as in paints, coatings, textiles, personal care products, building materials, electronics, etc. (Núñez et al. 2022). Due to their extensive production and usage, the global consumption of phthalates alone accounted for ~ 55% plasticizers in 2020 (Ai et al. 2023). As phthalates are used as additives, they are not chemically bonded with the host materials; hence phthalates leach out into the environment during the life cycle of the articles (Ye et al. 2020). Although phthalates are biodegradable and non-persistent in nature, due to their high production and use, they are widely found in water bodies (Ai et al. 2023; Cao et al. 2022; Kong et al. 2015). Hence, phthalates are called

“everywhere chemicals” and human exposure is imminent through air, food, and water (Paluselli and Kim et al. 2020).

Studies reported that foodstuffs and drinking water are the two major sources of phthalate exposure to humans (Ji et al. 2014; Das et al. 2014). Drinking water is also used in the preparation of food, and hence monitoring of phthalates in drinking water is essential to understand its contribution to daily intake. The occurrence of phthalates in the aquatic environments viz. industrial and municipal wastewaters (Kong et al. 2015), river water (Selvaraj et al. 2015), groundwater (Edjere et al. 2016), seawater (Paluselli and Kim et al. 2020), stormwater (Cao et al. 2022), drinking water (Liu et al. 2015), bottled water (Luo et al. 2018), and sediments (Weizhen et al. 2020) has been reported extensively. These studies indicated that phthalates are widely distributed in water resources, therefore assessment of phthalates in drinking water is essential for understanding their sources, and potential health impacts to ensure safe drinking water to the general population.

Exposure to phthalates has been associated with various health effects, viz. endocrine disruption, reproductive abnormalities, and developmental issues in infants and children (Rolland et al. 2023). Phthalates have also been linked to adverse effects on human organs such as the kidney, liver, and respiratory system (Paluselli and Kim et al. 2020). A comprehensive review, incorporating results from in-silico, in-vitro, in-vivo, and epidemiological studies concluded that exposure to phthalates is associated with reproductive disorders, potentially leading to transgenerational or multigenerational effects (Ji et al. 2014). Phthalates are recognized as endocrine disruptors, especially with the production of testosterone, which is crucial for the male reproductive system (Thacharodi et al. 2023). Among the various phthalates, di(2-ethylhexyl) phthalate (DEHP) has been classified as a probable carcinogen to humans (Group 2B) and BBP is classified as Group 3 by the “International Agency for Research on Cancer (IARC)” (IARC 2013). Further, the United States Environmental Protection Agency (USEPA) classified DEHP as a probable human carcinogen (Group B2,) whereas BBP is classified as a possible human carcinogen (Class C) (USEPA 1988, 2002). However, due to lack of sufficient evidence, the USEPA classified dibutyl phthalate (DBP), diethyl phthalate (DEP), and dimethyl phthalate (DMP) under class D i.e. not classifiable as to human carcinogens (USEPA 2006).

A study conducted on 207 elderly men from Taiwan found a significant correlation between urinary levels of DEHP metabolites and the incidence of benign prostate hyperplasia and prostatic enlargement (Edjere et al. 2016). An extensive study comprising more than 1500 Australian men found a direct link between total urinary phthalates and the prevalence of Type 2 diabetes, hypertension, cardiovascular disease, and chronic low-grade inflammation (Luo

et al. 2018). It has been estimated that phthalate exposure in the European Union (EU) leads to 53,900 cases of obesity in older women annually (Legler et al. 2015). Phthalate exposure in the United States (US) adults was estimated to result in 90,761–107,283 deaths during the period 2001–2015 (Trasande et al. 2022). Therefore, phthalates have been identified as the priority pollutants of widespread occurrence by regulatory agencies such as the US Environmental Protection Agency (US EPA 2014), the European Union (EU 2013), and China (Weizhen et al. 2020).

India does not have regulatory levels of phthalates in drinking water. Though limited efforts have been made to monitor and assess the health risks associated with phthalates in India, more comprehensive studies are required to understand the current scenario and to establish priorities for human health protection measures. The existing research on phthalates in drinking water in India has produced inconsistent findings, with wide variations in occurrence and levels of phthalates across different regions and water sources. Some studies have reported low concentrations of phthalates in source water, whereas other studies have identified higher levels, particularly in areas that are affected by industrial or agricultural activities (Kumawat et al. 2022; Selvaraj et al. 2015). These findings indicate that the presence of phthalates varies from place to place depending on their use in different products, concentrations, and leaching. Therefore, it is essential to assess phthalates in different regions to understand the extent of phthalate contamination. Considering India’s large population and the challenges posed by water pollution and inadequate waste management infrastructure, ensuring safe tap water for drinking purposes is of utmost importance. Hence, the objectives of the present study were to investigate the occurrence of phthalates in municipal drinking water of Nagpur City, India, and assess the associated health risks due to phthalate exposure through drinking water. The outcome of this research is expected to support policymakers, regulators, and public health professionals in formulating appropriate guidelines values for phthalates, and take appropriate regulatory actions to minimize the contamination of drinking water resources in India.

Materials and Methods

Chemicals

A standard mixture of fifteen phthalates viz. dimethyl phthalate (DMP), diethyl phthalate (DEP), di-iso-butyl phthalate (DIBP), dibutyl phthalate (DBP), di-(2-methoxyethyl) phthalate (DMEP), bis (4-methyl-2-pentyl) phthalate (BMPP), bis (2-ethoxyethyl) phthalate (BEEP), dipentyl phthalate (DPP), di-hexyl phthalate (DHP), benzyl butyl phthalate (BBP), bis (2-*n*-butoxyethyl) phthalate (BBEP), di-cyclohexyl phthalate

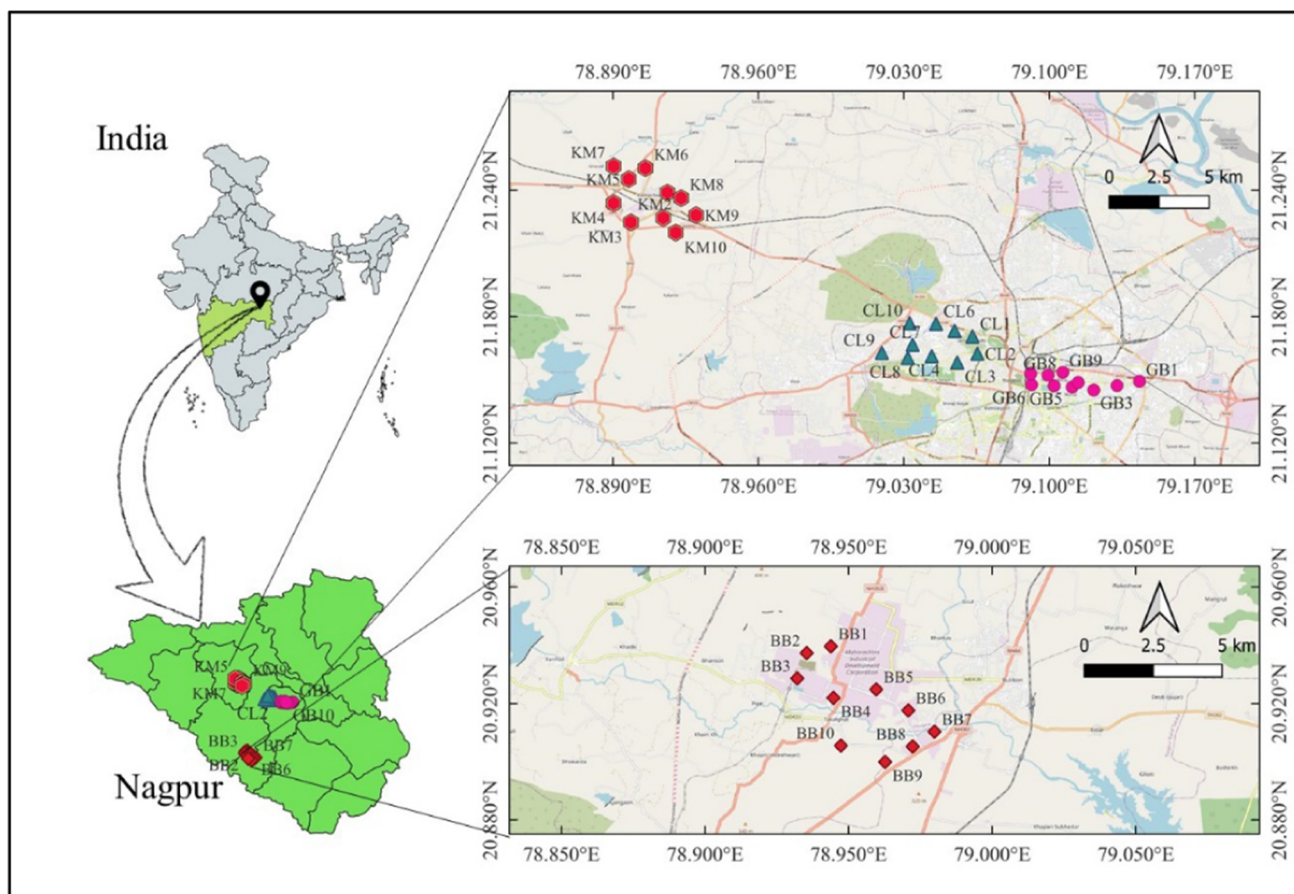


Fig. 1 The study area showing sampling sites in Nagpur City, India

(DCHP), di-*n*-octyl phthalate (DNOP), di-nonyl phthalate (DNP), and di(2-ethylhexyl) phthalate (DEHP) at a concentration of $100 \mu\text{g mL}^{-1}$ was purchased from AccuStandard Inc. (USA). Nine deuterated phthalates viz. DMP- d_4 , DEP- d_4 , DPP- d_4 , DIBP- d_4 , DBP- d_4 , DCHP- d_4 , DEHP- d_4 , DNOP- d_4 , and DNP- d_4 were also purchased from AccuStandard Inc. (USA). Pesticide residue grade dichloromethane (DCM), hexane, and acetone were purchased from Riedel-de Haën (Steinheim, Germany). Anhydrous sodium sulphate was obtained from Sigma Aldrich Chemicals Private Limited (Bangalore, India). Glass fiber filter paper ($0.1 \mu\text{m}$) was purchased from Pall Corporation (USA).

Study Area

The study area is Nagpur city, located in Central India. Within the Nagpur city, four areas representing; industrial zone: Butibori (BB), residential-commercial zone: Civil Lines (CL), rural: Kalmeshwar (KM), and a vibrant commercial area: Gandhibagh (GB) have been selected for sampling (Fig. 1). The selection of these four areas with distinct characteristics was aimed at providing insights on the

sources of phthalate contamination in different areas of the city. The three areas viz. GB, CL, and KM receive treated municipal water supply from Kanhan and Pench rivers and Gorewada lake, whereas BB receives municipal water from Wadgaon dam. The source water is treated by conventional treatment processes, and activated carbon filter (ACF), chlorinated, and distributed to different areas through pipelines. A total of 40 tap water samples were collected directly from the tap of houses located in BB, CL, GB, and KM areas (10 samples from each area) in 2.5 L amber-colored glass bottles. The distance between the two sampling points within the four areas varied between about 500–1000 m. All the samples were collected on the same day to minimize the artefacts of inter-day variations.

Extraction of Phthalates from Water Samples

The samples were collected by following USEPA 506 method for proper handling and minimizing potential sources of contamination (USEPA 1984; Kawahara 1995). Isotopically labeled phthalate standards ($10 \mu\text{g}$ each) were used as surrogate recovery standards and the percent

recovery obtained was in the range of 85–97% (Table ST1). Liquid–liquid extraction was followed for the extraction of phthalates from water samples. In brief, 1000 mL of water sample was taken in a 1 L separating funnel and isotopically labelled standards were added. The contents were shaken vigorously with 60 mL of dichloromethane (DCM) for 10 min and allowed to stand for 2 min for the separation of layers. The extraction was repeated thrice with fresh 60 mL portions of DCM to obtain maximum recovery of the analytes. The extracts were combined and passed through anhydrous sodium sulfate for moisture removal. The combined extracts were concentrated in a rotary evaporator (Heidolph Instruments GmbH & Co., Germany) and the solvent was exchanged to 1 mL of hexane (USEPA 1984; Otero et al. 2015).

Instrumentation

The targeted fifteen phthalates were analyzed by gas chromatography–tandem mass spectrometer (GC–MS/MS, Agilent, 7000DGC/TQ) in electron ionization (EI) mode (Agilent Technologies, Inc. USA). The column used was HP-5MS ultra-inert capillary column (30 m × 0.25 mm × 0.25 μm) (Agilent J&W GC Columns) and helium was used as the carrier gas at a flow rate of 1.0 mL min⁻¹. The split-less injector temperature was set at 280 °C. The temperature program of the oven was as follows: 100 °C, hold for 0.5 min, ramp 15 °C min⁻¹ to 235 °C, hold for 1 min, ramp 15 °C min⁻¹ to 265 °C, hold for 3 min, and ramp 15 °C min⁻¹ to 310 °C, hold for 1 min. The ion source and interface temperatures were set at 280 °C, and 300 °C, respectively. The solvent delay used for the MS was 3 min. The MS was operated in multiple reaction monitoring (MRM) mode with a focus on three characteristic ions for each analyte. More information on MS regarding the mass of the ions monitored for each phthalate, retention time (RT), limit of detection (LOD), and limit of quantitation limit (LOQ) are provided in Table 1.

Quality Control and Quality Assurance (QA/QC)

Strict quality control measures were followed throughout the sampling, sample preparation, and instrumental analysis. All glassware were cleaned by ultrasonication, washed with ultrapure water, solvent rinsed, oven-dried at 100 °C for 2 h, and cooled before use. No plastic-wares were used during the analysis to avoid any potential contamination. The analysis of procedural blanks and duplicates were also conducted to assess potential contamination, precision, and accuracy. The repeatability of analytical results calculated as percent relative standard deviation (%RSD) was in the range of 1.12–12.56%. The LOD and LOQ were defined as the concentration corresponding to the signal-to-noise ratio of 3 and 10, respectively. The LODs of fifteen phthalates were

in the range of 0.001–0.0066 μg L⁻¹ (Table 1). The analysis of each sample was repeated 3×, and the mean concentration was reported. The method was validated by spiking at three levels (5, 10, and 20 μg L⁻¹) and the percentage recovery of fifteen phthalates was in the range of 72–110% (Table ST 1).

Health Risk Assessment

Human exposure to phthalates via drinking water commonly occurs through oral ingestion, dermal contact, and inhalation of water vapors during bathing. As ingestion is the major route of exposure, the other two routes of exposure are negligible. Accordingly, the intake dose of phthalates via ingestion of drinking water was calculated using the Eq. (1):

$$ADI_{\text{oral}} = \frac{C_w \times IR_w \times EF \times ED}{BW \times AT} \quad (1)$$

where ADI_{oral} is the average daily intake of phthalates via oral ingestion of drinking water (μg kg⁻¹ day⁻¹), C_w is the concentration of phthalate in drinking water (μg L⁻¹), IR_w is the ingestion rate of drinking water (1.277 L day⁻¹ for adults of age 30–40 years and 0.455 L day⁻¹ for children of age 6–11 years) (US EPA 2019), EF is the exposure frequency (days/year) (365 days/year), ED is the lifetime exposure duration (49 and 5 years for adults and children, respectively), AT is the average time of exposure (EF × ED days) and BW is the body weight (kg) (Table ST2).

The chronic toxic risk (CTR) to adults and children associated with phthalates exposure through drinking water was estimated using the hazard quotient (HQ) (Eq. 2) and the cumulative CTR was estimated by hazard index (HI), which is the sum of the HQ of individual phthalates (Eq. 3). The reference doses (RfDs) of phthalates were used as the benchmark values of toxicity (Table ST3). The calculation of the cumulative risks was based on the assumption that all the phthalates show similar modes of toxic action (Liu et al. 2015; Xie et al. 2022).

$$HQ = \frac{ADI}{RfD} \quad (2)$$

The assessment of carcinogenic risk (CR) was carried out by using the linear low-dose carcinogenic risk equation:

$$\text{Risk} = ADI_{\text{oral}} \times SF \quad (3)$$

Risk is the probability of an individual developing cancer (unit less) and SF is the slope factor (μg/kg-bw/day)⁻¹. For CR calculation, SF for oral ingestion (μg/kg-bw/day)⁻¹ was used (Table ST3). The estimated CR levels < 1 × 10⁻⁶ indicate negligible risk, values between 1 × 10⁻⁴ and 1 × 10⁻⁶ indicate acceptable risk, and values > 1 × 10⁻⁴ indicate high risk (Xie et al. 2022).

Data Analysis

Statistical analysis was performed using two softwares viz. IBM statistical package for Social Sciences (SPSS version 26) and Microsoft Excel. The data obtained from the study were subjected to descriptive statistical analysis. The normality of the data distribution was assessed using the Shapiro–Wilk test, which indicated that the data does not follow a normal distribution. The Mann–Whitney U test and Kruskal–Wallis H test were conducted to compare data among two groups and multiple groups. The statistical significance level was set at $p < 0.05$, indicating the results are statistically significant.

Results and Discussion

Concentration of Phthalates in Tap Water

The concentrations of fifteen phthalates in tap water collected from the four areas of Nagpur city were presented in Fig. 2 and Table ST4. The total concentration of phthalates ($\sum_{15}\text{phthalates}$) ranged from 0.27 to 76.36 $\mu\text{g L}^{-1}$ with the median concentration of 3.92 $\mu\text{g L}^{-1}$. The area-wise distribution of $\sum_{15}\text{phthalates}$ concentration was as follows: 0.59 to 53.08 $\mu\text{g L}^{-1}$ in BB, 0.31 to 40.04 $\mu\text{g L}^{-1}$ in GB, 1.91 to 76.36 $\mu\text{g L}^{-1}$ in CL, and 0.27 to 42.24 $\mu\text{g L}^{-1}$ in KM (Fig. 2). Statistical analysis of the data showed that $\sum_{15}\text{phthalates}$ concentration did not differ significantly ($p = 0.33$) in BB, CL, GB, and KM areas.

Phthalates concentration in tap water showed wide variation with detection frequencies (DF) ranging from 40 to 100% in BB, 10 to 100% in GB, 0 to 100% in KM and 40 to 100% in CL, respectively. In the BB area, DEHP had the highest mean percentage concentration of 90.52%, while other phthalates showed comparatively lower mean percentages in the range of 0.01 to 2.57%. The mean percentage concentrations of DEHP in GB, CL, and KM areas were 81.53%, 58.64%, and 92.88%, respectively. The area-wise distribution (box-plot) of twelve phthalates was presented in Fig. 3. The distribution pattern of phthalates in tap water samples of BB area followed the order: DEHP > DIBP > DBP > DMEP > DEP > BEEP > BBP > BBEP > DNOP > BMPP > DMP > DPP > DNP > DHP \approx DCHP, indicating the predominance of DEHP. In GB, next to DEHP (81.53%), the mean percentage concentration followed the order: DNOP (6.42%), DBP (3.58%), BEEP (2.28%), DMEP (1.86%), and DIBP (1.26%). In CL, the major phthalates after DEHP were DIBP (19.93%), DNOP (18.18%), and DBP (0.97%), while, in KM, the mean percentage concentration of DIBP, DBP, and DEP were 2.08%, 1.48%, and 1.25%, respectively. The predominance of DEHP, DNOP, DIBP, and DBP and their distribution pattern in the tap water of four areas indicated that phthalates could have originated partly from the water distribution pipelines as these phthalates are used as plasticizers in PVC (Henkel et al. 2022; Jeon et al. 2016; Ye et al. 2020; Mohammadi et al. 2022).

Comparison with Other Published Results

To perceive the global scenario, we compared the concentration of phthalates in tap water of Nagpur city with other

Table 1 Gas chromatography–mass spectrometer method parameters

PAEs	Precursor ion (m/z)	Product ion (m/z)	Collision energy (V)	LOD ($\mu\text{g L}^{-1}$)	LOQ ($\mu\text{g L}^{-1}$)
DMP	163	77	5	0.0014	0.0042
DEP	149	93	7	0.0019	0.0057
DIBP	149	93, 121	13	0.0011	0.0033
DBP	149	93, 121	13	0.0011	0.0033
DMEP	149	93, 121	16	0.0052	0.0156
BMPP	149	93, 121	16	0.0065	0.0195
BEEP	149	93, 121	13	0.0034	0.0102
DPP	149	93, 121	13	0.0016	0.0048
DHP	149	65, 93	13	0.0014	0.0042
BBP	149	93, 121	13	0.0058	0.0174
DBEP	149	93, 121	13	0.0066	0.0198
DCHP	167	149	5	0.0014	0.0042
DEHP	149	65	5	0.0012	0.0036
DNOP	149	93, 121	13	0.0016	0.0048
DNP	149	93, 121	13	0.0013	0.0039

LOD, limit of detection; LOQ, limit of quantification

published results (Table ST6). It was observed that the concentration was significantly higher than that reported from Tehran, Iran (\sum_6 phthalates ranged from 0.45 to 1.08 $\mu\text{g L}^{-1}$) (Abtahi et al. 2019); Tianjin, China (\sum_3 phthalates ranged from 0.38 to 1.78 $\mu\text{g L}^{-1}$) (Chenchen. Wang et al. 2021a, b, c); Hanoi, Vietnam (\sum_{10} phthalates ranged from ND to 14.50 $\mu\text{g L}^{-1}$) (Le et al. 2021); South Florida, USA (\sum_6 phthalates ranged from 0.07 to 3.88 $\mu\text{g L}^{-1}$) (Cui et al. 2022); Madrid, Spain (\sum_6 phthalates ranged from <LOD to 0.91 $\mu\text{g L}^{-1}$) (Domínguez-Morueco et al. 2014); and France (\sum_5 phthalates ranged from 0.15 to 1.32 $\mu\text{g L}^{-1}$) (Bach et al. 2020). However, a recent study from Jabalpur, India reported very high concentration of phthalates in municipal drinking water samples (\sum_6 phthalates ranged from 1.86 to 1438.20 $\mu\text{g L}^{-1}$) (Kumawat et al. 2022). To the best of our knowledge, this is the first-ever study to report the highest concentration of \sum_6 phthalates in drinking water. The \sum_6 phthalates concentration reported by Kumawat et al. (2022) was about 10–20-fold higher than our results, however, the probable sources of such higher concentrations were not discussed by the authors. Barring the study of Kumawat et al. (2022), all other studies reported lower DEHP concentrations than the present study. The maximum DEHP concentration was found at the sampling locations BB4, BB10, GB4, GB5, GB6, CL3, CL9, KM2, KM3, and KM5 with concentrations in the range of 15.77 to 66.78 $\mu\text{g L}^{-1}$.

A nationwide survey of six phthalates in drinking water from waterworks across China reported the median concentration of phthalates in the range of 0.001 to 0.18 $\mu\text{g L}^{-1}$ with DBP and DEHP as the dominant phthalates (Liu et al. 2015). In the present study, DEP was the second most contributing phthalate with higher concentration found at the sampling points BB2, BB3, BB7, GB4, and KM2. The maximum concentration of DEP (0.35 $\mu\text{g L}^{-1}$) detected in the present study was higher than the previously reported studies by Wang et al. (2022) (0.098 $\mu\text{g L}^{-1}$); Jian et al. (2022) (0.028 $\mu\text{g L}^{-1}$); Liu et al. (2014) (0.23 $\mu\text{g L}^{-1}$); and Yang et al. (2022) (0.0068 $\mu\text{g L}^{-1}$). DIBP was the third most contributing phthalate in tap water with a median concentration ranging from ND to 0.12 $\mu\text{g L}^{-1}$. The DIBP concentration reported in the present study was similar to the concentration reported by Yang et al. (2022) (ND to 0.11 $\mu\text{g L}^{-1}$), and Santana et al. (2014) (ND to 0.17 $\mu\text{g L}^{-1}$). Comparatively higher concentration was reported by Le et al. (2021) (0.03 to 1.39 $\mu\text{g L}^{-1}$) and Wang et al. (2022) (ND to 3.31 $\mu\text{g L}^{-1}$). Other phthalates viz. DBP, BEEP, DMEP, BBEP, BMPP, and DNOP were detected with the median concentration ranging from 0.01 to 0.08 $\mu\text{g L}^{-1}$. The presence of these phthalates in tap water samples indicates their use in various products albeit to a lesser extent compared to DEHP. The distribution pattern of fifteen phthalates in the present study

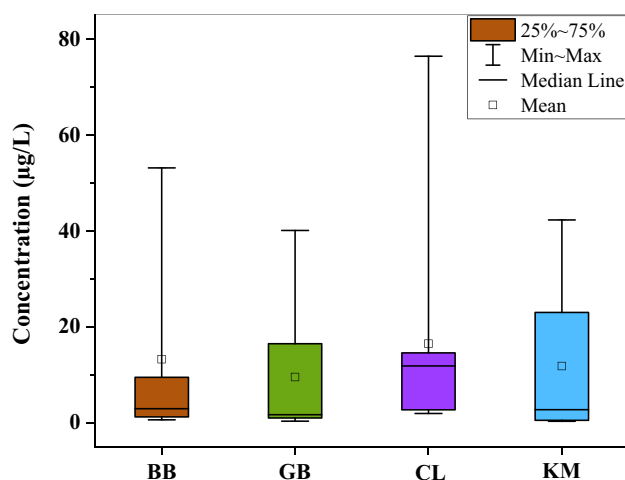


Fig. 2 Box-plot of \sum_{15} phthalates in tap water samples collected from Butibori (BB), Gandhibagh (GB), Civil Lines (CL), and Kalmeshwar (KM) areas of Nagpur city

more or less followed the global production and use pattern of phthalates in various applications (Nagorka et al. 2022).

Comparison with Water Quality Guidelines

Drinking water quality is assessed based on certain physical, chemical, and biological parameters. Indian standard for drinking water IS:10500 (2012) provides guideline values of these parameters, but phthalates and other emerging contaminants such as organophosphate esters, bisphenol-A, per- and poly-fluoroalkyl substances (PFAS), etc. have not been included. To minimize the risk of phthalate contamination in drinking water, several regulatory agencies have established guidelines. The World Health Organization has set 8.0 $\mu\text{g L}^{-1}$ as the acceptable limit for DEHP in drinking water (WHO 2003). The European Union (European Union 2013) and the United States Environmental Protection Agency (US EPA 2014) recognized six phthalates including DEHP as the priority pollutants. The USEPA maximum contaminant level (MCL) for DEHP has been fixed at 6 $\mu\text{g L}^{-1}$. In China, the acceptable limit for DBP and DEHP in source water has been set at 3.0 and 8.0 $\mu\text{g L}^{-1}$, respectively (Weizhen et al. 2020). In Japan, the respective MCL values for DEHP, DBP, and BBP are 100 $\mu\text{g L}^{-1}$, 200 $\mu\text{g L}^{-1}$, and 500 $\mu\text{g L}^{-1}$ (Wakayama 2004).

In the present study, 12 tap water samples (30%) exceeded the USEPA MCL limit of 6 $\mu\text{g L}^{-1}$. The average concentrations of DEHP in BB, GB, CL, and KM areas of Nagpur city were 11.95 $\mu\text{g L}^{-1}$, 7.74 $\mu\text{g L}^{-1}$, 9.64 $\mu\text{g L}^{-1}$, and 10.94 $\mu\text{g L}^{-1}$, respectively. These values were higher than the limit proposed by USEPA and WHO. In addition, the USEPA proposed ambient water quality criteria (AWQC) for priority phthalates as 2000 $\mu\text{g L}^{-1}$ for DMP; 600 $\mu\text{g L}^{-1}$

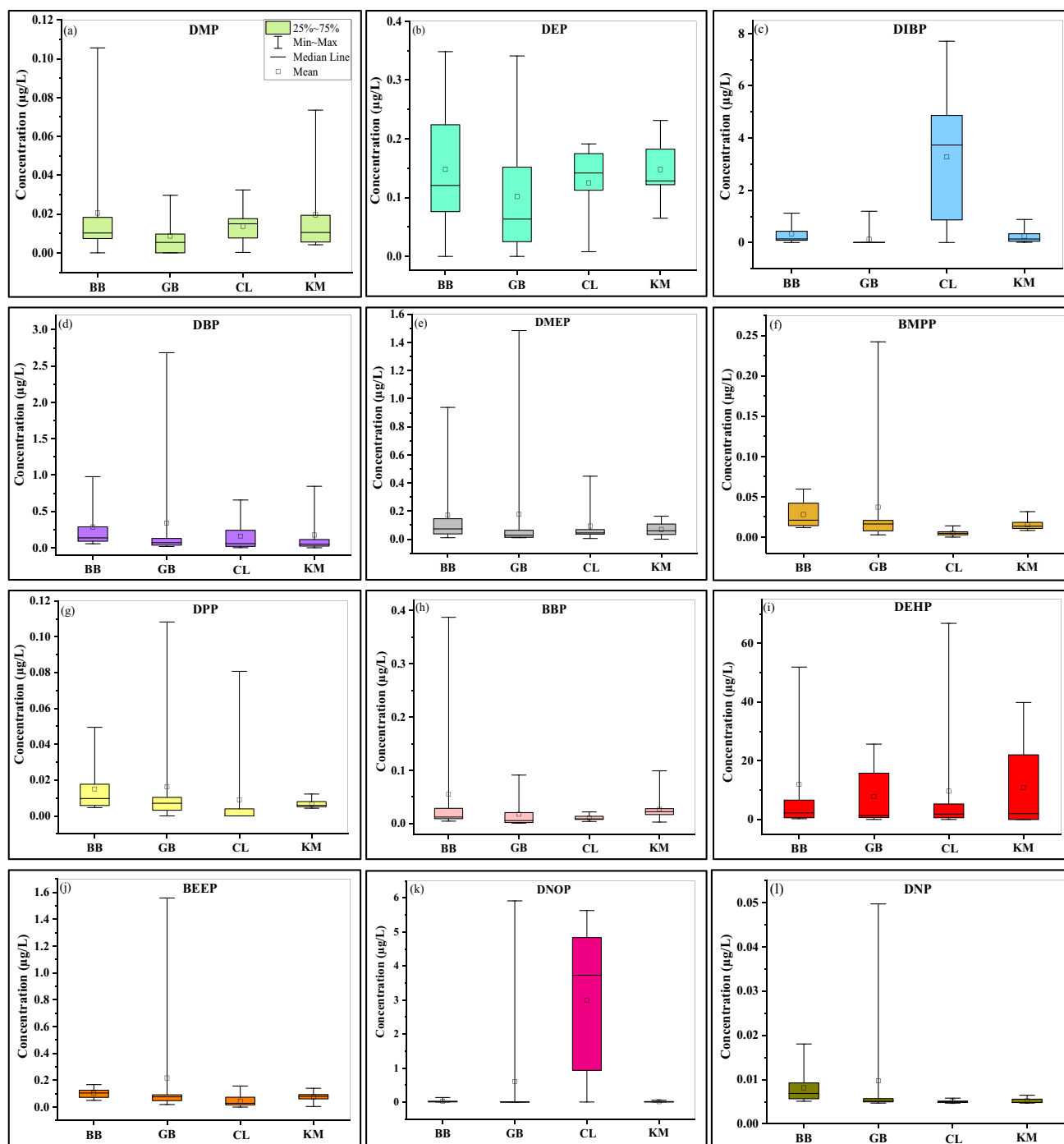


Fig. 3 Minimum, maximum, mean, and median concentration of **a** DMP, **b** DEP, **c** DIBP, **d** DBP, **e** DMEP, **f** BMPP, **g** DPP, **h** BBP, **i** DEHP, **j** BEEP, **k** DNOP, **l** DNP in tap water samples. (BB, Butibori; GB, Gandhibagh; CL, Civil Lines; KM, Kalmeshwar)

for DEP; $20 \mu\text{g L}^{-1}$ for DBP; $0.10 \mu\text{g L}^{-1}$ for BBP; and $0.32 \mu\text{g L}^{-1}$ for DEHP (USEPA 2015). The percentage of samples that exceeded the AWQC for BBP and DEHP was 5 to 77.50%. All the other phthalates were lower than the AWQC recommended by the (USEPA 2015).

Phthalates Contamination Profile

We analyzed the results of fifteen phthalates in the tap water to explore whether the phthalate content and profile were influenced by the geographical location and/or source water. Figure 4 provides the percentage contribution of fifteen phthalates from four areas of Nagpur city. The

median concentration of \sum_{15} phthalates was in the order: CL ($11.80 \mu\text{g L}^{-1}$) > KM ($2.69 \mu\text{g L}^{-1}$) > BB ($2.88 \mu\text{g L}^{-1}$) > GB ($1.65 \mu\text{g L}^{-1}$) (Table ST5). Among the fifteen phthalates, the median concentration of DEHP ranged between 1.39 to $2.27 \mu\text{g L}^{-1}$ and contributed 58.64% to 92.88% of \sum_{15} phthalates in the four areas. In BB, besides DEHP (90.51%), phthalate compounds DIBP (2.57%), DBP (2.15%), DMEP (1.71%), and DEP (1.48%) contributed substantially to the \sum_{15} phthalates. However, in GB, DNOP (6.01%), DBP (3.40%), BEEP (2.16%), and DMEP (1.77%) were the major contributing phthalates apart from DEHP. In CL, the contribution of DIBP and DNOP was 19.92% and 18.18%, respectively. This was followed by DBP (0.97%), DEP (0.76%), and DMEP (0.57%); however, in KM, DIBP (2.08%), DBP (1.48%), and DEP (1.25%) were the major contributing phthalates. Among all the phthalates, the sum of DEHP, DBP, and DIBP showed the maximum contribution to the total phthalate levels and accounted for 95.23%, 86.37%, 79.53%, and 96.44% in BB, GB, CL, and KM, respectively. This distribution pattern indicates the origin of phthalates from both PVC pipelines (DEHP, DIBP, DBP) as well as from source water (DMEP and DEP). Further, even though CL, GB, and KM areas receive water from the same source, only the tap water of the CL area showed a substantial contribution of DIBP (19.92%) and DNOP (18.18%) to \sum_{15} phthalates. This could be due to the recent replacement of several water supply pipelines in the area, which might contain the HMW phthalates DIBP and DNOP. However, this hypothesis requires further investigation.

The phthalate profile pattern *i.e.* dominance of DEHP, DBP, and DIBP observed in the municipal waters of this study, was similar to previous studies conducted on other environmental media *viz.* ambient air (Kashyap and Agarwal 2018); river water (Selvaraj et al. 2015); soil (Yue et al. 2022); and sediment (Neves et al. 2023). The similarity of phthalate profiles in several environmental media indicates that DEHP, DBP, and DIBP are the majorly used phthalates as plasticizers and additives in various applications including PVC pipes (Zhang et al. 2023). DEHP and DIBP have dominated the phthalate market constituting about 50% of the plasticizer market share, and DIBP has been applied as a replacement of DBP in recent years (Nagorka et al. 2022). While DMEP concentration was initially found in drinking water samples from Shanghai (Wu et al. 2013), its presence was not reported so far in surface water samples from India (Kumawat et al. 2022). As DMEP is mainly used in PCPs (Bao et al. 2015), its occurrence in most drinking water samples (DF: 97.50%) of this study indicates the contamination of source water.

Principal Component Analysis

Principal Component Analysis (PCA) was carried out to identify the sources of phthalates in tap water. PCA extracted five principal components (PC) with eigenvalues > 1, indicating their contribution to the observed variance of phthalates distribution in tap water. PC1 (45.8%), PC2 (12.8%), PC3 (11.1%), PC4 (7.4%), and PC5 (6.8%) explained 74% of the total variance. The loading and the score plots of all the samples were given in Fig. 5a, b, respectively. Figure 5a indicates that PC1 was the major source of phthalates contributed by DNP, DMEP, BMPP, BEEP, DPP, and BBEP, and moderate source for plasticizer phthalates DEHP, DIBP, DINP, and DNOP. PC1 also had moderate contributions from DHP, BBP, and DEP. Thus, PC1 is the indicator of “source water contamination” originating from non-point sources of contamination. Whereas, PC2 has strong loadings of plasticizer phthalates *viz.* DEHP, DIBP, DINP, and DNOP, which originate mainly from the water supply pipelines.

In the PCA loading plot, the fifteen phthalates clustered into four groups (Fig. 5a). The first group consists of DBP, DMEP, BMPP, BEEP, DPP, BBEP, and DNP, accounted for 36.2%, 35.3%, 36.4%, 36.2%, 32.5%, 34.5%, and 37%, respectively to the total proportion and showed a positive correlation in PC1. The second group consists of high molecular weight phthalates such as DEHP, DNOP, and DIBP which originated from water distribution PVC pipelines and source water. DNOP is used as an additive in general-purpose plasticizers (GPP) along with performance plasticizers such as DIBP, DBP, etc. (Eastman Chemical Company, 2023). The predominant phthalate DEHP, however, did not show a strong correlation with PC2, rather it accounted for only 17.5% of the total proportion possibly due to its origin from multiple sources. On the other hand, the third group comprising DEP, BBP, and DHP accounted for 21.3%, 16.4%, and 6.1%, respectively in PC1. DEP and DHP are commonly used as additives in plasticizers, cosmetics, and personal care products (U.S. Food and Cosmetics. 2022; Murat et al. 2019). In contrast to all phthalates, DMP and DCHP were distinctly isolated and showed no correlation with PC1 and PC2. This indicates that the source and use pattern of DMP and DCHP might be different from other phthalates or the lack of correlations with other phthalates.

The score plot (Fig. 5b) of PCA showed that the first component of PCA was strongly loaded with the tap water samples collected from CL, while samples from BB, GB, and KM dominated the second component. Moreover, specific samples *viz.* GB4 (located in the positive direction of PC1) and BB2, BB7, and BB9 (located in the negative direction of PC2), showed higher levels of certain phthalates, including DBP, DIBP, DMEP, BEEP, DEHP, and DNOP. These

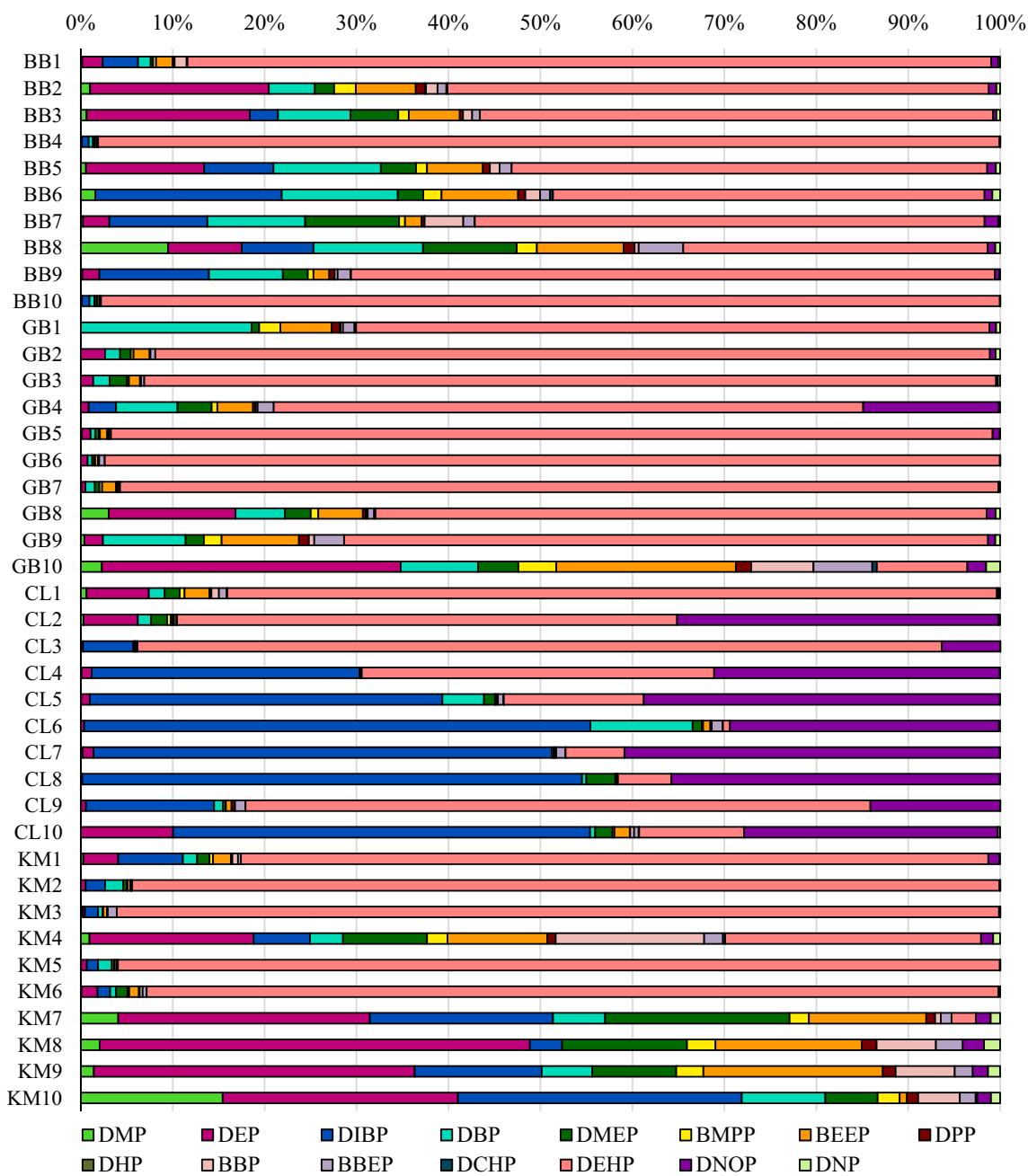


Fig. 4 Phthalates profile in tap water samples collected from Butibori (BB), Gandhibagh (GB), Civil Lines (CL), and Kalmeshwar (KM)

findings indicate that phthalates in tap water in these areas originated from multiple sources.

Probable Sources of Phthalates in Tap Water

The presence of diverse phthalate compositions in tap water suggests the influence of different contamination sources. Based on the phthalate profile and the results of PCA, two major sources contributed to phthalates in tap water viz. source water contamination resulting from

non-point sources, and leaching from pipelines. Some of the rivers and streams that originate in the catchment area receive untreated sewage and contribute to the source water (MPCB 2019). Untreated domestic sewage has been identified as the major contributor to the occurrence of elevated levels of phthalates and other organic contaminants in Indian rivers (Selvaraj et al. 2015; Mishra et al. 2023). Further, littering of single-use plastics, and open applications of phthalates containing products in building materials, paints, coatings, etc. contribute to urban

rainfall runoff that eventually ends up in surface water bodies. Thus, the source contamination factor identified as PC1 is the major sources of phthalates such as DMP, DEP, DBP, DMEP, BMPP, BEEP, DPP, BBP, and BBEP. PC1 has also contributed plasticizer phthalates DEHP, DIBP, DINP, and DNOP.

The second source identified by PC2 was the leaching from PVC pipelines which was the major contributor of DEHP, DIBP, DINP, and DNOP to the tap water. Several factors including pH, aging of pipes, contact time, water composition, temperature, biofilm etc. influence the leaching of phthalates and other additives from plastics (Abtahi et al. 2019; Wang et al. 2021a, b, c; Zhang et al. 2023; Liu et al. 2016; Mohammadi et al. 2022; Wang et al. 2021a, b, c; Zhang et al. 2023). However, we have not investigated these factors as it was beyond the scope of our study. Most of municipal water treatment plants (WTP) in India use ACF in addition to conventional treatment techniques (Sharma and Bhattacharya et al. 2017). Previous studies indicate that conventional water treatment, has removal efficiencies in the range of 15–65% for phthalates. For e.g. Choi et al. (2006) reported removal efficiency of 46%, 49%, and 53% for DEP, DBP, and DEHP, respectively. Another study reported 93% and 57% removal efficiency for DMP and DBP, respectively with overall removal efficiency of 62% for all phthalates using ACF (Liu et al. 2011). Kong et al. (2017) reported 15%, 24.3%, 59%, and 64.5% removal efficiency for DMP, DIBP, DEHP, and DBP, respectively in finished water and found that a substantial fraction of DEHP (mean: 150 ng/l) eludes water treatment process and make its presence in tap water (Kong et al. 2017). Although we have not investigated the phthalate removal efficiency, based on previous findings, we believe that incomplete removal in WTPs would have

also contributed to the prevalence of DEHP in tap water from Nagpur city.

Exposure of Phthalates in Humans

The exposure dose of phthalates to humans was estimated using factors such as the average body weight of the Indian population, the volume of water consumption per day, and the concentration of phthalates in tap water (Table ST2). The ADI values of fifteen phthalates to children through the consumption of tap water were 5.19×10^{-2} , 2.96×10^{-2} , 2.12×10^{-1} , and $4.84 \times 10^{-2} \mu\text{g kg}^{-1} \text{day}^{-1}$ in BB, GB, CL, and KM, respectively. Similarly, the ADI values for adults were 5.67×10^{-2} , 3.24×10^{-2} , 2.32×10^{-1} , and $5.29 \times 10^{-2} \mu\text{g kg}^{-1} \text{day}^{-1}$ in BB, GB, CL, and KM, respectively. No significant difference was observed on phthalate exposure between children and adults ($p = 0.39$). Among the fifteen phthalates, DEHP showed the highest mean exposure value of 1.35×10^{-1} and $1.48 \times 10^{-1} \mu\text{g kg}^{-1} \text{day}^{-1}$ for children and adults, respectively. This was followed by DIBP (children: $7.20 \times 10^{-2} \mu\text{g kg}^{-1} \text{day}^{-1}$ and adults: 7.86×10^{-2}), DNOP (children: $6.76 \times 10^{-2} \mu\text{g kg}^{-1} \text{day}^{-1}$ and adults: $7.38 \times 10^{-2} \mu\text{g kg}^{-1} \text{day}^{-1}$), DEP (children: $8.17 \times 10^{-2} \mu\text{g kg}^{-1} \text{day}^{-1}$ and adults: $8.92 \times 10^{-3} \mu\text{g kg}^{-1} \text{day}^{-1}$) and DBP (children: $5.58 \times 10^{-3} \mu\text{g kg}^{-1} \text{day}^{-1}$ and adults: $6.09 \times 10^{-3} \mu\text{g kg}^{-1} \text{day}^{-1}$). The estimated exposure of phthalates through the consumption of tap water was lower than the oral RfD (Table ST7). However, it was higher than that reported in Vietnam i.e. $3.94 \times 10^{-2} \mu\text{g kg}^{-1} \text{day}^{-1}$ (for DEHP) to $5.69 \times 10^{-2} \mu\text{g kg}^{-1} \text{day}^{-1}$ (for DBP) for the adult population (Le et al. 2021) and China i.e. $6.71 \times 10^{-6} \mu\text{g kg}^{-1} \text{day}^{-1}$ (for DBP) to $1.16 \times 10^{-5} \mu\text{g kg}^{-1} \text{day}^{-1}$ (for DEHP) (Zhang et al. 2023) via drinking water.

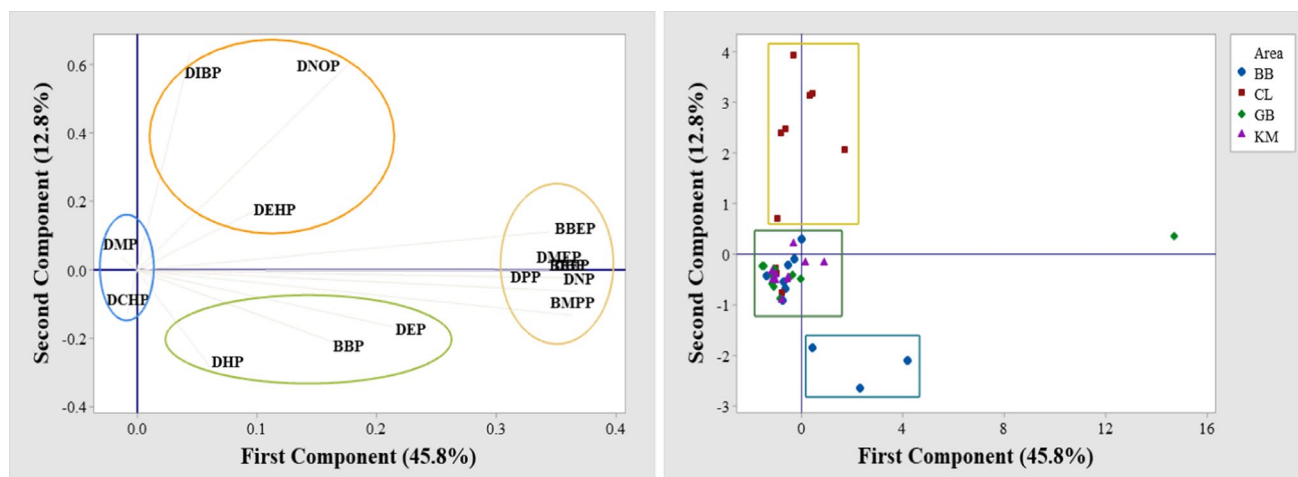


Fig. 5 Principal component analysis of phthalates in tap water showing **a** loading and **b** score plots (BB, Butibori; GB, Gandhibagh; CL, Civil Lines; KM, Kalmeshwar)

Table 2 Median hazard quotient and hazard index for children and adults

	Butibori (BB)		Civil Lines (CL)		Gandhibagh (GB)		Kalmeshwar (KM)	
	Hazard Quotient (HQ)							
	Children	Adults	Children	Adults	Children	Adults	Children	Adults
DMP	1.84×10^{-6}	2.01×10^{-6}	2.69×10^{-6}	2.94×10^{-6}	9.69×10^{-7}	1.06×10^{-6}	1.89×10^{-6}	2.06×10^{-6}
DEP	2.71×10^{-6}	2.96×10^{-6}	3.19×10^{-6}	3.48×10^{-6}	1.43×10^{-6}	1.56×10^{-6}	2.88×10^{-6}	3.15×10^{-6}
DIBP	8.54×10^{-5}	9.33×10^{-5}	2.34×10^{-3}	2.56×10^{-3}	0.00	0.00	8.39×10^{-5}	9.16×10^{-5}
DBP	2.44×10^{-5}	2.66×10^{-5}	1.01×10^{-5}	1.10×10^{-5}	1.24×10^{-5}	1.36×10^{-5}	8.89×10^{-6}	9.71×10^{-6}
DMEP	3.03×10^{-5}	3.31×10^{-5}	1.86×10^{-5}	2.03×10^{-5}	1.17×10^{-5}	1.27×10^{-5}	2.48×10^{-5}	2.71×10^{-5}
DPP	2.62×10^{-6}	2.87×10^{-6}	0.00	0.00	1.90×10^{-6}	2.08×10^{-6}	1.60×10^{-6}	1.75×10^{-6}
DHP	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
BBP	2.22×10^{-6}	2.42×10^{-6}	1.60×10^{-6}	1.75×10^{-6}	1.01×10^{-6}	1.11×10^{-6}	4.04×10^{-6}	4.42×10^{-6}
DEHP	2.04×10^{-3}	2.23×10^{-3}	1.65×10^{-3}	1.80×10^{-3}	1.25×10^{-3}	1.37×10^{-3}	1.82×10^{-3}	1.99×10^{-3}
DNOP	5.80×10^{-6}	6.33×10^{-6}	1.68×10^{-3}	1.83×10^{-3}	3.16×10^{-6}	3.46×10^{-6}	3.94×10^{-6}	4.31×10^{-6}
DNP	3.34×10^{-6}	3.65×10^{-6}	2.42×10^{-6}	2.64×10^{-6}	2.48×10^{-6}	2.71×10^{-6}	2.35×10^{-6}	2.56×10^{-6}
HI	2.14×10^{-3}	2.34×10^{-3}	6.74×10^{-3}	7.36×10^{-3}	1.28×10^{-3}	1.39×10^{-3}	2.01×10^{-3}	2.19×10^{-3}

HI, hazard index

Ingestion, inhalation, and dermal contact are the three routes of exposure to phthalates in humans. Therefore, we compared the exposure through drinking water of our study with other routes of exposure to find out the relative contribution to RfD. For e.g. the exposure of \sum_6 phthalates reported in China through food intake was 0.0036 and $0.0024 \mu\text{g kg}^{-1} \text{day}^{-1}$ for toddlers and adults, respectively which is about three and one orders magnitude lower than the present study (Wang et al. 2021a, b, c). Further, the mean \sum_{10} phthalates exposure via air in Vietnam was 0.42 and $0.21 \mu\text{g kg}^{-1} \text{day}^{-1}$ for children and adults, respectively, which was about an order magnitude lower for adults, and higher for children, respectively than the present study (Tran et al. 2017).

Human Health Risk Assessment

The CTR (HQ and HI) and CR due to phthalate exposure via drinking water were calculated and the results were given in Table 2. HQ results indicated that exposure from DEHP was highest followed by DIBP, DMEP, and DBP. The HQ values of DEHP in 55% of the sampling locations were comparatively higher than the other phthalates (HQ range: 1.04×10^{-3} to 6.56×10^{-2}) and the median HQ values ranged from 0.00 to 2.23×10^{-3} for adults and children. In comparison to the present study, Wang et al. (2022) reported median HQ values of 0.37 to 0.63 , and 0.55 to 0.73 , for adults and children, respectively (Wang

et al. 2022). Among the four areas, the HQ values followed the order $\text{CL} > \text{BB} > \text{KM} > \text{GB}$. The values of HI computed ranged between 3.52×10^{-5} to 1.00×10^{-3} for children and 5.57×10^{-5} to 1.09×10^{-3} for adults. The median of the HI values for children and adults were 2.66×10^{-3} and 2.91×10^{-3} , respectively indicating the CTR was within the threshold level (Table ST7).

The cumulative CR for children and adults varied from 7.85×10^{-5} to 7.73×10^{-3} and 8.54×10^{-5} to 8.44×10^{-3} , respectively (Fig. 6). According to USEPA, the CR value of 10^{-6} is regarded as the threshold level. The average CR for children and adults ranged from 4.79×10^{-4} to 7.67×10^{-4} and 5.43×10^{-4} to 8.38×10^{-4} , respectively which indicate significant CR due to phthalate exposure. It is important to highlight that the cumulative CR in some sampling sites ranged between 1.16×10^{-2} and 3.21×10^{-2} , exceeding the threshold level. Overall, the study found that while the detected phthalates in tap water were within acceptable limits in terms of CTR, the threshold level of CR exceeded in some sampling sites placing the population at higher risk. Hence, regulatory interventions are required to mitigate the exposure and reduce the potential risks of phthalate contamination.

Strengths and Limitations of the Study

The present study reports the occurrence of 15 phthalates in tap water and its associated health risks in both adults and

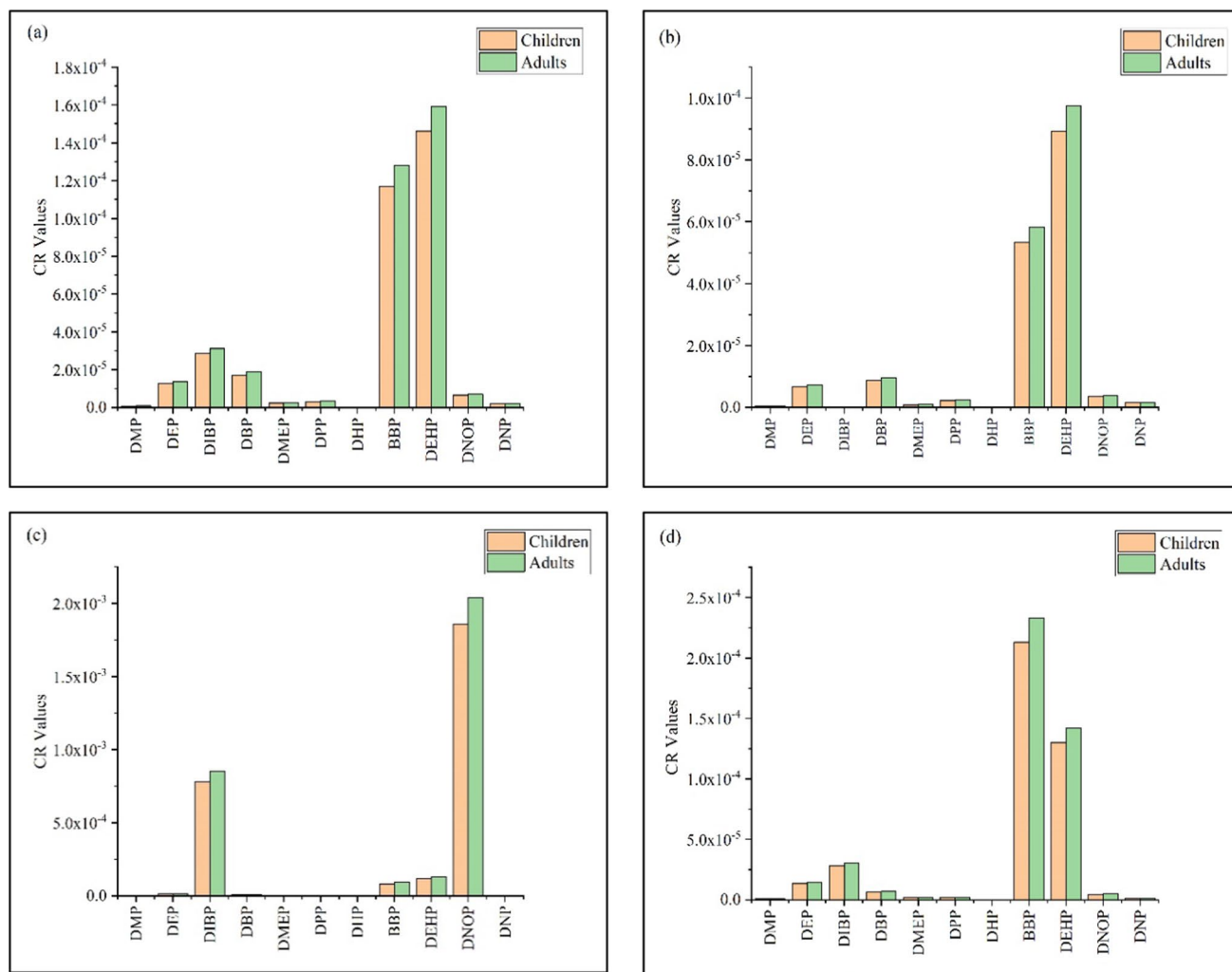


Fig. 6 Cancer risk in children and adults at **a** Butibori (BB), **b** Gandhibagh (GB), **c** Civil Lines (CL), and **d** Kalmeshwar (KM)

children of Nagpur City, India. The study presents insights into the distribution patterns of fifteen phthalates across four major areas of the city. This approach facilitated the identification of potential sources of contamination in these areas, contributing better understanding of phthalate exposure. By examining the potential health risks of phthalates in tap water, the study sheds light on the urgent need for guidelines and regulations to mitigate risks associated with phthalate exposure.

However, it is important to acknowledge the limitations of this study. Firstly, the findings reported in this study do not provide a comprehensive evaluation of phthalates exposure from all exposure routes. Migration of phthalates from plastic containers, water bottles, and kitchen wares are likely to contribute substantial phthalate exposure, which was not investigated in the present study. The study assumed a fixed ingestion rate of water per day for both adults and children. However, it is important to recognize that water

consumption can vary significantly across different seasons and individual behaviours. Finally, the authors considered only one class of chemical viz. phthalates, while the presence of other contaminants in tap water was not assessed. Therefore, it is not possible to conclude that tap water poses no CTR, despite the generally low risks calculated based on the phthalate data. Further research is needed to investigate the migration and leaching of phthalates and other additives from PVC pipes and other household plastic products into tap water. Such studies will provide a better understanding of the exposure and associated risks.

Conclusions

This study reported the occurrence of 15 phthalates in tap water samples collected from Nagpur City, India. Among the phthalates analyzed, the median concentration of DEHP was

higher than the other phthalates in all the samples. While DEHP is regulated in drinking water of several countries, no guideline values are proposed in India for monitoring and regulation. In addition to DEHP, substantial concentrations of DIBP, DBP, DnOP, and DNP were also detected in the tap water of all areas. These findings suggest that contamination of source water from non-point sources and leaching from PVC pipelines are the major sources of phthalates contamination. Though the presence of phthalates in tap water does not pose a chronic toxic risk to adults and children, it poses carcinogenic risk as the average CR exceeded the acceptable limits. The findings warrant appropriate risk reduction strategies such as regular monitoring of tap water, measures to minimize phthalate contamination of drinking water resources and establishment of regulatory levels.

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Author Contributions Nandini Shende: Conceptualization, formal analysis, Experiment, writing original draft, writing review and editing; Ishan Singh: Writing review and editing, formal analysis; Asirvatham Ramesh Kumar: Conceptualization, resources, writing review and editing, formal analysis; Girivankatesh Hippargi: Conceptualization, formal analysis, writing review and editing.

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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