



Ecological–Health Risk of Antimony and Arsenic in *Centella asiatica*, Topsoils, and Mangrove Sediments: A Case Study of Peninsular Malaysia

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The current study assessed the ecological–health risks of potentially toxic arsenic (As) and antimony (Sb) in the vegetable *Centella asiatica*, topsoils, and mangrove sediments sampled from Peninsular Malaysia. The As concentrations ranged from 0.21 to 4.33, 0.18 to 1.83, and 1.32 to 20.8 mg/kg dry weight, for the leaves, stems, and roots of the vegetable, respectively. The ranges of Sb concentrations were 0.31–0.62, 0.12–0.35, and 0.64–1.61 mg/kg dry weight, for leaves, stems, and roots of the vegetable, respectively. The children’s target hazard quotient (THQ) values indicated no non-carcinogenic risks of As and Sb in both leaves and stems, although children’s THQ values were higher than those in adults. The calculated values of estimated weekly intake were lower than established provisional tolerable weekly intake of As and Sb for both children and adult consumers. The carcinogenic risk (CR) values of As for children’s intake of leaves and stems of vegetables showed more public concern than those of adults. The levels of Sb and As in the topsoils were generally higher (although not significantly) than those in the mangrove sediments, resulting in a higher geoaccumulation index, contamination factor and ecological risk, hazard index, THQ, and CR values. This indicated that the

anthropogenic sources of Sb and As originated from the land-based activities before reaching the mangrove near the coast. The CR of As signifies a dire need for comprehensive ecological–health risks exposure studies, as dietary intake involves more than just vegetable consumption. Therefore, risk management for As and Sb in Malaysia is highly recommended. The present findings of the ecological–health risks of As and Sb based on 2010–2012 samples can be used as an important baseline for future reference and comparison.

Keywords: ecological risk, health risk, arsenic and antimony, soil and sediment pollution, Peninsular Malaysia

INTRODUCTION

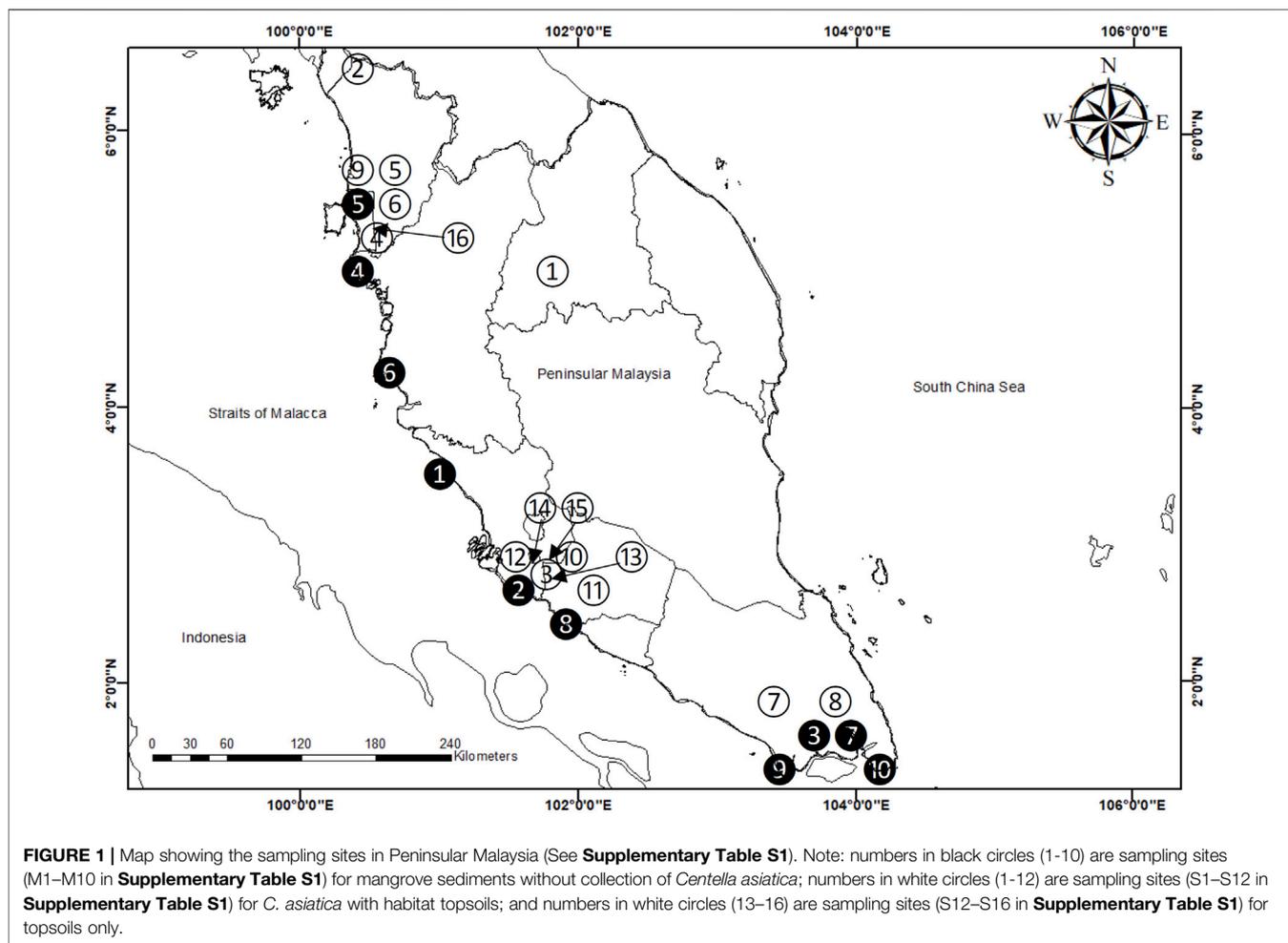
Reports on antimony (Sb) and arsenic (As) sources in Malaysia are scarce. In the central area of Sarawak, for example, Sb mineralization was found along the Lalang Fault Zone (Yeap et al., 1996), which also marks the boundary between the Upper Cretaceous Layar Member and the Eocene to Paleocene Kapit Member, both of which are part of the Belaga Formation. Pereira et al. (1994) also reported the use of soil geochemistry in the Buffalo Reef area, Kuala Medang, Pahang, to find Sb–Au mineralization. Recently, the concentrations of mineral-forming and trace elements including As and Sb were examined by Sia and Abdullah, (2012) in coal and coal ashes of the Balingian coalfield in Sarawak, Malaysia. However, the ecological and health effects of Sb and As in Malaysian soils were not addressed in the aforementioned studies.

In general, potentially toxic metals in the environmental media have received substantial concerns due to human health risk potentials (Egbueri et al., 2019; Ukah et al., 2019; Egbueri et al., 2020; Omeka et al., 2022). Murphy et al. (1989) developed risk assessment methodologies for two chronic As-contaminated soil exposure pathways: inhalation of fugitive dust emissions over a lifetime and unintentional soil/house dust intake. There is a wealth of knowledge about inorganic As oral toxicity, and several effects have been recorded, both non-carcinogenic and carcinogenic in nature. Fatigue, gastrointestinal complaints, irregular heart rhythm, bruises, and reduced nerve function can all occur due to ingestion (ATSDR, 2007). Other non-carcinogenic toxic effects due to As exposure include peripheral vascular diseases such as cyanosis, gangrene, and a condition known as “blackfoot disease” (Tseng et al., 2007). Oral ingestion of inorganic As has also been associated with an increased risk of cancer in the skin, liver, bladder, and lungs (Mandal, 2017). Bhattacharya et al. (2007) undertook a complete assessment of As in the environment, emphasizing the necessity for remediation of As-polluted groundwater, considering the carcinogenic toxicological effects on human health. According to Zhang et al. (2020), the combined effect of As in the atmosphere and groundwater may greatly worsen both the carcinogenic and non-carcinogenic effects associated with As pollution.

Sb is regarded as a valuable metal due to its vast range of applications, including catalysts, flameproof materials, ammunition, and bearings (Smichowski 2008; Reimann et al., 2010; Li et al., 2018; Wang et al., 2018). According to Zhou et al.

(2015), based on projection scenarios employing the worldwide Sb emission inventory for 1995–2010, the global Sb emissions are expected to grow by a factor of two between 2010 and 2050. Sb is now recognized as a global pollutant, and it has recently sparked global concern (Muhammad et al., 2021; Chang et al., 2022). Sb has gradually become one of the prime hazardous elements of environmental interest due to anthropogenic activities’ elevated input (Kawamoto and Morisawa, 2003; Casado et al., 2007; Qi et al., 2011a). Environmental concerns of Sb in soils have been identified in Chinese coal mines (Qi et al., 2011a). *In vitro* and *In vivo* Sb, like As, is known to be a genotoxic element. However, little is known about the transfer of Sb from the environment to humans and its health risk (Wu et al., 2011; Muhammad et al., 2021). In the Yodo River basin, Kawamoto and Morisawa, (2003) investigated the distribution of Sb in the river water environment, which included water, sediment, aquatic vegetation, and fish. They found that the biota in the Yodo River basin had elevated Sb levels. Sb pollution in soils is a significant environmental issue, and it is critical to understand how Sb migrates and transforms in soils (Mu et al., 2022). To acquire a better picture of the environmental risk caused by Sb, Serafimovska et al. (2013) investigated the distribution of Sb and its species in soil fractions. According to Mu et al. (2022), Sb is geochemically stable in soil due to its high presence in the residue fractions.

Wilson et al. (2010) reviewed the environmental chemistry of inorganic Sb in soils and compared and contrasted their findings with those of As. Sb is a hazardous trace element primarily linked to As (Shtangeeva et al., 2011). Due to its greater mobility in the soil than Pb, this metal is thought to provide a greater long-term risk than Pb (Tschan et al., 2010). Nevertheless, knowledge concerning the biogeochemical properties of Sb is relatively limited compared to other frequent hazardous elements (Shtangeeva et al., 2011). Although Sb and As are generally comparable in biogeochemical behavior; discrepancies in regulating variables impacting their mobility and bioavailability in soils were evident and warrant further investigation (Chang et al., 2022). In some mineralized soils, Sb can be found in high amounts. Sb’s action in plants is poorly understood (Hajiani et al., 2015). Casado et al. (2007) investigated the extent of Sb and As pollution, their bioavailability in mining-affected grassland soils, and their concentrations in plant aboveground sections. Humans can be exposed to excessive levels of As and Sb via ingestion of polluted soil, dust, or food plants, which leads to serious health risks (Clemente, 2013; Muhammad et al., 2021). Furthermore, the food chain is a



primary route for harmful substances to enter living creatures (Dubey et al., 2014). Tschan et al. (2009a) demonstrated that plants might absorb a considerable amount of Sb while appearing healthy.

Despite the growing concern about the ecological–health risks of As and Sb found in edible plants and mineralized topsoils, there is an acute lack of baseline knowledge regarding this aspect in Malaysia, or elsewhere in the Southeast Asia region. The vegetable plant *Centella asiatica* was selected given its reputation for being a good biomonitor of metals (Rainbow and Phillips, 1993; Ong, 2011; Ong, 2013). The aim of this study, therefore, was to assess the ecological–health risks of As and Sb in *C. asiatica*, topsoils, and mangrove sediments from Peninsular Malaysia.

2 MATERIALS AND METHODS

2.1 Study Area Description

In the present study, the mangrove sediments were collected from 10 sampling sites along the west coast of Peninsular Malaysia. These sites are flanked by a coastal fringe of

mangrove forest, primarily in the vicinity of the residential area, hydroelectric power plant, fishing villages, shipping areas, tourism spots, fish and shrimp farms, and industrial area. The sampling seasons ranged from March to June 2012, between the seasons of the first inter-monsoonal period and the Southwest monsoon (Britannica, 2022). *C. asiatica* plants, together with their habitat topsoils were collected from 12 sampling sites in Peninsular Malaysia. These sites are located near the residential area, agricultural areas such as oil palm plantation, industrial areas, highways, seaports, shop lots, and roadsides. The sampling seasons ranged from May to June 2010 during the Southwest monsoon season (Britannica, 2022).

2.2 Sampling

Samples of the plant *C. asiatica* and their habitat topsoils were collected between March and June 2010, while the mangrove sediments were collected from the west coast between March and June 2012, from Peninsular Malaysia (**Figure 1**; **Supplementary Table S1**). Plant samples were collected from three parts: leaves, stems, and roots, while soils and sediments of the top 0–5 cm layer were collected.

2.3 Neutron Activation Analysis

The leaves, stems, and roots from five plants were pooled for the analysis of As and Sb. Plants, topsoils, and mangrove sediment samples were dried at 65°C until the constant dry weight was achieved (approximately 5 days). Homogeneity of the dried samples was achieved by grinding them into a powder using an electronic agate homogenizer. The homogenous powder (0.15–0.20 g) of samples was manually shaken and sieved with a 63- μm sieve placed into polyethylene vials. These polyethylene vials were heat-sealed until analysis.

Sb and As were analyzed using the TRIGA MARK II reactor at the Malaysian Nuclear Agency, Bangi, Selangor, Malaysia. Neutron flux of $4\text{--}5 \times 10^{12}$ n/cm² was used for long irradiation of Sb as its half-life is 60.9 days. Following the irradiation, the samples were allowed to cool for a while using closed-end coaxial high purity germanium detectors (Model GC3018 CANBERRA Inc and Model GMX 20180, EG4G ORTEC Nuclear Instrument) and its related electronics. Counting of cooling time ranged from 3 to 6 days, and the lifetime counting for Sb was 3,600 s (IAEA-TECDOC-1360, 2003).

For quality control and quality assurance, acid-washed apparatus was used to avoid external contamination. The Certified Reference Material (CRM) IAEA-SOIL-7 was prepared under identical conditions and used as quality control for each batch. The CRM used for this study was IAEA-SOIL-7 for quality control purposes where the recovery of Sb was 73.58% (certified: 1.70 ± 0.09 mg/kg dry weight; measured: 1.25 ± 0.27 mg/kg dry weight), while the As recovery was 89.25% (certified: 13.40 ± 0.67 mg/kg dry weight; measured: 11.96 ± 2.16 mg/kg dry weight). To achieve greater sensitivity, precision, and accuracy of the analysis, we used a detection limit of 0.001 mg/g for As and Sb by NAA.

2.4 Data Treatment

2.4.1 Geoaccumulation Index

The geoaccumulation index (I_{geo}) is determined by using the following formula (Eq. (1)) (Muller, 1969):

$$I_{\text{geo}} = \text{Log}10\left(\frac{C_n}{1.5 \times B_n}\right) \quad (1)$$

where

C_n = concentration of element;

B_n = concentrations of background reference for Sb (0.31 mg/kg dry weight) and As (2.00 mg/kg dry weight) by Wedepohl (1995);

Factor 1.5 = background matrix correction factor due to lithogenic effects. The pollution intensities of I_{geo} were based on Muller (1969): “unpolluted” (<0); “unpolluted to moderately polluted” (0–1); “moderately polluted” (1–2); “moderately to strongly polluted” (2–3); “strongly polluted” (3–4); “strongly to very strongly polluted” (4–5); and “very strongly polluted” (>5).

2.4.2 Ecological Risk Assessment of Topsoils and Mangrove Sediments

Ecological risk (ER) of the individual element is an informative index reflecting the effects and toxicity of an element to the

environment (Hakanson, 1980; Wang et al., 2018). First, the contamination factor (CF) is calculated as in Eq. 2 (Hakanson, 1980):

$$\text{CF} = \frac{C_s}{C_n} \quad (2)$$

where CF = contamination factor of the element; C_s = the investigated metals in the soils or sediments; C_n = the background values for Sb and As by Wedepohl (1995).

Later, the ER is defined as the formula shown in Eq. 3 (Hakanson, 1980):

$$\text{ER} = \text{Tr} \times \text{CF} \quad (3)$$

The toxic coefficient (Tr) for As was calculated as 10 based on Hakanson (1980). Since the Tr for Sb was not provided by Hakanson (1980), the Sb Tr was adopted from Wang et al. (2018) (7.0 mg/kg), which was calculated based on Hakanson’s principles.

Classifications based on Hakanson (1980) for CF are: “low contamination” ($\text{CF} < 1$); “mild contamination” ($1 \leq \text{CF} < 3$); “considerable contamination” ($3 \leq \text{CF} < 6$); and “high contamination” ($\text{CF} \geq 6$). Classifications of the values of ER are (Hakanson 1980): “minimal potential ecological risk” ($\text{ER} < 40$); “mild potential ecological risk” ($40 \leq \text{ER} < 80$); “considerable potential ecological risk” ($80 \leq \text{ER} < 160$); “high potential ecological risk” ($160 \leq \text{ER} < 320$); and “very high ecological risk” ($\text{ER} \geq 320$).

2.5 Human Health Risk Assessments of *Centella asiatica*

For human health risk assessment (HHRA), the conversion factor (0.10) was applied to convert the dry weight (DW)-based data of As and Sb in leaves and stems of *C. asiatica* into wet weight (WW)-based data. To estimate the HHRA, three assessments were carried out, namely:

(a) Target hazard quotient

The estimated daily intake (EDI) and target hazard quotient (THQ) of elements were calculated to evaluate the once or long-term possible hazardous exposure to metals from eating edible leaves and stems of *C. asiatica* (USEPA, 1989) by the Peninsular Malaysian population. First, the EDI is calculated using the formula (Eq. 4):

$$\text{EDI} = \frac{\text{Mc} \times \text{consumption rate}}{\text{body weight}} \quad (4)$$

where Mc is the concentrations of the element (mg/kg wet weight; after converted from dry weight-based data) in leaves and stems of *C. asiatica*. The body weights were 17 kg and 69.2 kg for children and adults, respectively (Nurul Izzah et al., 2012; Chong et al., 2017), while the consumption rates were 17.0 and 34.0 g/day for children and adults, respectively (Chong et al., 2017).

Second, the THQ is calculated based on Eq. 5:

$$\text{THQ} = \frac{\text{EDI}}{\text{RfD}} \quad (5)$$

The oral reference portion (RfD) was compared with the EDIs ($\mu\text{g}/\text{kg}$ wet weight/day) of metals in vegetables. The RfD ($\mu\text{g}/\text{kg}$ wet weight/day) values used in this study were based on USEPA regional screening level (USEPA, 2022) for Sb (0.40) and As (0.30).

(b) Comparisons between estimated weekly intake and provisional tolerable weekly intake

The Joint FAO/WHO Expert Committee on Food Additives (JECFA) considered As at its 33rd meeting (FAO/WHO, 1989) and confirmed its earlier evaluation by establishing a provisional tolerable weekly intake (PTWI) of $15 \mu\text{g}/\text{kg}$ body weight. Therefore, the present study used the PTWI of $15 \mu\text{g}/\text{kg}$ body weight for As (FAO/WHO, 1989). Thus, the As PTWI values for 17 and 69.2 kg body weights for children and adults in Malaysia are equivalent to 255 and $1,038 \mu\text{g}/\text{week}$, respectively.

For Sb, based on the WHO (2017) and ATSDR (2019), which was initially based on drinking water quality guidelines ($0.02 \text{ mg}/\text{L}$) for Sb and compounds (WHO, 2017), the tolerable daily intake (TDI) for Sb was recommended as $6 \mu\text{g}/\text{kg}/\text{body weight}$. Therefore, 7 days of Sb TDI was calculated as $42 \mu\text{g}/\text{kg}/\text{body weight}$. An adult with a body weight of 69.2 kg would have a PTWI of $2,906 \mu\text{g}/\text{week}$, while a child with a body weight of 17 kg would have a PTWI of $714 \mu\text{g}/\text{week}$.

In order to evaluate the risk exposure from *C. asiatica* consumption, the estimated weekly intake (EWI) values of Sb and As are calculated as follows using Eq. 6:

$$\text{EWI} = \text{EDI} \times 7 \quad (6)$$

where $\text{EDI} = [\text{estimated daily intake calculated in Eq. 1}] \times 7$ (number of days in a week). The comparison between calculated EWI and established PTWI limits for children and adults will reveal if the EWI values are lower than the established PTWI values.

(c) Carcinogenic risk of As in *Centella asiatica*

The incremental probability of an individual developing cancer during a lifetime is determined by the cancer risks (CRs) for As. For example, a CR of 1.0×10^{-4} indicates a probability of 1 in 10,000 individuals developing cancer (USEPA, 1989). The CR of As in the leaves and stems of *C. asiatica* was calculated by multiplying the average daily intake of As (in $\mu\text{g}/\text{kg}/\text{day}$ over a lifetime) with a cancer slope factor (SF) according to Eq. (7).

$$\text{CR} = \left(\frac{\text{EDI}}{1000} \right) \times \text{SF} \quad (7)$$

Here, the SF actor for As after oral exposure has been set to $1.50 (\text{mg}/\text{kg}/\text{day})^{-1}$ by the USEPA (2022). According to USEPA methods, cancer risk lower than 1.0×10^{-6} is considered to be “negligible,” $>1.0 \times 10^{-4}$ is considered “unacceptable,” and in the range from 1.0×10^{-6} to 1.0×10^{-4} is considered “acceptable” (USEPA, 1989).

2.6 Human Health Risk Assessment of Topsoils and Sediments

(a) Non-carcinogenic risks of As and Sb

The non-carcinogenic risk (NCR) of As and Sb to humans was measured by HHRA of topsoils and sediments via three exposure pathways: ingestion, inhalation, and skin contact based on the guidelines and Exposure Factors Handbook of the US Environmental Protection Agency (USEPA, 1986, 1989, 1997, 2001). The average daily doses (ADDs) (mg/kg day) of As and Sb through ingestion (ADD_{ing}), inhalation (ADD_{inh}), and dermal contact (ADD_{der}) for both children and adults are calculated by using Eqs. 8–10 as follows:

$$\text{ADD}_{\text{ing}} = C_{\text{soil}} \left(\frac{\text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \right) \times 10^{-6} \quad (8)$$

$$\text{ADD}_{\text{inh}} = C_{\text{soil}} \left(\frac{\text{InghR} \times \text{EF} \times \text{ED}}{\text{PEF} \times \text{BW} \times \text{AT}} \right) \quad (9)$$

$$\text{ADD}_{\text{der}} = C_{\text{soil}} \left(\frac{\text{SA} \times \text{AF} \times \text{ABS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \right) \times 10^{-6} \quad (10)$$

where ADD_{ing} , ADD_{inh} , and ADD_{der} are the daily amounts of exposure to As and Sb (mg/kg day) through ingestion, inhalation, and dermal contact, respectively. In this study, NCR values of As and Sb were assessed by using the hazard quotient (HQ) and hazard index (HI) (Chabukdhara and Nema, 2013; Qing et al., 2015). The definition, exposure factors, and reference values used to estimate the intake values and health risks of As and Sb in topsoils and sediments are presented in **Supplementary Table S2**.

The HQ is the proportion of the ADD of a metal to its reference dose (RfD) for the three exposure pathway(s) (USEPA, 1989), as shown in Eq. 11:

$$\text{HQ} = \frac{\text{ADD}}{\text{RfD}} \quad (11)$$

The RfD (mg/kg day) is the maximum daily dose of As and Sb from three exposure pathways for both children and adults, which will not pose significant risks of adverse effects on sensitive individuals throughout the course of their lives. ADD values lower than the RfD value ($\text{HQ} \leq 1$) indicate no hazardous health effects, while higher ADD values than RfD values ($\text{HQ} > 1$) indicate potential hazardous health effects (USEPA, 1986, 2001). Despite substantial uncertainties, HQ values above 1 are regarded as an indicator of the potential risk of hazardous health effects to certain exposed individuals (USEPA, 1989).

(b) Carcinogenic risk of As in topsoils and sediments

The HHRA of topsoils and sediments was utilized to measure CR of As to humans by means of three exposure pathways, namely, ingestion, inhalation, and skin contact since the SF values were established for the three pathways for As but not for Sb (Cao et al., 2014; USEPA, 2022).

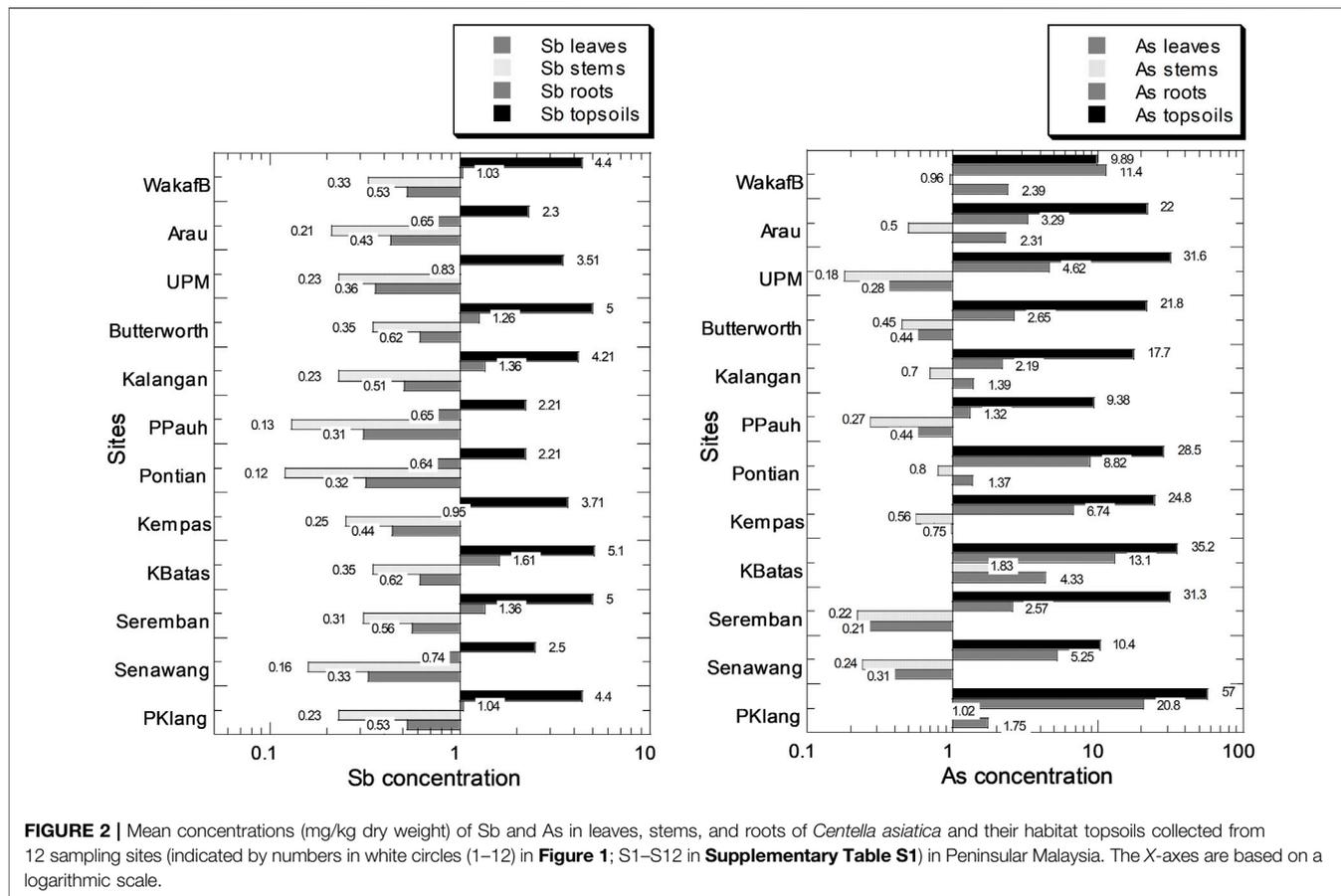


FIGURE 2 | Mean concentrations (mg/kg dry weight) of Sb and As in leaves, stems, and roots of *Centella asiatica* and their habitat topsoils collected from 12 sampling sites (indicated by numbers in white circles (1–12) in **Figure 1**; S1–S12 in **Supplementary Table S1**) in Peninsular Malaysia. The X-axes are based on a logarithmic scale.

The CR was calculated by multiplying the average daily intake of As (in $\mu\text{g}/\text{kg}/\text{day}$ over a lifetime) with an SF according to **Eq. 12**. The CR was estimated as the incremental probability of an individual developing cancer over a lifetime. For example, a CR of 1.0×10^{-4} indicates a probability of 1 in 10,000 individuals developing cancer (USEPA, 1989).

$$CR = \left(\frac{ADD}{1000} \right) \times SF \quad (12)$$

For both children and adults, the SF for As after oral exposure has been set to $1.5 \text{ (mg/kg/day)}^{-1}$ by the USEPA (USEPA, 1989; Cao et al., 2014; USEPA, 2022). For inhalation and dermal contact pathways, they were $15.1 \text{ (mg/kg/day)}^{-1}$ and $3.66 \text{ (mg/kg/day)}^{-1}$, respectively (Cao et al. (2014)). According to USEPA’s categories, a cancer risk lower than 1.0×10^{-6} is considered to be “negligible,” $>1.0 \times 10^{-4}$ is considered “unacceptable,” and in the range from 1.0×10^{-6} to 1.0×10^{-4} is considered “acceptable” (USEPA, 1989).

2.7 Statistical Analysis

KaleidaGraph (Version 3.08, Synergy Software, Eden Prairie, MN, United States) was utilized to obtain the overall statistics of the data and to create the graphical bar charts in this study.

3 RESULTS

3.1 Concentrations of Sb and As in *Centella asiatica* and Their Habitat Topsoils

Figure 2 and **Supplementary Table S3** show the levels of As and Sb. The As concentrations (mg/kg dry weight) ranged from 0.21 to 4.33, 0.18 to 1.83, and 1.32 to 20.8 for the leaves, stems, and roots of *C. asiatica*, respectively. The ranges of Sb concentrations (mg/kg dry weight) were 0.31–0.62, 0.12–0.35, and 0.64–1.61, for leaves, stems, and roots of *C. asiatica*, respectively. The As and Sb levels followed the order of roots > leaves > stems.

3.2 Health Risk Assessments of Sb and As in *Centella asiatica*

Figure 3 and **Supplementary Table S4** show the EDI values in the leaves and stem for both children and adults. The values of Sb EDI for children and adults in the leaves ranged from 3.10×10^{-2} to 6.20×10^{-2} and 1.50×10^{-3} to 3.10×10^{-2} , respectively. The values of Sb EDI in the stems ranged from 1.20×10^{-2} to 3.50×10^{-2} and 6.00×10^{-3} to 1.70×10^{-2} , for children and adults, respectively (**Supplementary Table S4**). The values of As EDI for children and adults in the leaves ranged from 2.10×10^{-2} to 4.33×10^{-1} and 1.00×10^{-2} to 2.13×10^{-1} , respectively. The values of As

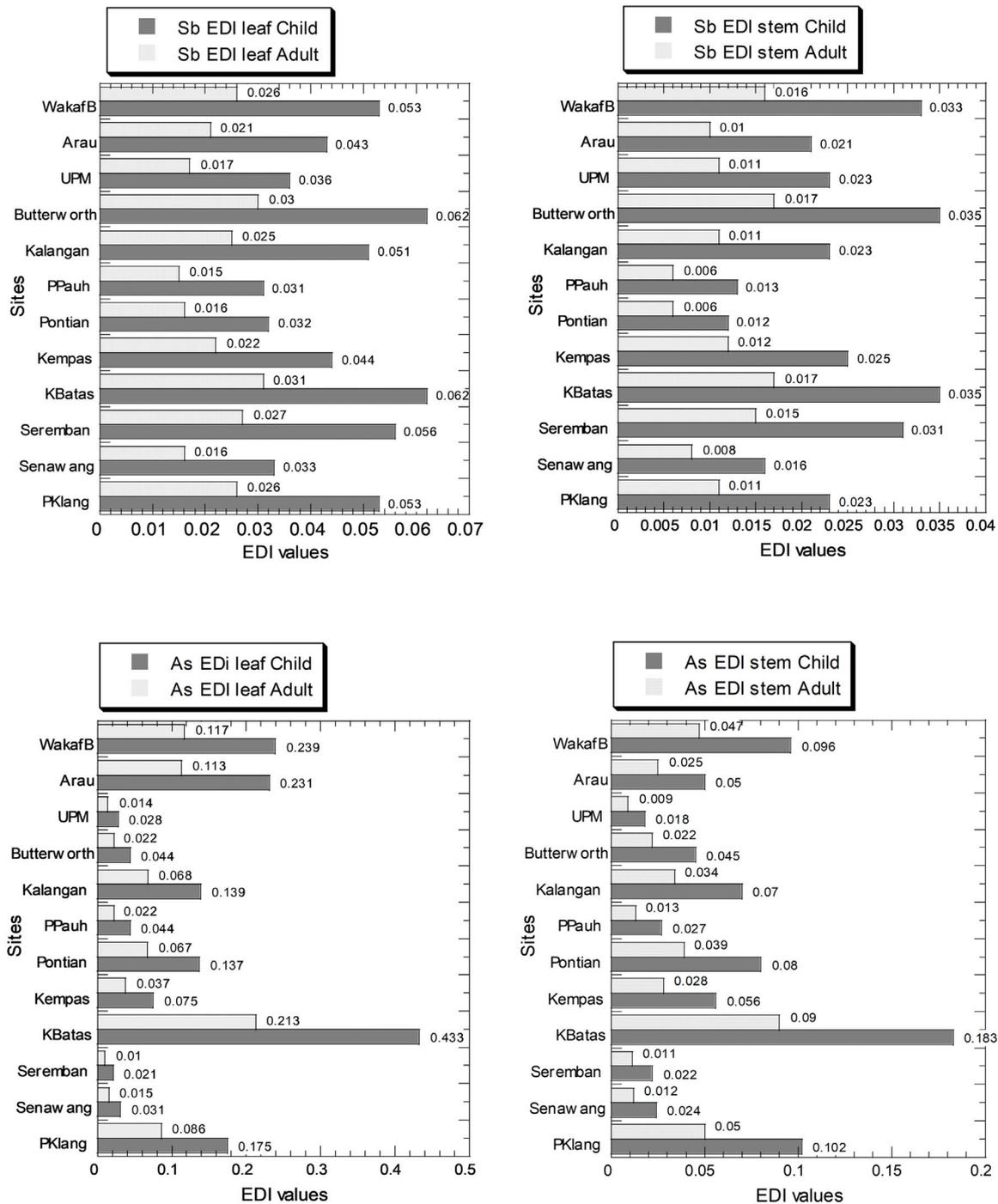


FIGURE 3 | Estimated dietary intake (EDI) values (µg/kg wet weight/day) of Sb and As in the leaves and stems of *Centella asiatica* collected from 12 sampling sites (numbers in white circles (1–12) in **Figure 1**; S1–S12 in **Supplementary Table S1**) in Peninsular Malaysia. The X-axes are based on a linear scale.

EDI in the stems ranged from 1.80×10^{-2} to 1.83×10^{-1} and 9.00×10^{-3} to 9.00×10^{-2} , for children and adults, respectively (**Supplementary Table S4**).

For Sb and As, the values of THQ in the leaves and stem in both children and adults are presented in **Figure 4**, and

Supplementary Table S5. The values of Sb THQ for children and adults in the leaves ranged from 7.90×10^{-2} to 1.55×10^{-1} and 3.90×10^{-2} to 7.60×10^{-2} , respectively. The values of Sb THQ for children and adults in the stems ranged from 2.90×10^{-2} to 8.80×10^{-2} and 1.40×10^{-2} to 4.30×10^{-2} ,

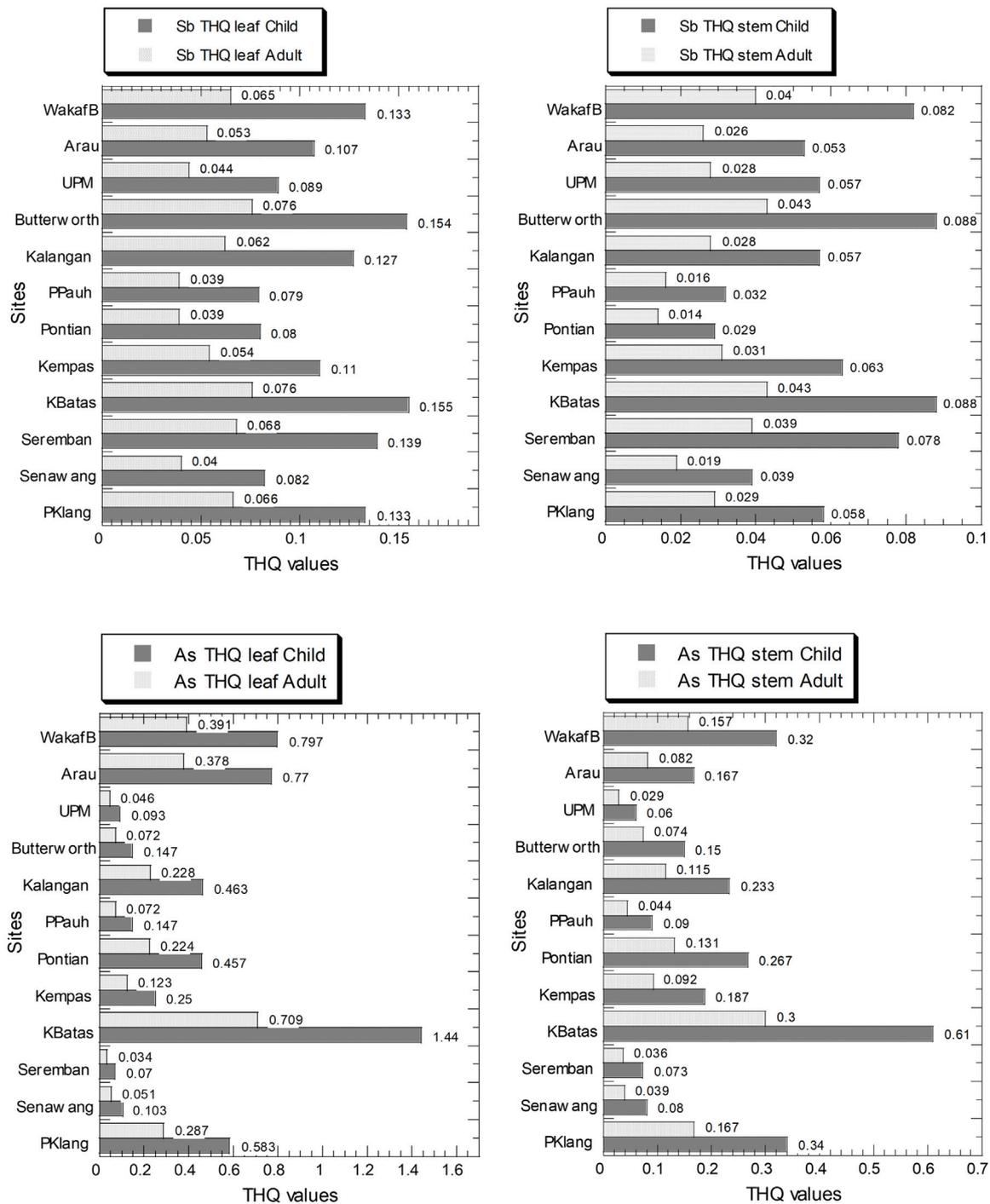


FIGURE 4 | Target hazard quotient (THQ) values (unitless) of As and Sb in the leaves and stems of *Centella asiatica* collected from 12 sampling sites (numbers in white circles (1–12) in **Figure 1**; S1–S12 in **Supplementary Table S1**) in Peninsular Malaysia. The X-axes are based on a linear scale.

respectively (**Supplementary Table S5**). The values of As THQ in the leaves ranged from 7.00×10^{-2} to 1.44 and 3.40×10^{-2} to 7.09×10^{-1} , for children and adults, respectively. The values of As THQ in the stems ranged from 6.00×10^{-2} to 6.10×10^{-1}

and 2.90×10^{-2} to 3.00×10^{-1} , for children and adults, respectively (**Supplementary Table S5**).

Two major patterns can be identified. First, the children’s THQ values in both leaves and stems were higher than those in

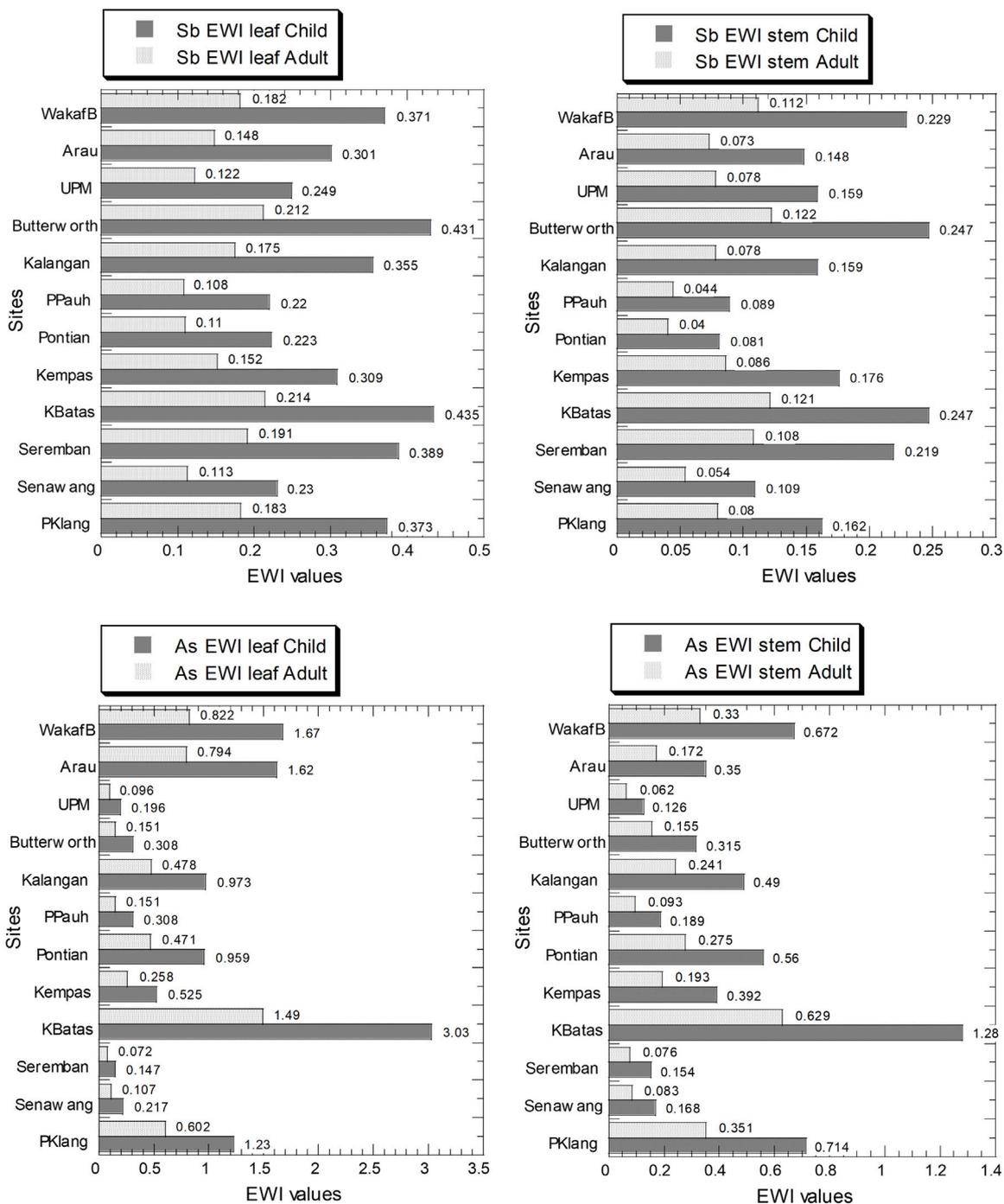


FIGURE 5 | Estimated weekly intake (EWI) values (µg/kg wet weight/week) of Sb and As in the leaves and stems of *Centella asiatica* collected from 12 sampling sites (numbers in white circles (1–12) in **Figure 1**; S1–S12 in **Supplementary Table S1**) in Peninsular Malaysia. The X-axes are based on a linear scale.

the adults. Second, all THQ values for Sb and As were below 1.0, except for As in children consuming the leaves collected from Kepala Batas (KBatas) in northwest Peninsular Malaysia.

Figure 5 and **Supplementary Table S6** show the values of EWI for Sb and As in the leaves and stems in both children and adults. The values of Sb EWI for children and adults in the leaves

ranged from 2.20×10^{-1} to 4.35×10^{-1} and 1.08×10^{-2} to 2.14×10^{-1} , respectively. The values of Sb EWI for children and adults in the stems ranged from 8.10×10^{-2} to 2.47×10^{-1} , and 4.00×10^{-2} to 1.22×10^{-1} , respectively (**Supplementary Table S6**). The Sb PTWI values for a 17 and 69.2 kg body weight for children and adults in Malaysia are equivalent to 714 and 2,906 µg/week,

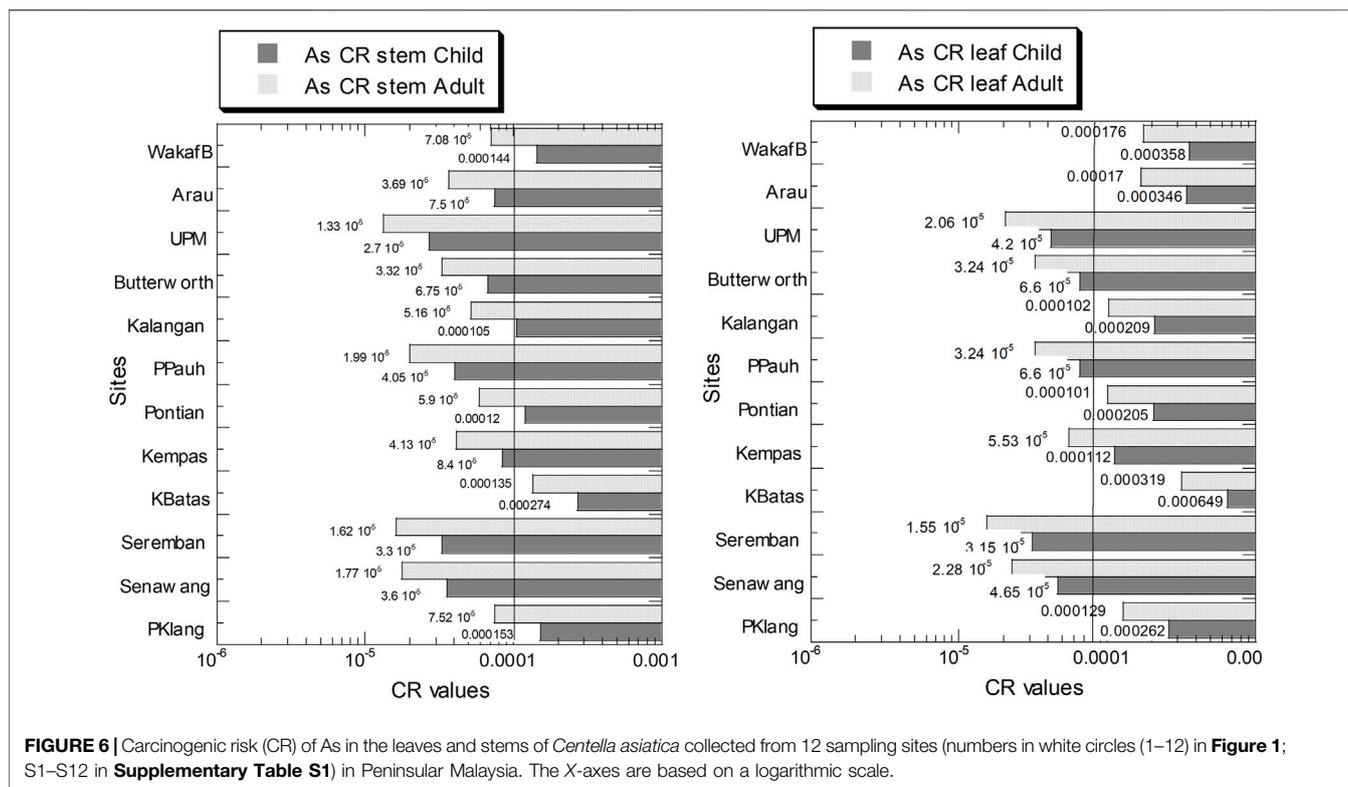


FIGURE 6 | Carcinogenic risk (CR) of As in the leaves and stems of *Centella asiatica* collected from 12 sampling sites (numbers in white circles (1–12) in **Figure 1**; S1–S12 in **Supplementary Table S1**) in Peninsular Malaysia. The X-axes are based on a logarithmic scale.

respectively. Therefore, the EWI values for children and adults in the leaves and stems of *C. asiatica* were significantly lower than those of PTWI values of Sb.

The values of As EWI for children and adults in the leaves ranged from 1.47×10^{-1} to 3.03, and 7.20×10^{-2} to 1.49, respectively. The values of As EWI for children and adults in the stems ranged from 1.26×10^{-1} to 1.28 and 6.20×10^{-2} to 6.29×10^{-1} , respectively (**Supplementary Table S6**). The As PTWI for a 17 and 69.2 kg body weight for children and adults in Malaysia are equivalent to 255 and 1,038 $\mu\text{g}/\text{week}$, respectively. Therefore, the EWI values for children and adults in the leaves and stems of *C. asiatica* were significantly lower than those of PTWI values of As.

For As, the values of CR in the leaves and stems in both children and adults were presented in **Figure 6** and **Supplementary Table S7**. The values of As CR for children and adults in the leaves ranged from 3.15×10^{-5} to 6.50×10^{-4} , and 1.55×10^{-5} to 3.19×10^{-4} , respectively. The values of As CR in the stems ranged from 2.70×10^{-5} to 2.74×10^{-4} , and 1.33×10^{-5} to 1.35×10^{-4} , for children and adults, respectively (**Supplementary Table S7**).

According to USEPA methods, cancer risk lower than 1.0×10^{-6} is considered to be “negligible,” $>1.0 \times 10^{-4}$ is considered “unacceptable,” and in the range from 1.0×10^{-6} to 1.0×10^{-4} is considered “acceptable” (USEPA, 1989). Based on **Figure 6**, the As CR values of all sites were considered “acceptable” except for the 5 sites (out of 10) in children consuming stems that exceeded 1.0×10^{-4} which is considered “unacceptable,” while only 1 site exceeded the threshold limit for adults in the

stem. For children consuming the leaves, six sites exceeded 1.0×10^{-4} which were considered “unacceptable” while five sites exceeded the threshold limit for adults in leaves. Therefore, these As CR values showed a public concern with 10–60% of the sampling sites exceeding the As CR threshold limit.

3.3 Concentrations of Sb and As and Ecological Risk Assessment in Topsoils and Mangrove Sediments

The levels of Sb and values of I_{geo} , Cf, and ER in both topsoils and mangrove sediments are presented in **Figure 7** and **Supplementary Table S8**. The Sb concentrations (mg/kg dry weight) ranged from 2.21 to 6.61 and 0.001 to 1.72, for topsoils and mangrove sediments, respectively. Overall, the average Sb level (4.00) in the topsoils was higher than 0.97 in the mangrove sediments.

The I_{geo} ranges (2.25–3.83) of Sb in the topsoils were categorized between “moderately to strongly polluted” and “strongly polluted” while those (-0.86–1.89) of the mangrove were all categorized as “unpolluted” and “moderately polluted” (Muller, 1969). The CF ranges (7.11–21.3) of Sb in the topsoils were all under the category of “high contamination” while those (0.003–5.55) of the mangrove were all categorized as “low contamination” and “considerable contamination” (Muller, 1969).

The ER ranges (49.8–149) of Sb in the topsoils fell between “mild potential ecological risk” and “considerable potential

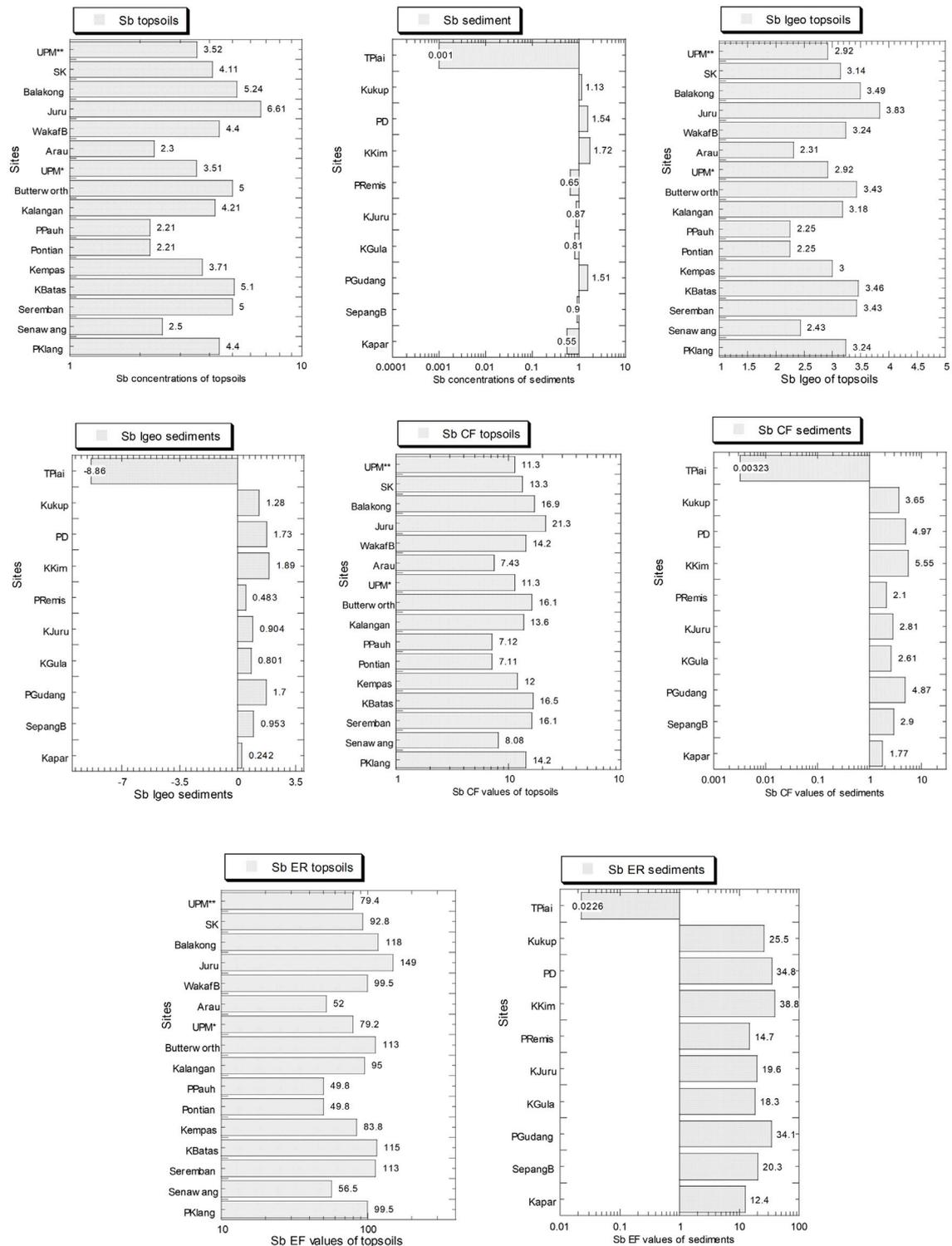


FIGURE 7 | Concentrations of Sb (mg/kg dry weight), geoaccumulation index (I_{geo}), contamination factor (CF), and potential ecological risk index (ER) for Sb in the topsoils (numbers in white circles (1–16) in **Figure 1**; S1–S16 in **Supplementary Table S1**) and mangrove sediments (numbers in black circles (1–10) in **Figure 1**; M1–M10 in **Supplementary Table S1**) from Peninsular Malaysia. All the X-axes are based on a logarithmic scale, except for the X-axes of I_{geo} values which are based on a linear scale.

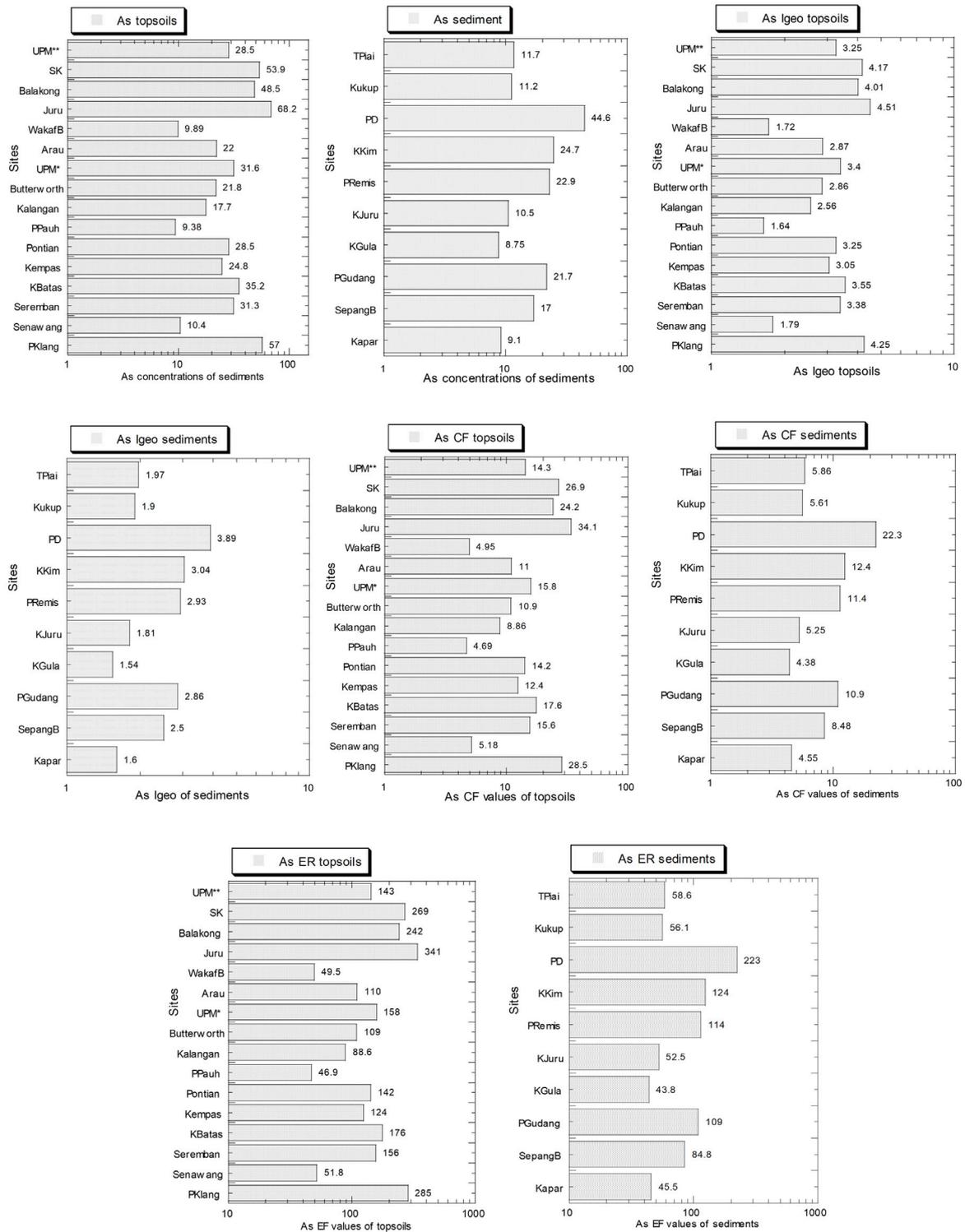


FIGURE 8 | Geoaccumulation index (I_{geo}), contamination factor (CF), and potential ecological risk index (ER) for As in the topsoils (numbers in white circles (1–16) in **Figure 1**; S1–S16 in **Supplementary Table S1**) and mangrove sediments (numbers in black circles (1–10) in **Figure 1**; M1–M10 in **Supplementary Table S1**) from Peninsular Malaysia. The X-axes are based on a logarithmic scale.

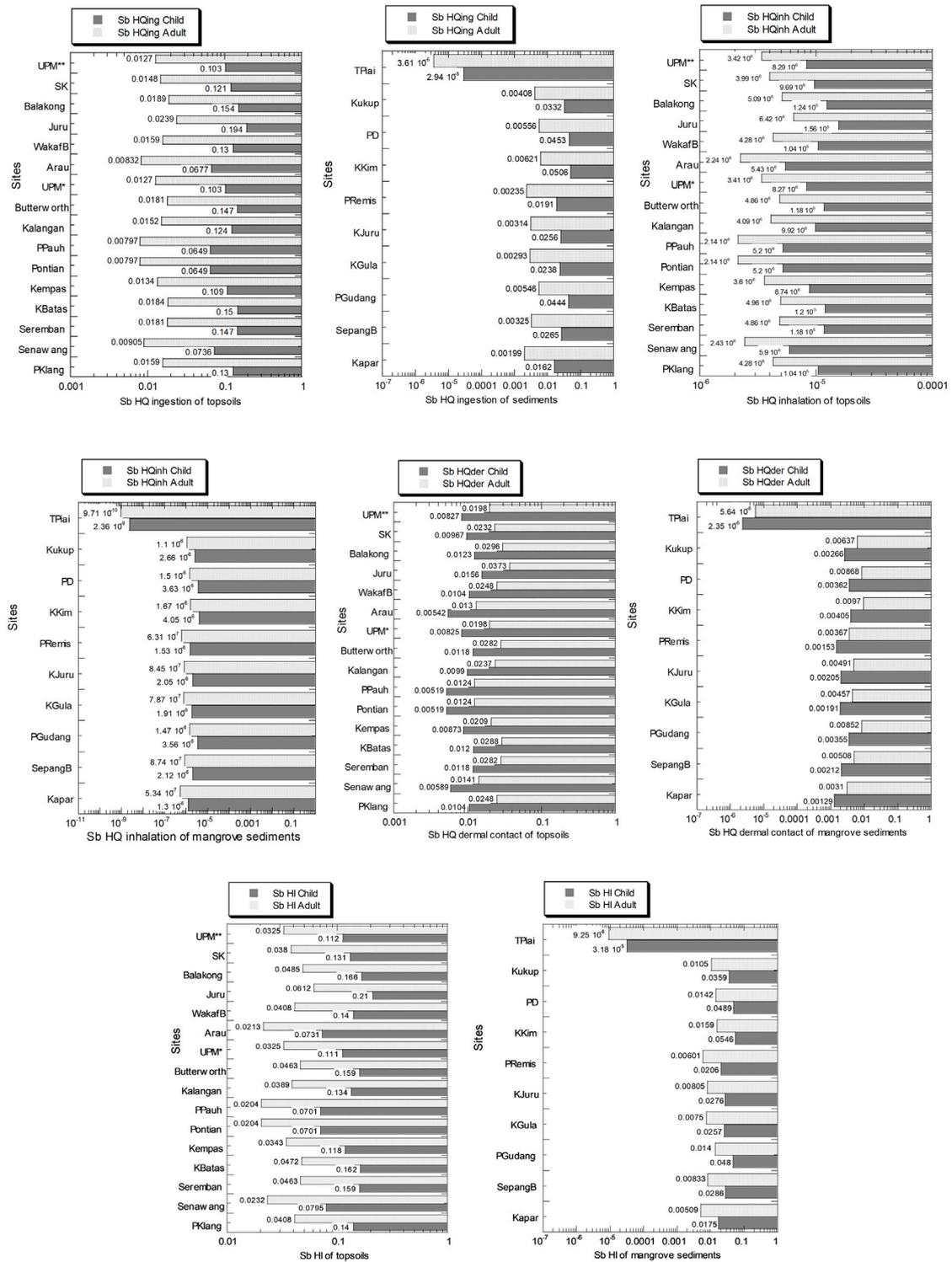


FIGURE 9 | Sb hazard quotient values of ingestion (HQ_{ing}), inhalation (HQ_{inh}), and dermal contact (HQ_{der}) pathways and overall hazard index (HI) in the soils (numbers in white circles (1–16) in **Figure 1**; S1–S16 in **Supplementary Table S1**) and mangrove sediments (numbers in black circles (1–10) in **Figure 1**; M1–M10 in **Supplementary Table S1**) from Peninsular Malaysia, in children and adults. The X-axes are based on a logarithmic scale.

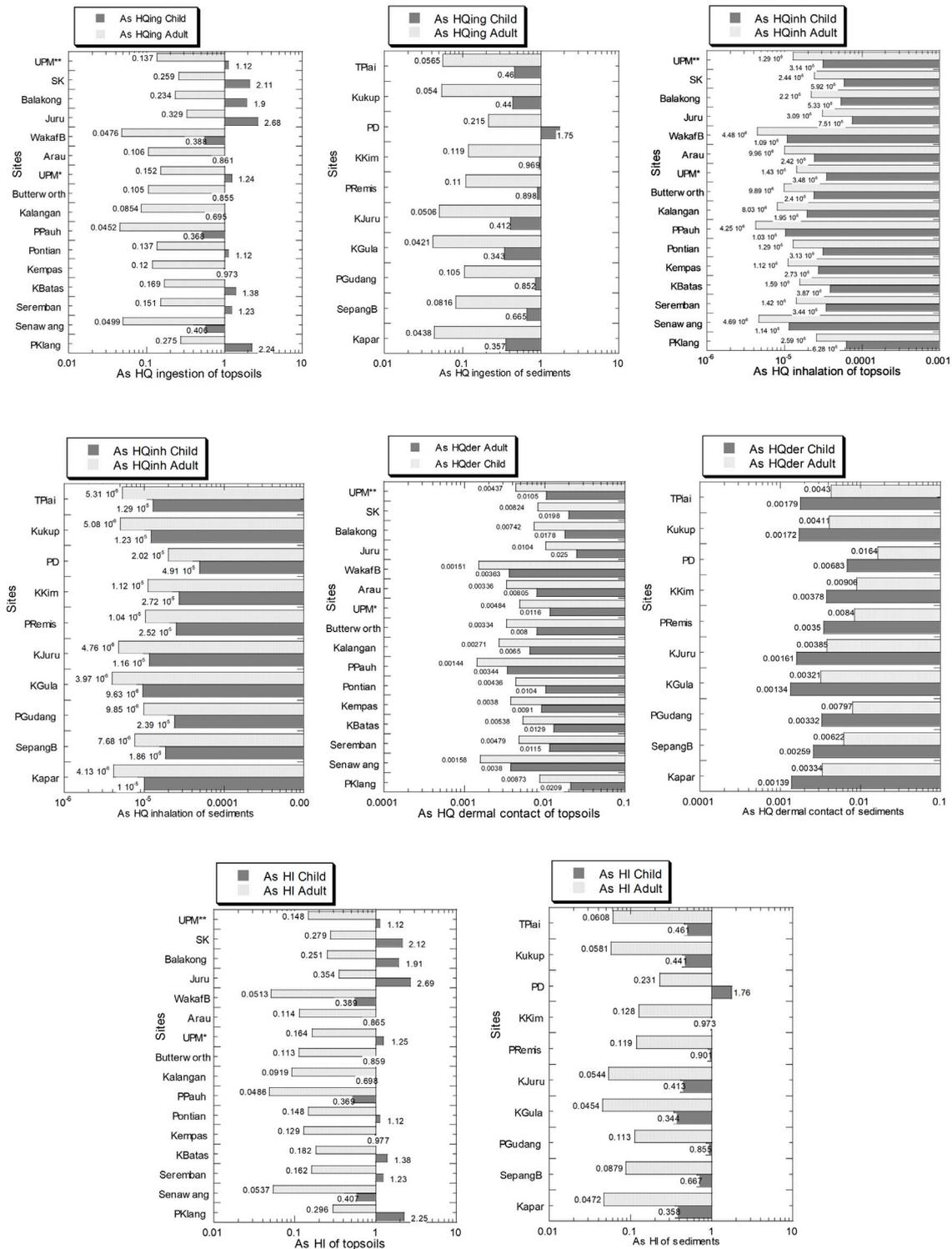


FIGURE 10 | As hazard quotient values of ingestion (HQ_{ing}), inhalation (HQ_{inh}), and dermal contact (HQ_{der}) pathways in the soils (numbers in white circles (1–16) in **Figure 1**; S1–S16 in **Supplementary Table S1**) and mangrove sediments (numbers in black circles (1–10) in **Figure 1**; M1–M10 in **Supplementary Table S1**) from Peninsular Malaysia, in children and adults. The X-axes are based on a logarithmic scale.

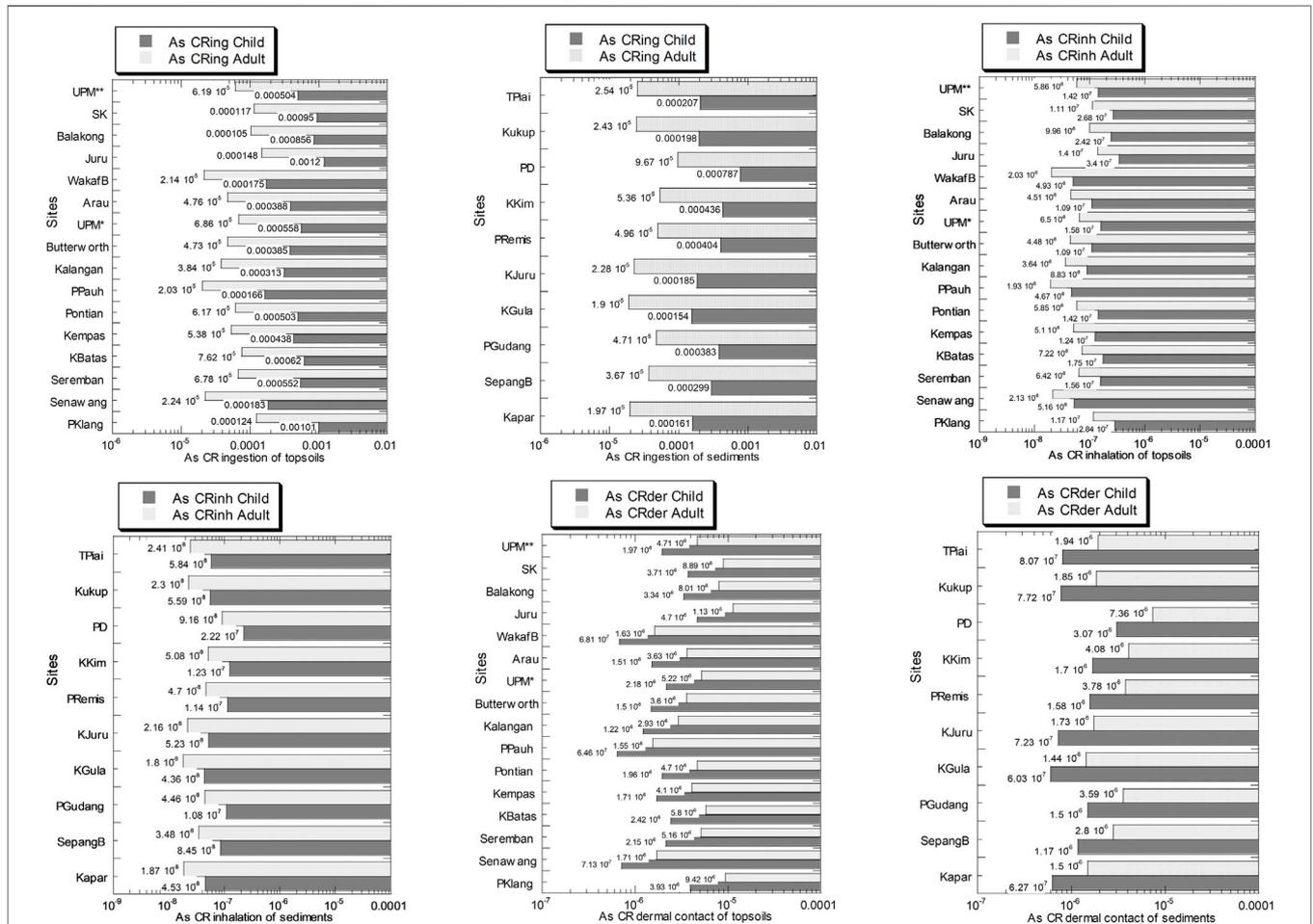


FIGURE 11 | As carcinogenic risk (CR) values of ingestion (CR_{ing}), inhalation (CR_{inh}), and dermal contact (CR_{der}) pathways in the soils (numbers in white circles (1–16) in **Figure 1**; S1–S16 in **Supplementary Table S1**) and mangrove sediments (numbers in black circles (1–10) in **Figure 1**; M1–M10 in **Supplementary Table S1**) from Peninsular Malaysia, in children and adults. The X-axes are based on a logarithmic scale.

ecological risk” while those (0.02–38.8) of the mangrove were all categorized as “minimal potential ecological risk” (Hakanson, 1980). Overall, the average Sb values of I_{geo} , CF, and ER in the topsoils were higher than those in the mangrove sediments.

Figure 8 and **Supplementary Table S9** show the levels of As and values of I_{geo} , CF, and ER in both topsoils and mangrove sediments. The As concentrations (mg/kg dry weight) ranged from 9.38 to 68.2 and 8.75 to 44.6, for topsoils and mangrove sediments, respectively. Overall, the average As level (31.2) in the topsoils was higher than that (18.2) in the mangrove sediments.

The I_{geo} ranges (1.64–4.51) of As in the topsoils were categorized between “moderately polluted” and “strongly to very strongly polluted” while those (1.54–3.89) of the mangrove were all categorized as “moderately polluted” and “strongly polluted” (Muller, 1969). The CF ranges (4.69–34.1) of As in the topsoils were categorized as “considerable contamination” and “high contamination” while those (4.38–22.3) of the mangrove sediments were categorized as

“considerable contamination” and “high contamination” (Hakanson, 1980).

The ER ranges (46.9–341) of As in the topsoils fell between “mild potential ecological risk” and “very high ecological risk” while those (43.8–223) of the mangrove sediments were categorized as “mild potential ecological risk” and “high potential ecological risk” (Hakanson, 1980). Overall, the average As values of I_{geo} , CF, and ER in the topsoils was higher than those in the mangrove sediments.

3.4 Health Risk Assessments of Sb and As in Topsoils and Mangrove Sediments

The Sb values of HQ_{ing} , HQ_{inh} , and HQ_{der} pathways and HI in the topsoils and sediments from Peninsular Malaysia in children and adults are presented in **Figure 9** and **Supplementary Table S10**. In the children, the Sb values of HQ_{ing} , HQ_{inh} , HQ_{der} , and overall HI in the topsoils ranged from 6.49×10^{-2} to 1.94×10^{-1} , 5.20×10^{-6} to 1.56×10^{-5} , 5.19×10^{-3} to 1.56×10^{-2} , and 7.01×10^{-2} to 2.10×10^{-1} , respectively. In the adults, the Sb values of HQ_{ing} ,

HQ_{inh}, HQ_{der}, and overall HI in the topsoils ranged from 7.97×10^{-3} to 2.39×10^{-2} , 2.14×10^{-6} to 6.42×10^{-6} , 1.24×10^{-2} to 3.73×10^{-2} , and 2.04×10^{-2} to 6.12×10^{-2} , respectively.

In children, the Sb values of HQ_{ing}, HQ_{inh}, HQ_{der}, and overall HI in the mangrove sediments ranged from 2.94×10^{-5} to 5.06×10^{-2} , 2.36×10^{-9} to 4.05×10^{-6} , 2.35×10^{-6} to 4.05×10^{-3} , and 3.18×10^{-5} to 5.46×10^{-2} . In adults, the Sb values of HQ_{ing}, HQ_{inh}, HQ_{der}, and overall HI in the mangrove sediments ranged from 3.61×10^{-6} to 6.21×10^{-3} , 9.71×10^{-10} to 1.67×10^{-6} , 5.64×10^{-6} to 9.70×10^{-3} , and 9.25×10^{-6} to 1.59×10^{-2} .

The Sb HI values based on the hazard quotients pathways of ingestion, inhalation, and dermal contact for Sb were all below 1.0 and greater in children compared to adults, and the Sb HI values in the topsoils were always higher than those in the mangrove sediments.

The As values of HQ_{ing}, HQ_{inh}, HQ_{der}, and HI in the topsoils and sediments from Peninsular Malaysia in children and adults are presented in **Figure 10** and **Supplementary Table S11**. In children, the As values of HQ_{ing}, HQ_{inh}, HQ_{der}, and HI in the topsoils ranged from 3.68×10^{-1} to 2.68, 1.03×10^{-5} to 7.51×10^{-5} , 1.44×10^{-3} to 1.04×10^{-2} , and 3.69×10^{-1} to 2.69, respectively. There are 9 sites out of 16 where the HI values were above 1.00 via the ingestion pathway in the topsoil samples. In adults, the As values of HQ_{ing}, HQ_{inh}, HQ_{der}, and HI in the topsoils ranged from 4.52×10^{-2} to 3.29×10^{-1} , 4.25×10^{-6} to 3.09×10^{-5} , 3.44×10^{-3} to 2.50×10^{-2} , and 4.86×10^{-2} to 3.54×10^{-1} , respectively.

In children, the As values of HQ_{ing}, HQ_{inh}, HQ_{der}, and HI in the mangrove sediments ranged from 3.43×10^{-1} to 1.75, 9.63×10^{-6} to 4.91×10^{-5} , 1.34×10^{-3} to 6.83×10^{-3} , and 3.44×10^{-1} to 1.76, respectively. Only 1 site out of 10 where the HI value was above 1.00 via the ingestion pathway is the mangrove sediments. In adults, the As values of HQ_{ing}, HQ_{inh}, HQ_{der}, and HI in the mangrove sediments ranged from 4.21×10^{-2} to 2.15×10^{-1} , 3.97×10^{-6} to 2.02×10^{-5} , 3.21×10^{-3} to 1.64×10^{-2} , and 4.54×10^{-2} to 2.31×10^{-1} , respectively.

Overall, there were 56% (9 out of 16 sites) in the topsoils for children with As hazard index values exceeding 1.00, in which almost all the As total HI values were contributed by the ingestion pathway. The HI values were greater in children than in adults, and the As HI values in the topsoils were always higher than those in the mangrove sediments.

The As values of CR_{ing}, CR_{inh}, and CR_{der} in the topsoils and sediments from Peninsular Malaysia in children and adults are presented in **Figure 11** and **Supplementary Table S12**. In children, the As values of CR_{ing}, CR_{inh}, and CR_{der} in the topsoils ranged from 1.66×10^{-4} to 1.20×10^{-3} , 4.67×10^{-8} to 3.40×10^{-7} , and 6.46×10^{-7} to 4.70×10^{-6} , respectively. In the adults, the As values of CR_{ing}, CR_{inh}, and CR_{der} in the topsoils ranged from 2.03×10^{-5} to 1.48×10^{-4} , 1.93×10^{-8} to 1.40×10^{-7} , and 1.55×10^{-6} to 1.13×10^{-5} , respectively.

In children, the As values of CR_{ing}, CR_{inh}, and CR_{der} in the mangrove sediments ranged from 1.54×10^{-4} to 7.87×10^{-4} , 4.36×10^{-8} to 2.22×10^{-7} , and 6.03×10^{-7} to 3.07×10^{-6} , respectively. In adults, the As values of CR_{ing}, CR_{inh}, and CR_{der} in the mangrove sediments ranged from 1.90×10^{-5} to 9.67×10^{-5} , 1.80×10^{-8} to 9.16×10^{-8} , and 1.44×10^{-6} to 7.36×10^{-6} , respectively.

Based on USEPA methods, a cancer risk value below 1.0×10^{-6} is considered “negligible,” $>1.0 \times 10^{-4}$ is considered “unacceptable,” and in the range from 1.0×10^{-6} to 1.0×10^{-4} is considered “acceptable” (USEPA, 1989). Based on **Figure 11**, all the As children CR values for both topsoils and mangrove sediments in all sites based on the ingestion pathway exceeded 1.0×10^{-4} which is considered “unacceptable.” For As adults’ CR values of the ingestion pathway, there are four sites (out of 16 sites) that exceeded the threshold limit in the topsoil samples, while all sites in the mangrove sediments were considered “acceptable.” All the CR values for inhalation pathways in both topsoils and mangrove sediments were considered “negligible” since they are lower than 1.0×10^{-6} . All the CR values for dermal contact pathways in both topsoils and mangrove sediments were considered “acceptable” since they were in the range from 1.0×10^{-6} to 1.0×10^{-4} , and some are “negligible.”

4 DISCUSSION

4.1 Comparable Levels of Sb in *Centella asiatica* With Reported Studies

The present ranges of As in the three parts of *C. asiatica* (0.31–1.61 mg/kg dry weight) from Peninsular Malaysia were comparable to various published research studies.

The present findings of As and Sb levels were in the order of roots > leaves > stems, which agrees with many reported studies on plants. Wilson et al. (2013) found substantially greater metalloid levels in the roots of plants compared to the sections above the ground, confirming our findings. Some researchers attributed Sb translocation from the roots to the upper part of the plants to Sb precipitation on the root membrane barriers, which hampered the transport of Sb solutions to the leaves (Tschan et al., 2009a). Tschan et al. (2009b) discovered the coherent relationship between plant Sb absorption and soluble Sb levels in the topsoil. The sites with Sb levels in *C. asiatica* from the present study indicated higher bioavailability of Sb in the sampling sites (Rainbow and Phillips, 1993). Plants growing near Sb pollution sources have greater Sb concentrations, according to Baroni et al. (2000).

Sb, being a non-essential element, is able to accumulate in plants (Ainsworth et al., 1991). Kabata-Pendias and Pendias, (2001) categorized plant tissue with 5–10 mg/kg of Sb as phytotoxic. Therefore, the present Sb range of 0.31–1.61 mg/kg dry weight in the three parts of *Centella* was not considered phytotoxic. According to Tschan et al. (2009a), the background ranges of Sb concentrations in the terrestrial plants were 0.20–50 mg/kg dry weight. This indicates that present Sb ranges were not elevated and hence can be considered the background level in the topsoils and mangrove sediments in Peninsular Malaysia. Kassem et al. (2004) reported that the Sb values ranged from 0.011 to 0.026 mg/kg dry weight in olive, eggplant, alfalfa, and cabbage leaves were grown in the Orontes River basin. Their data were significantly lower than those of the three parts in *C. asiatica* from the present study.

In the Pearl River Delta (PRD), South China, 112 pairs of soil and blooming cabbage were sampled from typical agricultural protection zones and vegetable-producing districts (Chang et al., 2022) to investigate the contamination levels of Sb and As in soils and harvested cabbages. Chang et al. (2022) reported that the ranges of Sb concentrations in the flowering cabbages across PRD were 0.015–0.20 mg/kg dry weight with a mean value of 0.038 mg/kg dry weight, exhibiting significant lower levels of Sb than radish leaves collected from the Xikuangshan mine area (range: 1.5–121 mg kg⁻¹) (He, 2007). This value is also below the tolerable concentration of plants for Sb (5 mg/kg dry weight) proposed by Eikmann and Kloke, (1993), and the levels between 5 and 10 mg/kg dry weight of Sb in plant tissue were reported to be phytotoxic (Kabata-Pendias and Pendias, 1985).

4.2 Comparatively Higher Levels of As in *Centella asiatica* With Reported Studies

The present ranges of As in the three parts of *C. asiatica* (0.21–20.8 mg/kg dry weight) from Peninsular Malaysia are comparatively higher than those in various published research studies.

Uddh-Söderberg et al. (2015) found that the As concentrations in lettuce ranged from 0.0022 to 4.3 mg/kg dry weight, while Bhattacharya et al. (2010) found that the total As concentrations in rice, wheat, pulses, and vegetables ranged from <0.0003 to 1.02 mg/kg dry weight in an arsenic-affected area of West Bengal (India). A reported study showed that the As concentrations measured for the lettuce, onion, radish, and bean greenhouse samples ranged from 0.00843–5.28 mg/kg wet weight (Ramirez-Andreotta et al., 2013). The present As ranges (0.2–20.8 mg/kg dry weight) in the leaves, stems, and roots of *C. asiatica*, in the current study, were greater than those reported in the studies mentioned above.

In Sweden and the European Union, there is no regulation for As in food (Uddh-Söderberg et al., 2015). The United Kingdom Food Standard Agency proposed a general As limit of 1.00 mg/kg in all foodstuffs (CAC, 2011). On the other hand, the Food Standard Agency of Australia and New Zealand (FSANZ, 2015) proposed a maximum level of total arsenic in cereal (1.00 mg/kg) and did not set restrictions on other food groups. The present As ranges (0.02–2.08 mg/kg wet weight) in the leaves, stems, and roots of *Centella*, from the present study, could be higher than the As limit by the United Kingdom Food Standard Agency (CAC, 2011) and the Food Standard Agency of Australia and New Zealand (FSANZ, 2015).

Haque et al. (2021) studied five heavy metals (including As) in the vegetables obtained from industrial, non-industrial, and As-contaminated regions and popular vegetable markets in Bangladesh. They reported that the As levels in vegetables were 0.15–0.55 mg/kg wet weight and 0.06–0.24 mg/kg wet weight for industrial and non-industrial areas, respectively. Qin et al. (2021) tested samples from vegetable plants cultivated in acidic mine water contaminated soil and found that the mean concentration of total As in the edible sections of leafy vegetables fell in the range of 0.41–7.61 mg/kg dry weight (mean: 2.39 mg/kg).

As concentrations in flowering cabbages across the PRD were reported to fall in the range of 0.021–1.20 mg/kg dry weight (mean: 0.15 mg/kg dry weight) (Chang et al., 2022). In comparison to previous reports, total As concentrations in the flowering cabbages in the current study were generally lower than those in the leafy vegetables collected near mining areas (range: 0.14–1.42 mg/kg dry weight with mean: 0.59 mg/kg dry weight for Shizhuyuan mine; range: 0.41–7.61 mg/kg dry weight with mean: 2.39 mg/kg dry weight for Dabaoshan mine) (Li L. et al., 2017; Qin et al., 2021). Furthermore, As levels in 96% of the flowering cabbages were lower than the Chinese National Standards' (GB 2762–2017) tolerance limit (0.50 mg/kg) for pollutants in foods (Chang et al., 2022).

Zheng et al. (2022) found a wide range of total As concentration in vegetable samples (0.52–15.94 mg/kg dry weight; mean: 6.55 ± 5.18 mg/kg dry weight) from samples collected from As-polluted farmlands, which were attributed to anthropogenic activities in Guangxi province, China. The highest As concentrations were found in Romaine lettuce (15.94 mg/kg dry weight), leaf lettuce (10.48 mg/kg dry weight), and leaf mustard (11.10 mg/kg dry weight).

Varol et al. (2022) found the mean As levels in some leafy vegetables grown in Malatya province (Turkey) were the highest in purple basil (0.297 mg/kg), purslane (0.161 mg/kg), and parsley (0.1 mg/kg wet weight). However, the mean concentrations of As in all crops were lower than the permissible concentrations (MPCs) set by the United Kingdom Food Standard Agency (CAC, 2012) (1.00 mg/kg wet weight) and the Chinese Health Ministry (MHPRC, 2013) (0.50 mg/kg wet weight).

4.3 No Non-Carcinogenic Risks of Sb and As in *Centella* but Are Higher in Children Than in Adults

The children's THQ values in both leaves and stems were higher than those in the adults. Second, all THQ values for Sb and As were below 1.0, except for As in children consuming the leaves collected from KBatas. This indicated that the THQ value (>1.00) of inorganic As in *C. asiatica* collected from KBatas exceeded the threshold value of 1, with a non-carcinogenic risk of As to the children. All EDI values for children and adults in the leaves and stems of *Centella* are lower than the PTWI for Sb and As which had further supported the aforementioned results of THQ values which were below 1.00.

Varol et al. (2022) reported that the As THQ of vegetables collected from Turkey was below the acceptable risk level of 1.00. Cheung et al. (2008) determined dietary exposure to three elements (including Sb) in food consumed by junior high school students in Hong Kong. The dietary intake of Sb was predicted as 0.567 µg (kg body weight)⁻¹ week⁻¹ for high-consumption high school students. Qin et al. (2021) reported that intake of vegetable plants cultivated on acidic mine water-polluted soils might pose considerable potential human health risk with a hazard quotient (HQ) of 2.7.

Wu et al. (2011) investigated the health risks and major routes of long-term human exposure to Sb on residents of the Tin Mine (XKS) Sb mine in China. They found that inhabitants near XKS

had an inorganic dietary intake of As (107 $\mu\text{g}/\text{day}$), below the PTWI of 15 $\mu\text{g}/\text{kg}$ body weight/week (equivalent to 129 μg A/day).

4.4 Carcinogenic Risk of As in Most Sampling Sites With the Unacceptable Status

The As CR values are concerning as 10–60% of the studied sites of *C. asiatica* had higher values than the As CR threshold limit. Bhatti et al. (2013) reported that the As concentration of spinach leaves was 1.6–6.4 times greater than the maximum concentration of inorganic As in China (0.05 mg/kg fresh weight). The HI values were above 1 and CR levels were also above the highest target of 1×10^{-4} , which identified the potential risk to humans from ingesting spinach leaves. Uddh-Söderberg et al. (2015) investigated the health risks from As in vegetables grown in private gardens close to 22 contaminated glass factories. They found that the increased levels of As in the soil of residential gardens were reflected in increased As levels of vegetables, and the intake of these vegetables might cause cancer risks in humans. They found that they did not exceed the threshold for chronic non-carcinogenic or acute effects; however, the reported As CR are inclusive of other non-native vegetables and total exposure.

Based on vegetables from Bangladesh, Haque et al. (2021) reported that the probabilistic risk, individual risk, and total cancer risk (TR) for As had surpassed the threshold level (1×10^{-4}) and safe limit (1×10^{-6}), denoting higher lifetime cancer risk due to long-term consumption of these vegetables.

4.5 No Non-Carcinogenic Risk of Sb in the Topsoils and Mangrove Sediments

The present ranges of Sb in the topsoils (2.21–6.61 mg/kg dry weight) and mangrove sediments (0.001–1.72 mg/kg dry weight) from Peninsular Malaysia are comparatively lower than those mentioned in many reported studies. I_{geo} , CF, and ER values for Sb are higher in the topsoils than in the mangrove sediments. The HI based on three pathways indicated no non-carcinogenic risks of Sb.

Overall, the topsoils' mean Sb level (4.00) was higher than 0.97 in the mangrove sediments. Chang et al. (2022) reported that the total Sb contents in soils from typical agricultural protection and vegetable-producing regions of the PRD ranged from 0.20 to 7.20 mg/kg dry weight (mean: 1.60 mg/kg dry weight). This range is within the mean Sb concentration (0.54–2.93 mg/kg dry weight) in soils from various regions across China (He et al., 2012). About 91% of the soil samples were within the range of background Sb levels (0.80–3.00 mg/kg dry weight) for China soils (He, 2007). Furthermore, 95% of the total soil samples were lower than the maximum allowed Sb concentration (3.50 mg/kg dry weight) proposed for soils in the Netherlands (Crommentuijn et al., 2000).

The Sb ranges (Mean: 4.00 in the topsoils and 0.97 in the mangrove sediments) in the current study were lower than those mentioned in many reports from the mining area of Sb such as South and North Sb mines (total Sb: 3,061 mg/kg dry weight; 74.2–16,389 mg/kg dry weight) as reported by Li et al. (2014) and

in the sediments of the Lianxi River (Xikuangshan Sb mine; Sb values of 57.11–7,315.7 mg/kg) (Wang et al., 2011) and an abandoned Sb ore deposit in Portugal (5,956 mg/kg dry weight; Pratas et al., 2005). In addition, the significantly higher Sb levels were also reported in the soil near the Xishan mine in Hunan province's Lengshuijiang city (5,949 mg/kg; Qi et al., 2011a), in the surface soils of the Swiss shooting range (13,800 mg/kg dry weight; Johnson et al., 2005). Since Sb and its compounds have long-range transport properties, higher concentrations of Sb in topsoils could be attributable to air dust settlement (Tian et al., 2014).

However, the present Sb ranges were comparable to those in Xuzhou (China), with a mean of 3.46 mg/kg of Sb in urban soils (Xue et al., 2005), and 1.23 mg/kg of Sb was found in Chinese farming soils (Niu et al., 2013). Jiang et al. (2020) examined the levels of eight heavy metals (including As) in soil, groundwater, air, and locally produced grain (wheat and corn) and vegetables in a village located close to a battery factory in Xinxiang, Henan Province, China. They found that the main input of As originated from activities associated with sewage irrigation and battery plant deposition. Based on soils collected from the coal mines in the Huaibei and Huainan areas of Anhui Province, China, Qi et al. (2011a) identified that the mean Sb content in the 33 samples was 4.00 mg/kg, which is lower than that in coals from this region (6.20 mg/kg).

Based on mining-affected grassland soils, Casado et al. (2007) found that the range of total Sb contents in soils was 60–230 mg/kg, indicating a high degree of pollution of soils. Sia and Abdullah (2012) studied the concentrations of mineral-forming elements and trace elements in coal and coal ashes from Balingian coalfield, Sarawak, Malaysia, and reported an enriched content of Sb (96 ppm) in Balingian coal relative to their respective coal Clarke.

Patyar and Moghanlo, two mining regions in Iran, were studied for their soil and vegetation (Hajiani et al., 2015). In Moghanlo, the total Sb concentrations in soil were 358–3,482 mg/kg, while in Patyar, the total Sb concentrations were 284–886 mg/kg. Sb levels in plant shoots ranged from 0.8 to 287 mg/kg and 1.3 to 49 mg/kg, respectively. *Achillea wilhelmsii* and *Matthiola farinosa* were the strongest Sb accumulators among these species, with 141 and 132 mg/kg Sb in their leaves, respectively.

Based on Baoji urban soil, Li X. et al. (2017) studied seven toxic elements (including As and Sb). They found that prolonged industrial activities have enriched the Sb content (26.00 mg/kg mean value) in urban soil. Based on nine surface soil samples contaminated from lead/zinc and iron smelting operations and coal-fired power plants, Serafimovska et al. (2013) reported that the main part of total Sb (2.5–105 mg/kg dry weight) was associated with the residual fraction in all soils. Chang et al. (2022) investigated the contamination levels of Sb and As in soils and harvested cabbages across the PRD (South China) by collecting 112 pairs of soil and flowering cabbage samples from agricultural protection zones and vegetable-producing districts in the region. The levels of Sb pollution in PRD cabbages and soils were generally low, although the soils were moderately polluted by As.

4.6 High Ecological Risk of As and Carcinogenic Risk of As in the Topsoils and Mangrove Sediments

The present ranges of As in the topsoils (9.38–68.2 mg/kg dry weight) and mangrove sediments (8.75–44.6 mg/kg dry weight) from Peninsular Malaysia are comparable to many reported studies in the literature. I_{geo} , CF, and ER values for As are also higher in the topsoils than those in the mangrove sediments. The hazard index based on three pathways indicated no non-carcinogenic risks of As. Higher and more As carcinogenic risk in the topsoils than in the mangrove sediments was observed.

Based on mining-affected grassland soils, Casado et al. (2007) found that total As contents in soils ranged from 42 to 4,530 mg/kg, denoting elevated levels of pollution in soils. Sia and Abdullah (2012) studied the concentrations of mineral-forming elements and trace elements in coal and coal ashes from Balingian coalfield, Sarawak, Malaysia, and reported an enriched content of As (as high as 181 ppm) in Balingian coal relative to their respective coal Clarke.

The mean As level (31.2) in the topsoils was higher (18.2) than in the mangrove sediments. The As levels from these two areas were higher than those (10 mg/kg dw) of the Swedish EPA (2009) generic soil guideline value for residential areas. Also, the As ranges in both areas were higher than the reported As UCC by Wedepohl (1995) of 2.00 mg/kg dry weight and close to EU recommended maximum acceptable limit for agricultural soil (20.0 mg/kg dry weight) (Rahman et al., 2007) for the mangrove sediments. Therefore, the topsoils have higher anthropogenic As sources that emptied into the coastal mangrove sediments in Peninsular Malaysia.

Chang et al. (2022) reported that the total As concentrations in soils collected from typical agricultural protection and vegetable-producing regions across the PRD ranged between 5.6 and 228 mg/kg dry weight. These values are similar to the range (15.2–174 mg/kg dry weight) reported in Dabaoshan mine and the surrounding areas (Qin et al., 2021) but much lower than those of the vegetable soils (360–1,054 mg/kg dry weight) collected from the vicinity of the Shizhuyuan mining area (Li L. et al., 2017). Furthermore, 52% of the studied soil contained As exceeding the risk screening value (40 mg/kg dry weight) proposed by the Chinese soil environmental quality risk control standard for soil contamination of agricultural land (GB15618-2018) (Chang et al., 2022).

Overall, 56% of topsoil samples (9 out of 16 sites) have As hazard index values exceeding 1.00 for children. Almost all the As total HI values are contributed by the ingestion pathway. The HI values are higher in children than in adults, and the As HI values in the topsoils are always higher than those in the mangrove sediments. All the As children CR values for both topsoils and mangrove sediments in all sites based on the ingestion pathway exceeded 1.0×10^{-4} , which is considered “unacceptable.” For As adults’ CR values of the ingestion pathway, 4 out of 16 sites exceeded the threshold limit in the topsoil samples, while all sites in the mangrove sediments are considered “acceptable.” All the CR values for inhalation

pathways in both topsoils and mangrove sediments are “negligible” since they are lower than 1.0×10^{-6} . All the CR values for dermal contact pathways in both topsoils and mangrove sediments are considered “acceptable” since they range from 1.0×10^{-6} to 1.0×10^{-4} , and some are “negligible.”

Cao et al. (2014) investigated children’s health hazard risks and exposure levels to 12 elements (including As and Sb) in water, soil, dust, air, and locally produced food using field sampling and questionnaire-based surveys in China’s largest coking area. The food intake and air inhalation exposure pathways each provided about half of As exposure. They concluded that children who reside near coking operations face higher health risks from As and Sb, and that food safety is necessary to protect these children.

Total arsenic (As) concentrations in poultry feed, manure, agricultural soils, and food plants from Khyber Pakhtunkhwa Province, Pakistan were examined by Muhammad et al. (2021). In the edible sections of food plants, the total As concentrations ranged from 0.096 to 1.25 mg/kg. The mean daily intake (ADI), HQ, HI, and CR were used to determine the risk to human health. The HI was three times higher in children (2.1) than in adults (0.79), indicating that the children face non-cancer health hazards (HI index >1). For children, the CR values were somewhat higher (1.53×10^{-4}) relative to USEPA and WHO limits (1×10^{-6} to 1×10^{-4}), but for adults, a low CR was found from ingestion of selected food plants. They have concluded that the source of pollution in food plant was attributed to poultry manure leading to non-CR and CR in agricultural areas.

4.7 Higher Ecological–Health Risks of Sb and As in the Topsoils Than in the Mangrove Sediments

The ecological risk assessments (I_{geo} , CF, and ER) and health risks (HQ and HI) for Sb and As in the topsoils were higher than those in the mangrove sediments. The higher ecological–health risks of Sb and As can be explained from three perspectives.

First, the anthropogenic sources from industrial activities started far from the coastal area. Therefore, the topsoils collected near the urban and industrial areas with observable human activities had speculatively contributed to the higher levels of Sb and As indicated by the present findings. Some of the topsoils were collected near rivers adjacent to industrial areas, such as Butterworth and Kepala Batas, and nearby active agricultural areas where the use of pesticides was highly possible. Yap and Pang (2011) previously reported the sediments collected from drainage and rivers that had higher levels of heavy metals than the coastal sediments. Yap et al. (2022) recently reported that land uses played an important role of a contributing factor in the elevation of heavy metals in the topsoils of Peninsular Malaysia. These two studies clearly explain why the levels of Sb and As are higher in the topsoils than in the mangrove sediments.

Second, it has been shown that the elemental levels will decrease with increasing distance from the sources of pollution (Yap et al., 2007; Yap and Al-Mutairi, 2022), which might explain the lower levels of Sb and As in the mangrove sediments than those in the topsoils adjacent to anthropogenic sources. Various

studies from other regions have reported similar findings (Qi et al., 2011a, 2011b; Hajiani et al., 2015; Qin et al., 2021).

Third, with the understanding that sediments are considered a sink of metal pollution (Szefer et al., 1995; Yap, 2018), the mangrove sediments are assumed to accumulate the highest levels of Sb and As. The present finding showed a reverse pattern indicating the mangrove forests acted as a phytoremediator of Sb and As that caused the lower levels of As and Sb in the mangrove sediments. Reasonably, the mangrove forests are effective phytoremediators of Sb and As since mangrove trees thrive in marshy ecosystems (Defew et al., 2005; Nazli and Hashim, 2010; Nirmal et al., 2011).

5 CONCLUSION

Concentrations of As and Sb in *C. asiatica*, topsoils, and mangrove sediments collected from Peninsular Malaysia were examined. The present findings indicated comparable levels of Sb in *C. asiatica* but comparatively higher levels of As in *C. asiatica* than those in previous studies. There were no non-carcinogenic risks of Sb and As in *C. asiatica*, although children are potentially exposed to higher risks than adults. The THQ values of As and Sb in the plant were below 1.00 for children and adult Malaysian consumers, denoting no non-carcinogenic risks of As and Sb via *C. asiatica* consumption, except for As in children consuming the leaves collected from the sampling site at Kepala Batas. We also found the calculated values of estimated weekly intakes below the established provisional tolerable weekly intakes of As and Sb for children and adults. However, the As CR of children in the leaves and stems of *C. asiatica* showed more public concern than that of adults, with 10–60% of the sampling sites exceeding the As CR threshold limit, which is considered “unacceptable.”

Risk assessments of As and Sb in topsoils and mangrove sediments indicated that the Sb ER ranges in the topsoils could reach “considerable potential ecological risk.” In contrast, those of As in the topsoils could reach “very high ecological risk.” The ER ranges of Sb in the mangrove sediments were all categorized as “minimal potential ecological risk,” while those of As in the mangrove sediments could reach “high potential ecological risk.” For the health risks of As and Sb in the topsoils and mangrove sediments, there was no non-carcinogenic risk of Sb in the topsoils and mangrove sediments. The Sb HI values were all below 1.0, although the values are higher in children than in adults. The Sb HI values in the topsoils are always higher than those in the mangrove sediments. Overall, there are 56% in the topsoils for children with As HI values exceeding 1.00. The ingestion pathway contributed almost all the As total HI values. Furthermore, a high ecological risk of As and carcinogenic risk of As in the topsoils and mangrove sediments were evident, with the HI values in children being higher than those in adults.

In conclusion, there were higher ecological–health risks of Sb and As in the topsoils than in the mangrove sediments. This indicates that anthropogenic sources of Sb and As originated from the land-based activities before reaching the mangroves. The unacceptable status based on As CR warrants future exposure research because dietary intake includes more than just *C. asiatica* and overall exposure results from several exposure pathways. Thus, minimizing As and Sb concentrations in vegetables is essential to safeguard human health and the environment from hazardous risks.

Therefore, establishing guidelines for Sb and As for agricultural soils and resourceful mangrove ecosystems in this region is highly recommended. This effort will ease the pollution caused by anthropogenic inputs to avoid similar disastrous chemical pollution as happened in Kim River, Southern Peninsular Malaysia (Yap et al., 2019).

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

Conceptualization, CKY; formal analysis, WHC; funding acquisition, CKY; investigation, KK and GHO; methodology, GHO, WHC, RH, and EC; project administration, CKY and WST; resources, WST, WMS, NAW, WHC, and MHI; supervision, CKY; validation, WST and RG; writing—original draft, CKY; writing—review and editing, CKY, WST, WHC, RN, RG, MHI, MM, HO, WC, FBE, HO, KAA, SAA, MS, MK, CFY, ARB, AB, HMMZ, TA, AN, MS, MAAR, GHO, GS, and LSW. All authors have read and agreed to the published version of the manuscript.

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SUPPLEMENTARY MATERIAL

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