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# Elemental characterization of medicinal plants of the Sundarban by INAA and AAS techniques: health risk assessments and statistical analysis

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**Abstract**
This study evaluates essential and toxic element contents in three widely used medicinal plants (*Acanthus ilicifolius*, *Avicennia officinalis*, and *Xylocarpus mekongensis*) of the Sundarban mangrove ecosystem and the possible health risks through the consumption of these plants. Total concentrations of 20 chemical elements (Al, As, Br, Ca, Cd, Ce, Co, Cr, Fe, K, La, Mn, Na, Pb, Sb, Sc, Sm, Th, V, and Zn) were determined by instrumental neutron activation analysis (INAA) and atomic absorption spectrometry (AAS) techniques. The determined concentrations of the elements were compared with available WHO permissible limits which indicate that the average concentration of Zn in *A. ilicifolius* (41.2 ± 10.1 mg/kg) was higher than that of the permissible WHO limit (27 mg/kg). The average concentrations of Cr in *A. ilicifolius* (3.35 ± 0.37 mg/kg) and *X. mekongensis* (4.02 ± 2.30 mg/kg) were also higher than that of the WHO permissible limit (2 mg/kg). However, the average daily intake of the toxic elements was below the maximum tolerable daily intake (MTDI) values. The target hazard quotient (THQ) values were also within the permissible limit (THQ < 1). The elemental transfer factors and pollution index for the medicinal plants were determined and statistical analyses were used to clarify the elemental correlations in the studied plants. Therefore, this study will give valuable information to the public about the elemental contents and health risks due to consumption of the medicinal plants.

**Keywords** Element contents · Instrumental neutron activation analysis · Atomic absorption spectrometry · Medicinal plants · Health risk assessment · Sundarban mangrove ecosystem

## Introduction

Herbal medicines are getting considerable interest in the global health sector [1]. In developing countries, some issues like the excessive fee for drug treatments, socio-economic, wide biodiversity, and the complexity of going through the health system, make herbal medicine more popular day by day. Therefore, stronger use is facilitated by easy access to plants and herbal medicines as alternatives to synthetic drugs [2, 3]. However, recent studies have proven that the toxic metallic contamination of medicinal plants has increased due to numerous human activities, including the use of chemical fertilizers and pesticides, irrigation with wastewater, and other industrialization processes like mining and smelting of non-ferrous metal during production [4–7]. Plants can absorb toxic elements from contaminated soils or deposits on parts of them exposed to the air in polluted environments [8, 9]. For plants, elements can generally be categorized into two types– essential (calcium, zinc, manganese, copper, iron, etc.) and toxic (cadmium, arsenic, lead, nickel, mercury, etc.) [10]. Due to nutritional, chemical, and biological properties, essential elements play a crucial role in the function of the body [11]. On the other hand, human and animal organs may damage due to some properties of toxic elements such as long biological half-lives, high accumulation behavior, and non-biodegradable behavior in diverse body parts [12, 13]. Moreover, non-carcinogenic, carcinogenic, and mutagenic effects in the body are led by the accumulation of toxic elements.

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Nuclear analytical techniques have the potential to analyze environmental and food samples. Among the different nuclear analytical techniques, instrumental neutron activation analysis (INAA) is one of the highly sensitive and multi-element analysis techniques to get precise and accurate element contents in a wide variety of samples [14–16]. This method is considered a primary method of measurement by metrology [17]. Moreover, the World Health Organization (WHO) has recommended NAA as an analytical method applicable to plant materials [18]. The INAA method can be used in conjunction with atomic absorption spectrometry (AAS) to determine a variety of elements in food samples.

Sundarban is the largest single-block mangrove forest in the world. It is a rich ecosystem with diverse flora and fauna. Three widely used medicinal plants of the Sundarban mangrove forest are *A. ilicifolius*, *A. officinalis*, and *X. mekongensis*. Among them, *A. ilicifolius* is a perennial plant, popularly known as "Holy leaved acanthus" which is locally known as a Hargoja. Phytochemical research on this plant has shown the presence of glycosides, alkaloids, flavonoids, triterpenoids, steroids, fatty acids, and saponins in it [19]. This plant is used as a crude drug to cure diabetes, asthma, dyspepsia, hepatitis, leprosy, paralysis, diuretics, snake bite, and rheumatoid arthritis [20, 21]. Besides this, *A. officinalis* is an evergreen species of mangrove that is mainly found in tropical countries but is not widely introduced elsewhere [22]. It is used as a folk remedy for boils and tumors [23]. The leaves of *A. officinalis* contain many important phytochemicals [24]. In the presence of such phytochemicals, *A. officinalis* can be a source of medicinal properties like analgesic, antioxidant, antimicrobial, or anticancer activities. Another important mangrove medicinal plant is *X. mekongensis*, which contains various essential minerals and phytochemicals [25]. As a result, it has been used traditionally for the treatment of fever, diarrhea, dysentery, inflammation, and abdominal disorders [25]. A few researches have been carried out on *A. ilicifolius*, and *A. officinalis* plants in the Indian part of Sundarban and estimated that the mangrove ecosystem is affected by potentially toxic elements [26]. There is no comprehensive research work on transfer factors and health risk assessments of the toxic elements of the medicinal plants of the Sundarban. The aims of this research are (i) to characterize the essential and toxic elements in these commonly used medicinal plants by INAA and AAS, (ii) to study the transfer factor of the elements and possible health risks related to the use of these therapeutic plants, and finally (iii) to apply statistical approaches to find out correlations of the elements in the studied plants.

## Materials and methods

### Study area

The Sundarban is located between 21° 32′ to 22° 40′ N and 88° 05′ to 89° 51′ E, covering an area of about 10,000 $km^2$ with 62% in Bangladesh and 38% in India [26]. The ecosystem is highly productive and diverse, as long as there are a variety of specific ecosystem resources for local people. On the other hand, mangroves function as a defense against the adverse due of various anthropogenic behaviors such as industrial effluents, fishing, tourism, port activities, agriculture, fish aquaculture, shipbreaking, etc. [26].

### Sample collection and preparation

Leaf samples of three different species (3 samples of each species from each location were used to make a composite sample) of medicinal plants, *A. ilicifolius*, *A. officinalis*, and *X. mekongensis*, and nine corresponding composite rhizosphere soil samples were also collected from 3 different locations of the Bangladeshi Sundarban. The sampling locations are shown in Fig. 1. Leaves of these plants are extensively used for the treatment of different diseases. To remove soil and dust particles, the leaf samples were washed several times initially with deionized water. Both soil and leave samples were heated discretely at 40 °C in the microwave oven until attain a constant weight. Then the samples were grounded using a motor and pestle, sieved, homogenized, and pulverized. The pulverized fraction was stored in clean, dry, and airtight bottles for further analysis.

### Sample analysis

For quantification of the elements by INAA, samples were irradiated along with reference materials at the Pneumatic Transfer System and Dry Central Thimble irradiation facilities of the TRIGA MARK-II research reactor of Bangladesh Atomic Energy Commission (BAEC). The irradiated samples' gamma-ray spectrometry was performed using a high-resolution HPGe detector system (40% relative efficiency). The energy resolution of the detector is 1.8 keV at 1332 keV energy peak of $^{60}Co$ gamma-ray line and the peak-to-Compton ratio is 63:1. For element determination using INAA, approximately 50 mg of each dried powder soil and 150 mg of plant leaf sample were measured in a polyethylene bag and heat sealed. In this study, one certified reference material (CRM) from International Atomic Energy Agency (IAEA): IAEA-336 (Lichen), and one standard reference material NIST-1547 (Peach leaves) along with the plant samples were analyzed. Similarly, soil samples were


**Fig. 1** Sampling locations at the Sundarban mangrove ecosystem

irradiated with IAEA-Soil-7 and IAEA-SL-1. For the determination of element concentrations, the relative standardization method of INAA was applied. Targeting the short and long half-life radioactive nuclei short-term and long-term types of irradiation were performed. Short and long irradiation of each sample/standard was performed at a thermal neutron flux of $1.77 \times 10^{12}$ cm$^{-2}$·s$^{-1}$ for 1 min at 250 kW and $1.70 \times 10^{13}$ cm$^{-2}$·s$^{-1}$ for 7 min at 2.4 MW, respectively. The neutron flux gradient inside the sample stack is measured by inserting IRMM-530RA Al-0.1% Au (0.1 mm foil) monitor foils placed on the bottom, in the middle, and on top of the stack. For AAS analysis, one gram of each medicinal plant and soil sample and the same standards used for INAA were digested. The concentrations of Cd and Pb in medicinal plant and soil samples were determined by a graphite furnace AAS following the standard procedures [27, 28].

## Quality control and quality assurance

To ensure reported data quality (accuracy and precision) repeated analyses of reference materials NIST-1547 and IAEA-Soil-7 were performed along with the medicinal plant and rhizosphere soil samples. To determine laboratory performance U-score and deviation (%) of concentration values from the certified/information values of the standards were calculated. The following equation was used to calculate U-score [29]:

$$\text{U} - \text{Score} = \frac{\left|X_{lab} - X_{ref}\right|}{\sqrt{\mu_{lab}^2 + \sigma_{ref}^2}}$$

where, $X_{lab}$, $X_{ref}$, $\mu_{lab}$, and $\sigma_{ref}$ are the laboratory result, reference value, uncertainty due to standard deviation (1σ) of the laboratory result, and standard uncertainty with the reference value, respectively. If the value of U-score ≤ 1, the laboratory performance is satisfactory, and if U-score > 1 (result and certified value are not in agreement) then the laboratory performance is unsatisfactory.

## Average daily intake

The United States Environmental Protection Agency (USEPA) developed a method to assess the nature and probability of health hazards in humans that may have been exposed to pollutants. The average daily intake (ADI) of heavy metal (mg/kg/day) through medicated diet is referred to as ADI which was calculated by the following equation:

$$\text{ADI} = \frac{EF \times ED \times IR \times C_{element}}{B_{weight} \times AT}$$

where EF is the exposure frequency (365 day/a; a = annum) [30], ED is the exposure duration (30 a) [31], IR is the ingestion rate of herbs (0.009 kg/d) [32], $C_{element}$ is the element concentrations in herbs (mg/kg), AT is the average time for non-carcinogens (365 × ED day) [33], and $B_{weight}$ is the person's body weight (60 kg).

### Target hazard quotient

The health risks associated with the use of herbal medicines by local people were determined by using THQ [34]. There is a health risk that can occur if the total THQ value is equal to or greater than 1. THQ values are calculated as follows:

$$THQ = \frac{ADI}{RfD}$$

Here, 0.0003, 0.0001, 0.003, 0.14, and 0.3 mg/kg/day are the oral reference doses (RfD) for As, Cd, Cr, Mn, and Zn, respectively [34]. For identifying the non-carcinogenic health risks, THQ plays a vital role.

### Target carcinogenic risk

For estimating concentration-dependent lifetime cancer risk the following equation was employed:

$$TCR = CSF \ X \ ADI$$

where, CSF is the carcinogenic slope factor and ADI is the estimated average daily intake of elements in the body, respectively. The slope factors of As and Cr are 1.5 and 0.5 $(mg/kg/day)^{-1}$, respectively [34].

### Determination of transfer factor

The transmissions of elements from soil to plant tissues are studied using an index called Transfer Factor (TF) which is calculated using the following equation [35]:

$$TF = C_{plant}/C_{soil}$$

where $C_{plant}$ is the metal concentration in plant tissue and $C_{soil}$ is the metal concentration in soil.

### Pollution index

The pollution index (PI) value is used to estimate the metal contamination status of herbal medicine. The following equation was used to determine PI.

$$P_i = C_i/S_i$$

where $P_i$ is the pollution index of certain metals, $C_i$ is the estimated value of certain heavy metal concentrations in plants and $S_i$ is the evaluation standard values. The values of the evaluation standard for As, Cd, Cr, Pb, and Zn in herbal medicines are 10, 0.2, 2, 10, and 50 mg/kg, respectively according to FAO/WHO standards [36].

## Results and discussion

### Concentrations of the elements in medicinal plants

Elemental concentrations (mg/kg) of the studied medicinal plants *A. ilicifolius*, *A. officinalis*, and *X. mekongensis*, and rhizosphere soil samples were determined using INAA and AAS techniques. The quality control of the analytical data was assessed by analyzing reference standards and given in Tables S1 and S2 (Supplementary data). The overall calculated U-score and percentage deviations ensured that laboratory results were satisfactory. Twenty chemical elements (Al, As, Br, Ca, Cd, Ce, Co, Cr, Fe, K, La, Mn, Na, Pb, Sb, Sc, Sm, Th, V, and Zn) were determined in the studied medicinal plants and rhizospheric soil samples tabulated in Tables 1 and 2. These elements could be divided into three categories: essential elements (Ca, Co, Fe, K, Mn, Na, and Zn); toxic elements (As, Br, Cd, Cr, and Pb); other elements (Al, Ce, La, Sb, Sc, Sm, Th, and V). The human body desires several elements which are taken into consideration as important for its growth; these elements are necessary components and play an important physiological function in which Ca is a vital mineral for human health, contributing to the formation of bones and teeth. The concentrations of Ca in *A. ilicifolius*, *A. officinalis*, and *X. mekongensis* were 3220, 3520, and 6580 mg/kg, respectively. The K is the most common place for intracellular ions in human and animal cells and is also abundant in dietary count [37, 38]. It is noteworthy that, the contents of K in *A. ilicifolius*, *A. officinalis*, and *X. mekongensis* were found to be 20,600, 15,300, and 25,300 mg/kg which indicates that they are the potential source for K. Among the studied medicinal plants, the highest concentrations of Ca and K were found in the *X. mekongensis* plant. Iron is a vital element, which is an important component of hemoglobin that transports oxygen [39]. In the present study, Fe concentrations varied from 157 to 1160 mg/kg; the maximum concentration of iron was found in *A. ilicifolius*, (1160 mg/kg) as shown in Table 1. Therefore, *A. ilicifolius* could be a possible natural source/supplement for this micronutrient.

Zinc is another vital trace element needed for proper growth, thyroid function, blood clotting, and protein and DNA synthesis. But excessive Zn can be harmful and cause toxicity to human health [40]. Excessive amounts of Zn may also lead to abdominal pain, diarrhea, vomiting, and nausea [41], while Zn deficiency leads to hypogonadism and growth retardation [42]. The Zn concentrations for this study were found to be 41.2 mg/kg, 18.7 mg/kg, and 11.2 mg/kg for *A. ilicifolius*, *A. officinalis*, and *X. mekongensis*, respectively (Table 1). The Zn concentration in *A. ilicifolius* is greater than the WHO's maximum permissible

**Table 1** Mean values (n = 3) of elemental concentrations (mg/kg) determined in three medicinal plants by the INAA and AAS

| Elements | *A. ilicifolius* | *A. officinalis* | *X. mekongensis* | WHO/FAO values [43] |
|---|---|---|---|---|
| *Essential elements* | | | | |
| Ca | 3220 ± 154 | 3520 ± 430 | 6580 ± 1180 | |
| Co | 0.431 ± 0.015 | 0.664 ± 0.099 | 0.532 ± 0.087 | 0.48 |
| Fe | 1160 ± 122 | 157 ± 34 | 837 ± 404 | |
| K | 20,600 ± 1710 | 15,300 ± 5190 | 25,300 ± 2990 | |
| Mn | 104 ± 22 | 54.5 ± 17.8 | 37.4 ± 10.3 | |
| Na | 16,700 ± 1800 | 13,600 ± 790 | 13,000 ± 2500 | |
| Zn | 41.2 ± 10.1 | 18.7 ± 0.3 | 11.2 ± 1.6 | 27 |
| *Potential toxic elements* | | | | |
| As | 0.0182 ± 0.0047 | 0.0433 ± 0.0164 | 0.0144 ± 0.0044 | 5.0 |
| Br | 132 ± 4 | 122 ± 11 | 146 ± 22 | |
| Cd[a] | 0.026 ± 0.002 | 0.021 ± 0.002 | 0.020 ± 0.002 | 0.3 |
| Cr | 3.35 ± 0.37 | 0.829 ± 0.272 | 4.02 ± 2.30 | 2 |
| Pb[a] | 0.939 ± 0.078 | 1.00 ± 0.09 | 1.12 ± 0.07 | 10 |
| *Other elements* | | | | |
| Al | 1790 ± 173 | 207 ± 62 | 1820 ± 1020 | |
| Ce | 1.99 ± 0.15 | < 0.187 | 1.88 ± 1.11 | |
| La | 1.49 ± 0.23 | 0.325 ± 0.060 | 1.47 ± 0.72 | |
| Sb | 0.0458 ± 0.0151 | 0.0313 ± 0.0048 | 0.0180 ± 0.0134 | |
| Sc | 0.376 ± 0.051 | 0.0388 ± 0.0081 | 0.273 ± 0.138 | |
| Sm | 0.167 ± 0.038 | < 0.024 | 0.147 ± 0.053 | |
| Th | 0.603 ± 0.047 | 0.0406 ± 0.0027 | 0.633 ± 0.379 | |
| V | 2.75 ± 0.32 | < 0.648 | 3.10 ± 2.05 | |

[a]Determined by AAS technique. Uncertainties are due to standard deviation (1σ) for n = 3

limit (27 mg/kg) [43]. Concentrations of Zn were obtained to be 111 mg/kg, and 97.4 mg/kg for *A. ilicifolius* and *A. officinalis* medicinal plants in the Indian part of Sundarban which were higher than the presently studied values [26]. Besides this, Co is an important micronutrient needed for the metabolic processes of humans, animals, and plants. It is available with vitamin B12, which is required in producing red blood cells and in the prevention of anemia [41]. In this study, Co concentrations were found to be 0.431 mg/kg, 0.664 mg/kg, and 0.532 mg/kg for *A. ilicifolius*, *A. officinalis*, and *X. mekongensis*, respectively. Therefore, the concentrations of Co except in *A. ilicifolius*, all medicinal plants were slightly higher than the WHO permissible limit of 0.48 mg/kg [43]. Although Co is a vital element, it can be harmful if consumed in larger amounts. Excess consumption of Co can cause blood pressure, cardiomyopathy, diarrhea, pulmonary disorders, vomiting, slowed respiration, and so on, whereas, its deficiency consequences in anemia, loss of weight, and retarded growth [41]. Besides this, Al is an element that has a negative impact on plants [44]. The Al concentrations were obtained in the present study by 1790, 207, and 1820 mg/kg in *A. ilicifolius*, *A. officinalis*, and *X. mekongensis*, respectively. Manganese is an important element as well as a significant activator of the enzyme. The concentrations of Mn were found to be 104, 54.5, and 37.4 mg/kg for *A. ilicifolius*, *A. officinalis*, and *X. mekongensis*, respectively whereas the concentration of the same element in *A. ilicifolius*, and *A. officinalis*, plants were found to be 178, and 163 mg/kg in the Indian part of Sundarban [26] which are much higher than the same plants in Bangladeshi part of the Sundarban.

The elements As, Cd, and Pb are considered toxic elements, and Br, Sb, and Cr are occasionally also considered toxic elements. Arsenic is carcinogenic and can lead to cancer of the liver, bladder, lungs, and skin; besides this, inorganic arsenic intake at lower concentrations can cause abnormal heart rhythm and reduced red and white blood cell production [45]. The As concentrations in the present study were obtained for *A. ilicifolius*, *A. officinalis*, and *X. mekongensis* plants as 0.018 mg/kg, 0.043 mg/kg, and 0.014 mg/kg, respectively, which are below the WHO/FAO permissible limit 5 mg/kg [43]. Besides this, the Cd concentrations were found to be 0.026 mg/kg, 0.021 mg/kg, and 0.020 mg/kg for *A. ilicifolius*, *A. officinalis*, and *X. mekongensis* plants, whereas the WHO/FAO permissible limit is 0.3 mg/kg [43], and the concentrations of the same element for *A. ilicifolius*, and *A. officinalis* in the Indian part of Sundarban [26] were determined as 1.6 mg/kg and 1.9 mg/kg which

**Table 2** Elemental concentrations (mg/kg) in the rhizosphere soil samples

| Elements | *A. ilicifolius* | *A. officinalis* | *X. mekongensis* |
|---|---|---|---|
| Al | 87,300 ± 9100 | 96,200 ± 3700 | 88,700 ± 3600 |
| As | 9.58 ± 0.05 | 10.2 ± 0.1 | 8.87 ± 0.82 |
| Br | 18.1 ± 4.8 | 16.9 ± 1.2 | 21.1 ± 7.2 |
| Ca | 10,600 ± 3700 | 15,500 ± 1200 | 12,100 ± 2000 |
| Ce | 77.9 ± 2.9 | 94.3 ± 3.3 | 90.2 ± 2.3 |
| Cd[a] | 0.43 ± 0.03 | 0.45 ± 0.02 | 0.36 ± 0.01 |
| Co | 15.7 ± 0.7 | 18.9 ± 0.2 | 18.2 ± 0.1 |
| Cr | 82.7 ± 4.8 | 99.3 ± 2.4 | 96.0 ± 0.9 |
| Fe | 38,900 ± 1800 | 47,500 ± 1000 | 44,300 ± 1900 |
| K | 33,000 ± 3500 | 36,000 ± 1600 | 37,300 ± 1400 |
| La | 42.0 ± 3.7 | 42.7 ± 1.7 | 40.1 ± 3.0 |
| Mn | 740 ± 51 | 927 ± 16 | 943 ± 12 |
| Na | 9100 ± 690 | 8900 ± 1600 | 8400 ± 890 |
| Pb[a] | 15.8 ± 0.5 | 19.9 ± 0.3 | 14.6 ± 0.4 |
| Sb | 0.794 ± 0.044 | 1.42 ± 0.72 | 0.856 ± 0.067 |
| Sc | 14.1 ± 0.8 | 16.9 ± 0.2 | 16.1 ± 0.4 |
| Sm | 6.75 ± 0.92 | 6.40 ± 0.18 | 6.52 ± 0.18 |
| Th | 17.6 ± 0.8 | 20.8 ± 0.8 | 20.1 ± 0.6 |
| V | 108 ± 8 | 122 ± 7 | 116 ± 2 |
| Zn | 90.2 ± 11.2 | 100 ± 1 | 89.1 ± 12.6 |

[a]Determined by AAS technique. Uncertainties are due to standard deviation (1σ) for n = 3

are greater than the Bangladeshi part of Sundarban. Among the toxic elements, Pb is very harmful to humans, plants, and microorganisms [46]. Exposure to Pb can cause serious disturbances resulting in permanent complications with brain problems [41]. In the present study, the Pb concentrations in *A. ilicifolius*, *A. officinalis*, and *X. mekongensis* plants were obtained to be 0.94 mg/kg, 1.01 mg/kg, and 1.1 mg/kg, respectively, where WHO/FAO permissible limit for medicinal plants is 10 mg/kg [43] but in the Indian part of Sundarban, the concentration of this element for *A. ilicifolius*, and *A. officinalis*, was estimated 27.3 mg/kg and 36.6 mg/kg which are higher than that of the Bangladeshi part of Sundarban as well as WHO/FAO permissible limit. Likewise, the Cr concentrations in *A. ilicifolius*, *A. officinalis*, *and X. mekongensis* were 3.3 mg/kg, 0.83 mg/kg, and 4.0 mg/kg, respectively. According to the WHO/FAO, the maximum permissible limit of Cr is 2 mg/kg [43]; the concentrations levels of Cr obtained from this study for *A. ilicifolius*, *and X. mekongensis*, were found to be higher than the tolerable limit of the WHO/FAO limit. Therefore, it can be said that the metal concentrations of studied plants were higher in the Indian part of Sundarban compared to the Bangladeshi part of Sundarban. On the other hand, the elemental concentrations in the rhizosphere soil samples are given in Table 2, where most of the elemental concentrations were higher than the determined values in the previous study for surface sediments in Sundarban [27]. The average concentrations of As, Cr, and Pb were obtained in the present study as 8.47 mg/kg, 92.6 mg/kg, and 16.8 mg/kg whereas the concentrations of the same elements were found in the previous study [27] as 6.76 mg/kg, 67 mg/kg, and 15.8 mg/kg, respectively. Therefore, concentrations of these toxic elements in Sundarban soils have increased which may affect their concentrations in medicinal plants of this mangrove ecosystem.

**Table 3** Comparison of the average daily intake (ADI, mg/day) of the elements in medicinal plants with the corresponding maximum tolerable daily intake (MTDI) in the Bangladeshi population

| Elements (mg/kg) | *A. ilicifolius* | *A. officinalis* | *X. mekongensis* | MTDI value |
|---|---|---|---|---|
| As | $2.7 \times 10^{-6}$ | $6.5 \times 10^{-6}$ | $2.2 \times 10^{-6}$ | 0.126 |
| Cd | $3.9 \times 10^{-6}$ | $3.2 \times 10^{-6}$ | $3.0 \times 10^{-6}$ | 0.021 |
| Cr | $5.0 \times 10^{-4}$ | $1.2 \times 10^{-4}$ | $6.0 \times 10^{-4}$ | 0.2 |
| Mn | $1.6 \times 10^{-2}$ | $8.2 \times 10^{-3}$ | $5.6 \times 10^{-3}$ | 2–5 |
| Pb | $1.4 \times 10^{-4}$ | $1.5 \times 10^{-4}$ | $1.7 \times 10^{-4}$ | 0.21 |
| Zn | $6.1 \times 10^{-3}$ | $2.8 \times 10^{-3}$ | $1.7 \times 10^{-3}$ | 60 |

*MTDI* Maximum tolerable daily intake

## Health risk assessments

### Average daily intake values

The estimated ADI values for *A. ilicifolius*, *A. officinalis*, *and X. mekongensis* were tabulated in Table 3. The ADI value of As in these medicinal plants was found in the range of $2.2 \times 10^{-6}$ to $6.5 \times 10^{-6}$ mg/day, whereas the maximum tolerable daily intake (MTDI) is 0.126 mg/day [47]. Similarly, ADI values for Cd, Cr, Mn, Pb, and Zn in *A. ilicifolius*, *A. officinalis*, *and X. mekongensis*, plants were obtained in the range of $3.0 \times 10^{-6}$ to $3.9 \times 10^{-6}$ mg/day, $1.2 \times 10^{-4}$ to $6.0 \times 10^{-4}$ mg/day, $5.6 \times 10^{-3}$ to $1.6 \times 10^{-2}$ mg/day, $1.4 \times 10^{-4}$ to $1.7 \times 10^{-4}$ mg/day, and $1.7 \times 10^{-3}$ to $6.1 \times 10^{-3}$ mg/day, respectively while the maximum daily allowances for above elements are 0.021, 0.2, 2–5, 0.21 and 60 mg/day [48, 49]. Therefore, the ADI values of the elements for all studied medicinal plants were found less than the MTDI values.

### Target hazard quotient values

The target hazard quotient values for the As, Cd, Cr, Mn, and Zn are given in Table 4. It is observed that THQ values for the elements are below 1.0. The total target hazard quotient values for the considered elements in *A. ilicifolius*, *A. officinalis*, and *X. mekongensis* samples were found to be $3.5 \times 10^{-1}$, $1.6 \times 10^{-1}$, and $2.8 \times 10^{-1}$, respectively (Table 4).

**Table 4** Target hazard quotient (THQ) and target carcinogenic risk (TCR) values of the elements for consuming medicinal plants

| Target hazard quotient (THQ) | | | | Target carcinogenic risk (TCR) | | |
|---|---|---|---|---|---|---|
| Elements | *A. ilicifolius* | *A. officinalis* | *X. mekongensis* | *A. ilicifolius* | *A. officinalis* | *X. mekongensis* |
| As | $9.1\times10^{-3}$ | $2.2\times10^{-2}$ | $7.2\times10^{-3}$ | $4.1\times10^{-6}$ | $9.7\times10^{-6}$ | $3.2\times10^{-6}$ |
| Cd | $3.9\times10^{-2}$ | $3.2\times10^{-2}$ | $3.0\times10^{-2}$ | – | – | – |
| Cr | $1.7\times10^{-1}$ | $4.1\times10^{-2}$ | $2.0\times10^{-1}$ | $2.5\times10^{-4}$ | $6.2\times10^{-5}$ | $3.0\times10^{-4}$ |
| Mn | $1.1\times10^{-1}$ | $5.8\times10^{-2}$ | $4.0\times10^{-2}$ | | | |
| Zn | $2.1\times10^{-2}$ | $9.4\times10^{-3}$ | $5.6\times10^{-3}$ | | | |
| Total | $3.5\times10^{-1}$ | $1.6\times10^{-1}$ | $2.8\times10^{-1}$ | | | |

In the present study, the total THQ value for every medicinal plant was found below 1.0, which indicates that no non-carcinogenic risk is associated to consume these medicinal plants [49, 50].

### Target carcinogenic risk values

The TCR values for As and Cr in *A. ilicifolius*, *A. officinalis*, and *X. mekongensis* samples were calculated and also given in Table 4. According to the latest regional screening level (RSL) Tables of the US-Environmental Protection Agency (EPA) [34], among the considered elements only these two elements have available CSF values. It should be mentioned that calculations of THQ and TCR values for As and Cr were done based on RfD and CSF values of inorganic As and Cr(VI) contents because of the unavailability of these values for total As and Cr contents in the latest RSL tables of EPA. So, THQ and TCR values calculated in this study may be overestimated. The range of TCR values for As and Cr of *A. ilicifolius*, *A. officinalis*, and *X. mekongensis* were $3.2\times10^{-6}$ to $9.7\times10^{-6}$ and $6.2\times10^{-5}$ to $3.0\times10^{-4}$, respectively. If the TCR value is above $1\times10^{-6}$, it indicates the carcinogenic risk associated with the consumption of these medicinal plants [48, 51]. It is observed that the estimated TCR values of As and Cr for the studied medicinal plants were found above $1.0\times10^{-6}$. This indicates some extent of carcinogenic risk to consume these medicinal plants by people. However, the carcinogenic risk also depends on different factors like exposure frequency, duration, and amounts of consumption of medicinal plants.

### Transfer factor and pollution index values

The capability of a plant species to transfer metal from the soil to the plant is indicated as a transfer factor (TF). The factors are considered on the root uptake of the metal and reduce the foliar absorption of atmospheric metal deposits. Higher TF values ($\geq 1$) indicate higher metal absorption from the soil by plants that can cause harm to human health due to the consumption of the plants. On the contrary, a TF value below 1 indicates a poor plant response to metal absorption [52]. The estimated TF value is given in Table 5.

**Table 5** Transfer factor for medicinal plant samples *A. ilicifolius*, *A. officinalis*, and *X. mekongensis*

| Elements | *A. ilicifolius* | *A. officinalis* | *X. mekongensis* |
|---|---|---|---|
| Al | 0.021 | 0.002 | 0.021 |
| As | 0.002 | 0.004 | 0.002 |
| Br | 7.29 | 7.24 | 6.93 |
| Ca | 0.305 | 0.227 | 0.545 |
| Cd | 0.061 | 0.046 | 0.056 |
| Co | 0.027 | 0.035 | 0.029 |
| Cr | 0.040 | 0.008 | 0.042 |
| Fe | 0.030 | 0.003 | 0.019 |
| K | 0.624 | 0.425 | 0.679 |
| La | 0.036 | 0.008 | 0.037 |
| Mn | 0.141 | 0.001 | 0.040 |
| Na | 1.83 | 1.52 | 1.54 |
| Pb | 0.059 | 0.051 | 0.077 |
| Sb | 0.058 | 0.022 | 0.021 |
| Sc | 0.026 | 0.002 | 0.017 |
| Th | 0.034 | 0.002 | 0.031 |
| Zn | 0.457 | 0.186 | 0.125 |

Concentrations of Ce, Sm, and V in plant leaves were below the detection limits of INAA

In this study, the TF values obtained for three medicinal plants were 0.002–0.021 (Al), 0.002–0.004 (As), 6.93–7.29 (Br), 0.277–0.545 (Ca), 0.046–0.061 (Cd), 0.027–0.035 (Co), 0.008–0.042 (Cr), 0.003–0.030 (Fe), 0.425–0.679 (K), 0.008–0.037 (La), 0.001–0.141 (Mn), 1.52–1.83 (Na), 0.051–0.077 (Pb), 0.021–0.058 (Sb), 0.002–0.026 (Sc), 0.002–0.034 (Th) and 0.125–0.457 (Zn), respectively. The TF values for Br and Na were higher than 1.0 (Table 5) indicating the absorptions of Br and Na were higher than the other elements for studied medicinal plants. Besides this, the TF values obtained for all heavy metals were below 1.0, which implied the low absorption capacity of the heavy metals by the medicinal plants. It is also observed that for an element, transfer factors for different medicinal plant species were different.

The safety level consideration by using PI value was divided into five categories: safe ($PI \leq 0.7$), guard line

(0.7 < PI < 1), mild contamination (1 < PI < 2), moderate contamination (2 < PI < 3), and serious contamination (PI ≥ 3) [53]. The PI values obtained in this study are shown in Table 6. For *A. officinalis*, the PI values of As, Cd, Cr, Pb, and Zn were below the safety level. On the other hand, for *X. mekongensis*, the PI value of Cr was 2.0 which indicates mild contamination whereas PI for other elements is within safe limits. Besides this, for *A. ilicifolius*, the value of PI for As, Cd, and Pb was at a secure level but for Zn, it was in the guard line, and for Cr, it was at a mild contamination level.

## Statistical analysis

Pearson correlation matrix was calculated to study the inter-metal relationships in the medicinal plants of the Sundarban and tabulated in Tables S3–S5. For *A. ilicifolius* and *X. mekongensis* plants, it is observed that Al has strong positive correlations with most of the elements, and for *A. ilicifolius* plant Fe has also strong positive correlations with most of the metals (Table S3). This can be attributed that Al and Fe-oxides in the plant soils are one of the main factors that affect plant uptake of trace elements from soils [54]. Zinc has negative or poor correlations with every metal at *A. officinalis* (except Co and Mn) whereas, at *X. mekongensis*, it has strong positive correlations with most metals. This indicates the complex correlation nature of the elements in different species of medicinal plants [55]. From Table S3–S5, it is also observed that for *A. ilicifolius and A. officinalis*, As has a negative or poor correlation with the majority of the elements which may show the different sources of this metal in Sundarban mangrove soils and also in plants. The major element Ca exhibits negative or poor correlations with most of the metals at *A. ilicifolius and A. officinalis* which indicates that they can result from diverse sources of contamination of the Sundarban soils. For *A. officinalis* significant positive correlation between Cd and Pb (r = 0.99, $P < 0.1$) points to their geochemical similarities (chalcophile behavior) in the soil as well as their similar transfer factors (discussed in the previous section) for the medicinal plants.

**Table 6** Pollution index value for the toxic metals in medicinal plants

| Elements | *A. ilicifolius* | *A. officinalis* | *X. mekongensis* |
|---|---|---|---|
| As | *0.002* | *0.004* | *0.001* |
| Cd | 0.13 | 0.10 | 0.10 |
| Cr | 1.7 | 0.41 | 2.0 |
| Pb | 0.094 | 0.10 | 0.11 |
| Zn | 0.82 | 0.37 | 0.22 |

## Conclusions

In this study, total concentrations of 20 chemical elements (Al, As, Br, Ca, Cd, Ce, Co, Cr, Fe, K, La, Mn, Na, Pb, Sc, Sb, Sm, Th, V, and Zn) were determined by INAA and AAS techniques. This study indicates that Cr concentrations were slightly higher than WHO/FAO tolerable levels in *A. ilicifolius, and X. mekongensis*. However, average daily intake (ADI) values compared with MTDI and target hazard quotient (THQ) values were within the safety levels. On the other hand, calculated TCR values and PI values of Cr for *A. ilicifolius, and X. mekongensis* plants were slightly higher than the risk level which reveals some extent of risk to consumers if consumed for a long time. Although these medicinal plants might be considered a good source of Ca, K, and Zn, the consumption rate of those plants must be monitored and strictly controlled by the proper authority to avoid potential toxicity to the consumers.

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## Declarations

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

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