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# Evaluation of heavy metal contamination in fish from the rivers of Gujarat and assessment of health risk associated with their consumption

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## ABSTRACT

This study was conducted to assess heavy metal contamination in fish and the potential risks to human health associated with fish consumption. The fish samples were collected from the urban rivers of Gujarat (Narmada River and Sabarmati River) during the summer season in 2017. Quantification of fourteen metals (Al, As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sn and Zn) was carried out using inductively coupled plasma-optical emission spectrometry (ICP-OES). The obtained results indicate that none of metal exceeds the recommended maximum acceptable levels proposed by Food Safety and Standards Authority of India (FSSAI 2016) except Lead (Pb) in Narmada River and Sabarmati River. Target hazard quotients (THQs) and hazard index (HI) were calculated to assess the health risks to the local population through the consumption of heavy metal-contaminated fish. The values of THQ and HI were less than 1 which means that consumption of these fish is not hazardous for human health. The findings of the present study may lead to measures that will be useful for regulatory bodies to control the potential health risks associated with heavy metal pollution in the future.

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**KEYWORDS:** fish, heavy metals, inductively coupled plasma-optical emission spectrometry (ICP-OES), bioaccumulation, dietary intake, target hazard quotients (THQs), hazard index (HI).

## 1. INTRODUCTION

The rapid industrialization process along with population growth and agricultural activities has increased the risk of pollution in the natural environment. Around the globe, the marine environment is heavily polluted with various organic and inorganic contaminants [1] such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), heavy metals [2] and organochlorine pesticides (OCPs) due to their bioaccumulation [3] and biomagnification in aquatic food or aquatic organisms [4]. One such pollution is aquatic pollution by heavy metals. Heavy metal contamination of aquatic ecosystem is acknowledged as a serious environmental concern [5] because of its potential toxicity, abundance, persistence and tendency to subsequent bio-accumulation [6-8].

Heavy metals are usually found in the earth's crust, which cannot be degraded or destroyed [9]. Contamination by heavy metals occurs naturally (direct atmospheric deposition, geologic weathering, run-off from adjacent agriculture lands) and anthropogenic activities (discharge through agriculture, municipal, residential/domestic or industrial waste, acid mine drainage) lead to the increase in their levels in the environment [10-14].

The consumption of fish worldwide has increased rapidly in recent years, particularly with the awareness of its nutritional and therapeutic benefits. Fish are considered the top of the food chain [14-17] and the most important organisms for the aquatic environment [18].

The major route of heavy metal exposure in humans is food consumption rather than other routes. Fish are enriched with amino acids, nutrients and omega-3 fatty acids and provides the main source of proteins [18, 19].

Fish are considered as the best indicator of contamination by pollutants because of the biomaculation and biomagnification of these pollutants in their tissues [20].

Fish have been reported as excellent indicators for heavy metal contamination in aquatic and marine environments because they occupy different levels of the food chain) [21]. In addition, heavy metals are non-biodegradable and therefore can easily be accumulated in the living organisms including fish [22]. Consequently, human beings are potentially exposed to these contaminants through the food chain with the consumption of fish [23].

The aim of the present study is to evaluate the concentration levels of heavy metals in fish samples of Narmada River and Sabarmati River of Gujarat and the human health risk associated with consumption of contaminated fish.

## 2. MATERIALS AND METHODS

### 2.1. Study area

The total area of Sabarmati basin is 21,674 $km^2$ and in Gujarat it covers about 18,550 $km^2$. The length of the river from its origin (Rajasthan) to outfall into the Gulf of Khambhat (Arabian Sea) is about 371 km. Sei, Wankal, Harnay, Hathmati, Vatrak and Meshwa are the major tributaries of the Sabarmati River. The Narmada River is the fifth longest river in India and it is known as 'Rewa' in central India. The Narmada River flows westwards a length of 1,312 km before reaching the Gulf of Khambhat (Arabian Sea). Fish samples were collected from these rivers at sites near the cities of Ahmedabad, Gandhinagar and Bharuch in the summer month of 2017 (Figure 1).

### 2.2. Sample collection and preparation

From each study site (i.e. Sabarmati and Narmada River) five fish samples were collected with the help of local fishermen. These collected fish species represent the local breed from each river, consumed in the study area. Samples were stored at -20 °C till analysis.

The fish samples were washed properly, de-scaled (where applicable) and homogenized using a blender. A portion (1.0 gm) of each homogenized sample was taken in a Teflon vessel containing 7 ml Conc. $HNO_3$ and 1 ml $H_2O_2$ and digested in MARS One microwave digestion system. The digested samples were allowed to cool and made up to a final volume of 50 ml with Milli-Q Water and filtered through 0.45 μm PTFE syringe filter. Multi elemental analysis was carried out by ICP-OES analysis.

### 2.3. Chemicals and instruments

#### 2.3.1. Reagents and standard solutions

All reagents were of highly pure analytical grade: $HNO_3$ (69% W/V, Merck); $H_2O_2$ (30% W/V, Merck); $H_2O$ (Milli-Q Water). Standard solution was prepared from the stock solution (V23 Wear Metals - 500 µg $g^{-1}$, Multi-Element Metallo-Organic CRM), purchased from VHG Labs (LGC, UK) which is traceable to the National Institute of Standard and Technology (NIST) CRMs.

#### 2.3.2. Instrumental analysis and quality assurance of samples

Heavy metal concentration quantification was done using inductively coupled plasma optical emission spectrometry (Thermo Scientific iCAP 7000 Plus Series ICP-OES) for all the digested fish samples. The wavelength lines of the ICP-OES used for the analysis of each metal were arsenic (As) 189.042, aluminium (Al) 308.215, barium (Ba) 493.409, cadmium (Cd) 226.502, cobalt (Co) 228.616, chromium (Cr) 205.560, copper (Cu) 324.754, Iron (Fe) 259.940, manganese (Mn) 257.610, molybdenum (Mo) 203.844, nickel (Ni) 231.604, lead (Pb) 220.353, tin (Sn) 189.989 and zinc (Zn) 213.856.

Certified reference materials (CRMs) were used for the quality control (QC) of heavy metal analysis by ICP-OES. The calibration curve was

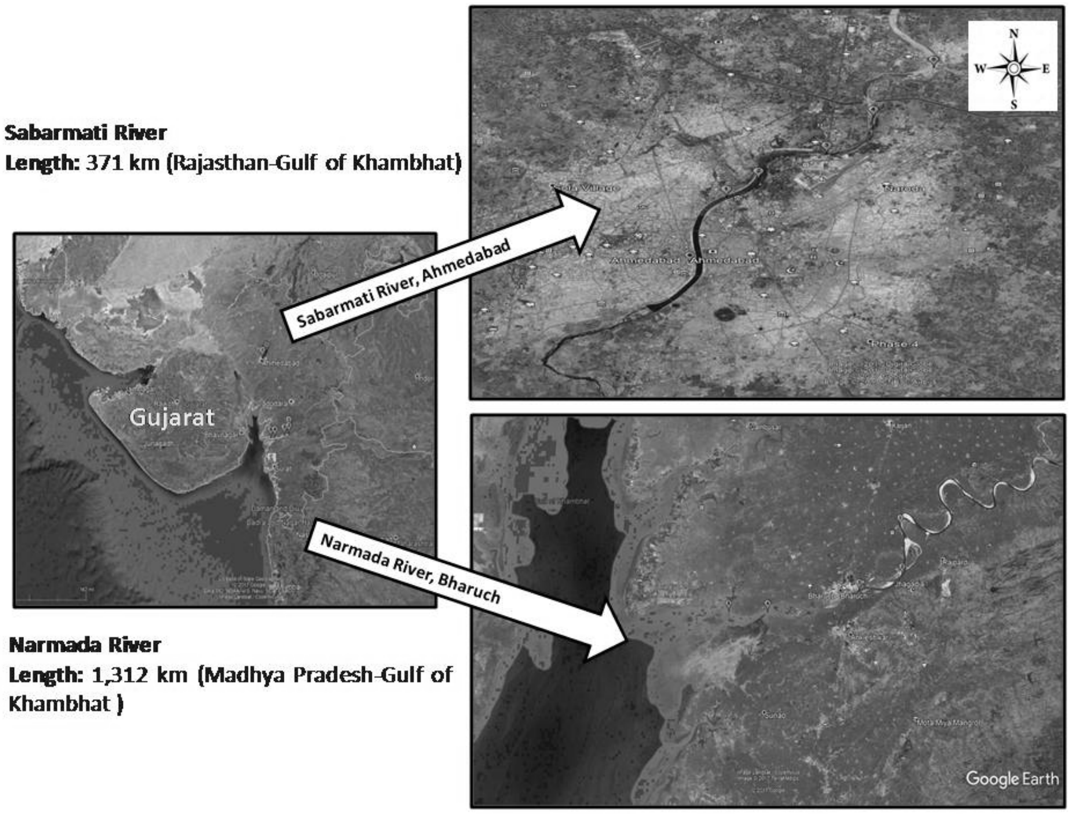


**Figure 1.** A map of the study area and sampling locations of Narmada and Sabarmati Rivers of Gujarat.

linear and the correlation coefficients ($R^2$) were $\geq$ 0.999 for all metals except aluminium (Al) and iron (Fe) which were 0.9270 and 0.9263, respectively. Method accuracy, precision and recovery % were determined by spiking the blank sample at a fortification level of 0.050 mg $kg^{-1}$ and these parameters were also used as a quality control (QC). The relative standard deviation (% RSD) was $\geq$ 16.446% and the relative recovery % of heavy metals ranges between 88.49-129.01% except for aluminium (Al) and iron (Fe) which were 553.52% and 589.30%, respectively as shown in Table 1. Recovery study was used to compensate for any losses during digestion of the sample. In all cases, the respective reagent blank was also analysed to correct the analytical data. All samples were analysed in triplicate for a good reproducibility.

### 2.4. Human health risk assessment

In the present work, the heath risk assessment due to ingestion of heavy metals from fish consumption was performed by calculating the estimated daily intake (EDI), target hazard quotient (THQ) and hazard index (HI). EDI) and THQ were calculated as described by Yabanli and Alparslan (2015) [24] and Yabanli *et al.* (2016) [25].

#### 2.4.1. Target hazard quotient (THQ)

The target hazard quotient (THQ) is used to quantify the amount of metal ingested. The target hazard quotient was calculated by using the formula given by Luczynska J. *et al.* [26].

$$THQ = 10^{-3} \times \frac{Efr \times EDtot \times FIR \times C}{RfDo \times BWa \times ATn}$$

where, Efr - exposure frequency (365 days/year), EDtot - exposure duration (69 years) [27], FIR - average daily consumption of fish (gm $day^{-1}$), C is the concentration of metals in fish (mg $kg^{-1}$), BWa is the average body weight (60 kg), RfDo - oral reference dose (mg/kg/day) (USEPA - 2015) [28]

**Table 1.** ICP-OES analytical parameters and quality check of certified reference materials (CRMs).

| Sr. no. | Elements | Emission line/ wavelength (nm) | Plasma view | Correlation coefficients ($R^2$) | Certified value (mg $kg^{-1}$) | Observed value (mg $kg^{-1}$) | % RSD | Recovery (%) |
|---|---|---|---|---|---|---|---|---|
| 1 | Al | 308.215 | Axial | 0.9270 | 0.050 | 0.277 | 0.742 | 553.52 |
| 2 | As | 189.042 | Axial | 0.9937 | 0.050 | 0.062 | 1.585 | 124.26 |
| 3 | Ba | 493.409 | Axial | 0.9940 | 0.050 | 0.065 | 0.596 | 129.01 |
| 4 | Cd | 226.502 | Axial | 0.9952 | 0.050 | 0.055 | 0.177 | 109.96 |
| 5 | Co | 228.616 | Axial | 0.9948 | 0.050 | 0.055 | 0.265 | 110.29 |
| 6 | Cr | 205.560 | Axial | 0.9959 | 0.050 | 0.058 | 0.068 | 115.83 |
| 7 | Cu | 324.754 | Axial | 0.9964 | 0.050 | 0.054 | 0.806 | 108.37 |
| 8 | Fe | 259.940 | Axial | 0.9263 | 0.050 | 0.295 | 3.466 | 589.30 |
| 9 | Mn | 257.610 | Axial | 0.9956 | 0.050 | 0.056 | 0.424 | 111.78 |
| 10 | Mo | 203.844 | Axial | 0.9979 | 0.050 | 0.044 | 2.142 | 88.49 |
| 11 | Ni | 231.604 | Axial | 0.9955 | 0.050 | 0.056 | 0.122 | 111.53 |
| 12 | Pb | 220.353 | Axial | 0.9946 | 0.050 | 0.055 | 0.954 | 110.67 |
| 13 | Sn | 189.989 | Axial | 0.9971 | 0.050 | 0.053 | 16.446 | 105.31 |
| 14 | Zn | 213.856 | Axial | 0.9933 | 0.050 | 0.061 | 0.324 | 121.77 |

**Table 2.** Heavy metal concentration (mg $kg^{-1}$) in fish samples of Narmada and Sabarmati Rivers, Gujarat.

| | Heavy metals | Al | As | Ba | Cd | Co | Cr | Cu | Fe | Mn | Mo | Ni | Pb | Sn | Zn |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Narmada River (n = 5) | **Min.** | 13.6027 | 0.3235 | 0.5926 | 0.0064 | 0.0672 | 2.9181 | 0.9678 | 37.3108 | 2.2243 | 0.1818 | 1.3268 | 0.3111 | 0.0000 | 13.2072 |
| | **Max.** | 505.3091 | 4.0803 | 1.3083 | 0.1025 | 0.7320 | 5.2427 | 2.6661 | 868.2640 | 14.4647 | 0.4185 | 2.2288 | 2.0289 | 0.0483 | 45.5957 |
| | **Average** | 139.3067 | 1.7613 | 0.8546 | 0.0410 | 0.2599 | 3.9432 | 1.6160 | 273.1396 | 5.8423 | 0.2795 | 1.5762 | 1.0602 | 0.0483 | 21.4351 |
| | **S.D.** | 211.6349 | 1.4960 | 0.2832 | 0.0471 | 0.2877 | 0.9504 | 0.8036 | 362.1235 | 5.0612 | 0.0953 | 0.3763 | 0.7960 | 0.0216 | 13.6143 |
| Sabarmati River (n = 5) | **Min.** | 10.9959 | 0.4065 | 0.5227 | 0.0048 | 0.0600 | 3.0597 | 0.8781 | 29.9129 | 3.8938 | 0.2444 | 1.4190 | 0.2347 | 0.0000 | 7.9132 |
| | **Max.** | 23.9022 | 0.5831 | 1.2987 | 0.0798 | 0.1368 | 3.6958 | 1.9423 | 86.4632 | 11.7025 | 0.4194 | 2.2405 | 0.3937 | 0.1963 | 19.2356 |
| | **Average** | 18.2521 | 0.4875 | 0.8082 | 0.0350 | 0.0916 | 3.3278 | 1.5160 | 54.2480 | 7.9963 | 0.3253 | 1.7986 | 0.3010 | 0.1295 | 12.2442 |
| | **S.D.** | 5.0811 | 0.0773 | 0.3044 | 0.0310 | 0.0317 | 0.2380 | 0.4017 | 21.2954 | 2.7928 | 0.0809 | 0.3798 | 0.0641 | 0.0766 | 4.1916 |
| | **FSSAI (2016)** | - | 76.0000 | - | 0.3000 | - | 12.0000 | - | - | - | - | - | 0.3000 | 200.0000 | - |

Min. means minimum; Max. means maximum; S.D. means standard deviation;
Note: The obtained concentrations are based on wet weight of fish.

**Table 3.** Estimated target hazard quotient (THQ) and hazard index (HI) of individual metals in samples (n = 5) collected from Narmada and Sabarmati Rivers of Gujarat.

| **Heavy metals** | | **Al** | **As** | **Ba** | **Cd** | **Co** | **Cr** | **Cu** | **Fe** | **Mn** | **Mo** | **Ni** | **Pb** | **Sn** | **Zn** |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **RfDo (mg/kg-day)** | | 1.0000 | 0.0003 | 0.2000 | 0.0001 | 0.0003 | 1.5000 | 0.0400 | 0.7000 | 0.1400 | 0.0050 | 0.0200 | 0.0036 | 0.6000 | 0.3000 |
| **Narmada River** | **THQ** | 0.0040 | 0.1696 | 0.0001 | 0.0119 | 0.0250 | 0.0001 | 0.0012 | 0.0113 | 0.0012 | 0.0016 | 0.0023 | 0.0085 | 0.0000 | 0.0021 |
| | **HI** | 0.2388 | | | | | | | | | | | | | |
| **Sabarmati River** | **THQ** | 0.0005 | 0.0469 | 0.0001 | 0.0101 | 0.0088 | 0.0001 | 0.0011 | 0.0022 | 0.0017 | 0.0019 | 0.0026 | 0.0024 | 0.0000 | 0.0012 |
| | **HI** | 0.0796 | | | | | | | | | | | | | |

THQ and HI represent target hazard quotient and hazard index respectively; RfDo is oral reference dose.

and ATn - average exposure for non-carcinogens in the year (365 days/year × 69 years). If the THQ value is below 1, it indicates that no adverse effect for human health [26] is likely to occur as a result of the ingestion of fish. However, as the value of the THQ increases, the level of the health risk associated with the ingestion of fish also increases.

#### 2.4.2. Hazard index (HI)

It is calculated as the sum of THQ values of all metals. It is considered that there would be no significant risk of non-carcinogenic effects if the value of HI < 1, but if it is > 1, then the probability of occurrence of non-carcinogenic health effects would exist.

$$HI = THQ_{(Al)} + THQ_{(As)} + THQ_{(Ba)} + THQ_{(Cd)} + THQ_{(Co)} + THQ_{(Cr)} + THQ_{(Cu)} + THQ_{(Fe)} + THQ_{(Mn)} + THQ_{(Mo)} + THQ_{(Ni)} + THQ_{(Pb)} + THQ_{(Sn)} + THQ_{(Zn)}$$

## 3. RESULTS AND DISCUSSION

The obtained average concentrations of analyzed heavy metals for Al, As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sn and Zn were 139.3067, 1.7613, 0.8546, 0.0410, 0.2599, 3.9432, 1.6160, 273.1396, 5.8423, 0.2795, 1.5762, 1.0602, 0.0483 and 21.4351 (mg $kg^{-1}$ww) in Narmada River and 18.2521, 0.4875, 0.8082, 0.0350, 0.0916, 3.3278, 1.5160, 54.2480, 7.9963, 0.3253, 1.7986, 0.3010, 0.1295, 12.2442 (mg $kg^{-1}$ww) in Sabarmati River (Table 2). When comparison was done between the samples of both the study sites (i.e. Narmada and Sabarmati River), the level of all the heavy metals was significantly lower in Sabarmati than the Narmada river except for Manganese (Mn), Molybdenum (Mo), Nickel (Ni) and Tin (Sn). Furthermore, the concentrations of analyzed metals in the tested fish samples did not exceed the recommended maximum acceptable levels proposed by Food Safety and Standards Authority of India (FSSAI 2016) [29] except for Lead (Pb - 1.0602 and 0.3010 mg $kg^{-1}$ww in Narmada and Sabarmati River, respectively). In terms of the target hazard quotients (THQs) of individual metals, none of metal exceeded 1, which indicate that it is safe for human health. Hazard index (HI) values for Narmada River and Sabarmati River were 0.2388 and 0.0796, respectively. There is no potential health risk due to consumption of fish because hazard index (HI) was also less than 1 (Table 3).

## 4. CONCLUSION

The present study identified the presence of a wide range of heavy metals in fish samples consumed in study sites. The overall results show that the fish of the Narmada River is more contaminated by heavy metals than the Sabarmati River. The level of Lead (Pb) in both rivers was significantly higher, which is alarming for human health. Therefore consumption of fish may have a potential adverse health impact in human. However, THQ and HI values were less < 1, which reflects no harm for human health. This present work could help in understanding the current status of possible health risk associated with heavy consumption in general population as well as aid the regulatory bodies to control excessive use/emission of heavy metals and further to take control measures to reduce public health risk in future.

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## CONFLICT OF INTEREST STATEMENT

The authors declare that there is no conflict of interest.

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