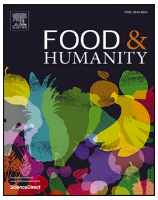
Food and Humanity 6 (2026) 101200

Contents lists available at ScienceDirect

Food and Humanity

journal homepage: www.editorialmanager.com/foohum/journal_overview.html



Dietary exposure to toxic trace elements through cooked rice in high rice consuming populations: A comprehensive risk assessment in Sri Lanka

Jayani Wathsala Gunawardana [a], Inoka Chinthana Perera [a], Nekadage Don Amal Wageesha [c], Chamindri Witharana [b], Sameera Anuruddha Gunawardena [d,*]

[a] *University of Colombo Faculty of Science, Sri Lanka*
[b] *University of Colombo Faculty of Medicine, Sri Lanka*
[c] *Sabaragamuwa University, Faculty of Medicine, Sri Lanka*
[d] *International Medical University, Kuala Lumpur, Malaysia*

ABSTRACT

Toxic Trace Element (TTE) exposure is an increasing public health concern, and rice, Sri Lanka's primary staple, requires ongoing monitoring. This study analysed Cd, As, Pb, and Cr in 25 rice grain composites (from 54 samples) across most commonly consumed varieties in Sri Lanka, assessing differences by pericarp colour, parboiling status, and variety types. Median (IQR) concentrations of TTEs in raw rice were; Cd: 0.032 (0.062 – 0.008), As: 0.049 (0.063 – 0.035), Pb: 0.095 (0.170 – 0.047), and Cr: 0.376 (0.532 – 0.166) mg kg^{-1} . While no samples exceeded JECFA limits for Cd or As, one exceeded the Pb limit, and 20% surpassed the stricter 0.1 mg kg^{-1} Cd; a rigorous threshold specific to several rice consuming countries. Cooking significantly reduced TTE availability ($p < 0.001$). Estimated daily intakes (EDIs) for a typical Sri Lankan adult upon average cooked rice consumption were; Cd: 0.271 ± 0.279 μg kg^{-1} bw^{-1}, As: 0.297 ± 0.126 μg kg^{-1} bw^{-1}, Pb: 0.643 ± 0.500 μg kg^{-1} bw^{-1} and Cr: 0.379 ± 0.333 μg kg^{-1} bw^{-1}respectively, with the JECFA-PTMI for Cd exceeded by 8.3% and the EFSA-TDI by 25% of samples. Non-carcinogenic risk was evident only for Pb, while Cd and As indicated potential carcinogenic risks. Findings highlight the need to establish maximum country-specific threshold/cut-off values for continuous surveillance of TTE contamination in Sri Lankan rice to safeguard consumers with higher rice consumption habits.

1. Introduction

Trace elements occur naturally in the Earth's crust, some of which are essential to metabolism while others referred to as Toxic Trace elements (TTEs), such as; Cadmium (Cd), Arsenic (As), Lead (Pb) and Chromium (Cr) considered hazardous for human health. TTEs function as potent metabolic disruptors that accumulate in tissues and organs, implicated in numerous acute and chronic disorders even at ultra - low concentrations (Jaishankar et al., 2014; Mitra et al., 2022). As non – biodegradable, persistent environmental contaminants with extensive biological half-lives, TTEs are biomagnified through food webs, rendering humans highly vulnerable to intoxication (Ali & Khan, 2019; Hajeb et al., 2014) primarily through the ingestion of contaminated food and water (Jeyasanta & Patterson, 2025; Scutarașu & Trincă, 2023)

The World Health Organization (WHO) identifies Cd, As, and Pb among the ten chemicals of major public health concern (WHO, 2020). Chronic low-grade exposure to the nephrotoxin Cd is causative of renal, hepatic, and skeletal damage, including the Itai-itai disease (Genchi et al., 2020). While organic As is readily metabolized, inorganic As is highly toxic, inducing hypertension, diabetes, and various malignancies (Fatoki & Badmus, 2022; Popowich et al., 2016). Similarly, Pb exposure targets renal, hepatic (Collin et al., 2022) and the nervous system, notably contributing to cognitive dysfunction and impaired development in children (Ramírez Ortega et al., 2021). Furthermore, while Cr is an essential metabolic cofactor, excessive intake of its hexavalent form (Cr (VI)) can cause severe damage to the respiratory system, gastrointestinal tract, and skin (Shin et al., 2023).

Dietary staples represent critical vectors for chronic TTE exposure due to their high consumption frequency and quantity. Therefore, several statutory threshold limits have been established globally to monitor the levels of TTEs to ensure the safety of food items for human consumption (Scutarașu & Trincă, 2023). Rice (*Oryza sativa* L.) is a staple cereal for over half the global population, with 95% of its production consumed in developing nations (Solh, 2024; U.S. Department of Agriculture., 2025). In Sri Lanka, rice and its products take a predominant role in the household diet, with an annual per capita consumption of 114 – 120 kg (RRDI Sri Lanka, 2024). This daily intake of ~300 g ranks Sri Lanka among the top ten rice-consuming nations

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* Corresponding author.

E-mail addresses: jayani.zoology@stu.cmb.ac.lk (J.W. Gunawardana), icperera@sci.cmb.ac.lk (I.C. Perera), awageesha@med.sab.ac.lk (N.D.A. Wageesha), chamindri@bmb.cmb.ac.lk (C. Witharana), sameera@imu.edu.my (S.A. Gunawardena).

https://doi.org/10.1016/j.foohum.2026.101200
Received 21 March 2026; Accepted 25 April 2026
Available online 7 May 2026
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globally, as the population relies heavily on rice for primary caloric and protein requirements (Kadupitiya et al., 2022; Xu et al., 2020).

Sri Lankan rice cultivation is diversified across all climatic zones and two distinct seasons, *Yala* (May – August) and *Maha* (September – May) (Department of Census & Statistics - Sri Lanka, 2025). While the country is home to over 2000 nutrient-rich 'Traditional/ heirloom' cultivars, valued for their nutritional and medicinal properties (Gunawardana et al., 2024), the modern market is dominated by high-yield '*Improved*' varieties which were hybridized from indigenous breeds since 1950s (Ginigaddara & Disanayake, 2018; Rambukwella & Priyankara, 2016). Both categories produce grains with red and white pericarps that undergo various post-harvest treatments, including parboiling, husking, and varying degrees of milling, before reaching commercial retail markets. These factors are critical as they influence the final elemental concentrations available for human consumption (Mishra et al., 2023; Samarajeewa, 2022).

The contamination of Sri Lankan rice has been extensively debated in the context of Chronic Kidney Disease of Unknown Etiology (CKDu) (Fernando et al., 2020; Kulathunga et al., 2022; Lockwood et al., 2024; Lunyera et al., 2016; Navarathna et al., 2021). Although many studies suggest TTE levels in rice remain within international regulatory limits (Diyabalanage et al., 2016; Jayatilake et al., 2013; Lockwood et al., 2024; Lunyera et al., 2016), these benchmarks (e.g., FAO/WHO Joint Expert Committee on Food Additives - JECFA, European Food Safety Authority - EFSA) are often based on a global average intake of ~128 g person^{-1} day^{-1}. This standard significantly underestimates health risks for South Asian populations where median daily intake reaches ~630 g person^{-1} day^{-1} (Bhavadharini et al., 2020). Evaluating the concentration of toxic trace elements (TTEs) in the '*food as consumed*' format is critical for accurate health risk modeling, as the majority of existing literature relies on raw grain analysis overlooking the impact of domestic preparation (Meharg et al., 2013; Mwale et al., 2018; Norton et al., 2014).

To address these limitations, this study first quantifies Cd, As, Pb, and Cr levels in raw commercial grains and evaluates the impact of a standardized domestic cooking process on levels remaining in cooked rice. Elemental concentrations and transitions are compared across rice varietal categories, further differentiating by pericarp color and parboiling treatment. These cooked-grain data are then utilized to conduct a localized toxicological risk assessment, investigating the potential for chronic dietary TTE exposure and related health effects for the Sri Lankan adult consumer, providing the empirical basis necessary to establish regional safety standards that reflect the true chronic exposure risks for populations dependent on rice as a primary caloric and protein source.

2. Methods

2.1. Sample collection

A total of 54 samples of consumer–ready, husked rice grains were purchased from selected Dedicated Economic Centers (DECs) in Sri Lanka (Fig. 1).

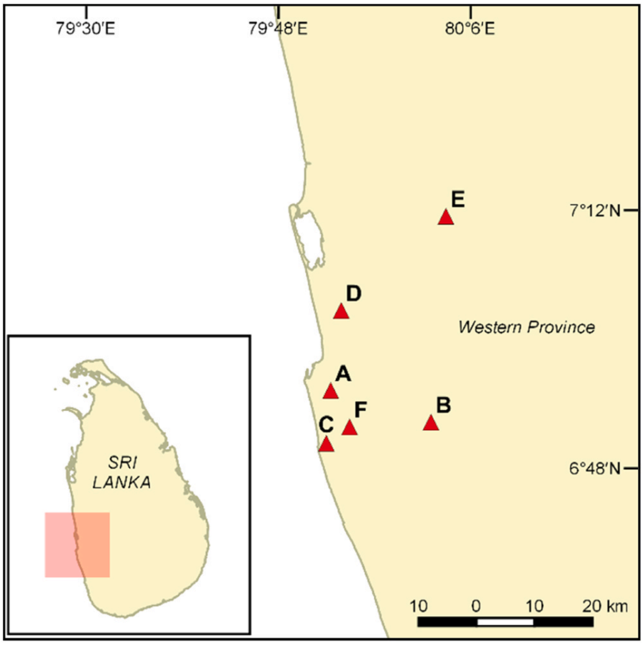

Fig. 1. Sampling locations – Dedicated Economic Centers (DECs) in Sri Lanka; Western province, (A: Narahenpita, B: Meegoda, C: Ratmalama, D: Welisara, E: Veyangoda, F: Bokundara); Scale bar shows kilometers, where 1 cm represents 10 km.

The DECs are a market space for farmers, intermediaries and wholesale vendors to directly sell key food commodities (Ministry of Agriculture and Plantation Industries - Sri Lanka, 2024). These 'natural distributors' (Weerakoon, 2021) play a key role in the retail of rice that was harvested and milled in all geographical regions in Sri Lanka. The ten most commonly consumed, locally produced varieties in Sri Lanka (Department of Census & Statistics, Ministry of Economic Policies & Plan Implementation, 2019; Department of Census & Statistics, Ministry of National Policies & Economic Affairs Sri Lanka, 2016; Department of Statistics & Ministry of Finance & Planning Sri Lanka, 2010; Galappattige, 2020; Kankanamge and Beillard, 2024) were included in the study. The grain samples encompassed both parboiled and non-parboiled varieties with varying degrees of milling representing both red and white pericarp colors (Table S1). There were three varieties in the Traditional/ heirloom category: Kaluheenati ($n = 5$), Suwandel ($n = 4$), Pachchaperumal (Siyapath-el) ($n = 3$), six varieties in the Improved category: Red Nadu ($n = 6$), White Nadu ($n = 6$), Red Samba ($n = 4$), White Samba ($n = 6$), Red Kekulu ($n = 7$), White Kekulu ($n = 8$). The non-locally produced Imported Indian Basmati (n = 5) was also included as a comparison. Rice samples were transferred to sealable high – density polyethylene (HDPE) bags and stored in room temperature for immediate processing. After evaluating the grain morphological and varietal indexes such as; grain length, width, elongation ratio, grain weight, they were categorized accordingly with shapes and sizes as well as pericarp colour, parboiling status and retail identifiers as guided by the Rice Research and Development Institute (RRDI)- Bathalegoda Sri Lanka (Table: S1), to pool the total sample cohort of 54 into 25 analytical composites which were aliquoted, proceeded for cooking and stored in -20 °C for lyophilisation.

2.2. Domestic cooking procedure

A weighed standard portion of rice was cooked using a standardized domestic cooking procedure (Gunawardana et al., 2023; Gunawardana et al., 2025). Briefly, 1.5 cups of rice were washed using three independent rinse and drain cycles and cooked using 1: 2.25 V/V of rice: water in a domestic electric rice cooker. After the cooking cycle, the cooked rice was allowed to cool. Aliquots of washed and cooked rice were stored at -20 °C in metal free (10% HNO_3 overnight bath and washed using deionized water (Milli Q 18 Ω, Millipore, USA)) High Density Polypropylene (HDPP) sealed containers until lyophilisation.

2.3. Lyophilisation, microwave digestion, elemental profiling and quality control measures

Approximately ~50 g aliquots of raw, washed and cooked grain fractions were lyophilized using a freeze dryer (Lanconco -FreezeZone, USA) at -40 °C under vacuum until a constant weight. Freeze - dried grains were powdered, homogenized and stored sealed at -20 °C for digestion.

Elemental profiling of digested rice samples were carried out using standardized methods (Gunawardena et al., 2020) with modifications. An analytical portion of ~0.2 g (analytical balance, 0.001 g, Satorius, Germany) lyophilized grain powders were subjected to microwave – assisted acid digestion (Sigma- Aldrich Trace Select®, Germany; HNO_3, HCl and high purity H_2O_2) at 180–200 °C for 30 min using a high-pressure microwave digester (CEM/MARS-6, XP-1500, USA). Digests were filtered (Whatman® Grade 1, 11 μm, UK) and diluted to 50 ML volume with deionized water, re-filtered (disposable membrane filters, 0.45 μm) to remove total dissolved solids and stored at 4 °C until elemental profiling. Method blanks and external quality control material; matrix matched Certified Reference Material (CRM) (European Commission IRMM – 804 Rice Flour) were de-identified and similarly processed and profiled within samples runs.

Water used for washing and cooking (Colombo Municipal Council general water supply) of each rice sample (50 ML) was collected into HDPP containers and immediately stabilized with an addition of 0.5 ML of HNO_3 (5% made from Trace Select® 70% HNO_3 using Milli Q 18 Ω water) and sealed. The samples were then mixed gently through inversion and at stored at 4 °C until analysis. Prior to profiling with ICP-MS, the water samples were filtered through 0.45 disposable membrane filters.

Elemental profiling was conducted using Inductively Coupled Plasma Mass Spectrometry (ICP – MS) (Agilent 7900- ASX-500, USA), calibrated with 2 A rare earth element panel (internal calibration std. 0, 0.5, 1, 5, 20,50, 100, 250, 500 and1000 ppb) (Table S2), using No gas mode for Cd, Pb and Cr and He mode for As using standard ICP-MS operation conditions (Table S3) in accordance with routine elemental analyses on complex matrixes (Gunawardena et al., 2020, 2021). Per each 30-sample analysis round, 0.5 ppb and 20 ppb standards were re-analysed, where $\leq$ 3 standard deviations (SD) were considered as the run is in control, alongside the within-day precision estimates. The Limit of Detection (LOD) for each element was calculated as 3.29 times the standard deviation of the method blanks ($n = 7$) while the Limit of Quantification (LOQ) was calculated as the 10 times the standard deviation of the method blanks ($n = 7$).

2.4. National rice consumption data

Rice consumption rates for the Sri Lankan adults were estimated by drawing on multiple published data sources (Department of Census & Statistics, Ministry of Economic Policies & Plan Implementation, 2019; Fernando et al., 2020; Jayatissa et al., 2014; Jayawardena and Herath, 2017; Jayawardena et al., 2012; Ministry of Health Sri Lanka, 2020; Xu et al., 2020; Bandara et al., 2021; Ministry of Health Sri Lanka, 2021; Kadupitiya et al., 2022; Saddhananda, 2022; Kankanamge & Beillard, 2024; RRDI Sri Lanka, 2024).

Data on cooked rice consumption specific to Sri Lankan adult population was scarce in published literature. Jayawardena et al. (2012), (2017) reported a cooked rice consumption of 0.8 kg person^{-1} day^{-1} (a consumption pattern reflective of 200 – 600 g consumed twice daily).

The majority of the reviewed literature reported rice consumption on a raw rice basis, expressed either as daily or yearly intakes for adults (Fernando et al., 2020; Jayatissa et al., 2014; Ministry of Health Sri Lanka, 2020; Xu et al., 2020; Bandara et al., 2021; Kadupitiya et al., 2022; Saddhananda, 2022; Kankanamge & Beillard, 2024; RRDI Sri Lanka, 2024). The mean raw rice intake from these studies was 0.3095 kg person^{-1} day^{-1}.

The mean raw rice intake was subsequently converted to cook rice intake equivalent using a raw-to-cook weight conversion factor (yield factor) of 3.01, established specifically for Sri Lankan rice (Gunawardana et al., 2023) and supported by other studies (Adikari & Thamilini, 2018; Aydin, 2022; Lisciani et al., 2022). The converted cooked rice intake equivalence was 0.9316 kg person^{-1} day^{-1}.

To derive a representative consumption value for use in exposure estimations, the cooked rice equivalent obtained from raw rice conversion (0.9316 kg person^{-1} day^{-1}) and the directly reported cooked rice consumption (0.8 kg person^{-1} day^{-1}, Jayawardena et al., 2012, 2017) were averaged yielding a final cooked rice consumption estimate of 0.918 kg for a Sri Lankan adult. This value was used for all subsequent dietary exposure calculations.

2.5. Exposure risk assessment for dietary intake of TTEs

2.5.1. Maximum permissible values (MLs), tolerable weekly intake values (TWIs) & provisional tolerable monthly intakes (PTMIs), country specific thresholds

Exposure risk assessment for TTEs in rice was conducted using; WHO/FAO Joint Expert Committee for Food Additives and Contaminants (JECFA) – Maximum Permissible Levels (MLs) for specific elements (JECFA, 2023) and further compared with country specific contaminant threshold values by India (Food Safety & Standards Authority of India, 2011), China (Food Safety Authority China, 2023; National Health Commission of the People's Republic of China, 2017), Japan (Japan External Trade Organization, 2010) and New Zealand (Board of Food Standards Australia New Zealand, 2015).

The monthly and weekly intakes of TTEs were calculated using cooked rice consumption data (Eq. 1 & Eq. 2) and evaluated against the Codex Alimentarius statutory Provisional Tolerable Monthly Intake (PTMI) limits and Tolerable Weekly Intake (TWI) by European Food Safety Authority (EFSA) – (EFSA Panel on Contaminants in the Food Chain (CONTAM), 2011) respectively. For the toxicological assessment the body weight categories of 50, 55, 61.4 (average Sri Lankan body weight) (Ministry of Health, Nutrition & Indigenous Medicine and World Health OrganizationWHO, 2015) and 70 kg (global standard body weight) were considered.

$$PTMI = (C \times \quad RCR_{month}) \quad /BW \tag{1}$$

$$TWI = (C \times \quad RCR_{week})/BW \tag{2}$$

C = concentration of the element (mg kg $^{-1}$ ww cooked rice)
RCR_{month} = cooked rice consumption per month per person (kg person^{-1})
RCR_{week} = cooked rice consumption per week per person (kg person^{-1})
BW = Average adult body weight (kg)

2.5.2. Non-carcinogenic risk assessment

Total As was considered fully as inorganic, relevant to worst-case risk estimation scenario, supported by speciation studies showing predominantly inorganic forms in Sri Lankan rice (Senarathne et al., 2023). This conservative approach aligns with regional hydrogeochemistry favoring inorganic As uptake under paddy conditions (Diyabalanage et al., 2016; Jayasumana et al., 2015).

Total chromium was risk-assessed as Cr (III) the less hazardous valency (Health Security Agency- United Kingdom, 2016; US EPA, 2016) as Sri Lankan studies indicating low levels of Cr primarily as Cr (III) (Kodikara et al., 2023; Lockwood et al., 2024). Moreover, Cr (III) is the naturally occurring form of Cr in food (CONTAM, 2014). Despite the low levels previously being reported, the non – carcinogenic risk assessment was conducted to evaluate the possibility of hazardous exposures upon high rice consumption patterns.

The non-carcinogenic risk was modelled using Estimated Daily Intake (EDI) (Eq.3) and appraised through several toxicological guidelines; EFSA – Tolerable Daily Intake (EFSA-TDI), U.S Food and Drug Administration (FDA) Interim Reference Levels (IRLs) FDA Toxicological Reference Values (TRVs) issued for each element.

The Hazard Quotients were calculated (E.q. 4 & 6) based on U.S. Environmental Protection Agency (US-EPA) Oral Reference Doses (RfD) and Agency for Toxic Substances and Disease Registry (ASTDR) for all TTEs. Minimal Risk Levels (MRLs) were only calculated for Cd. There is no difference between the two guidelines for As while there are no MRLs established for Pb and Cr (III) to evaluate chronic exposure outcomes.

The RfD values of TTEs used for the assessment were Cd: 1 μg kg^{-1} day^{-1}, As: 0.3 μg kg^{-1} day^{-1}, Pb: 4 μg kg^{-1} day^{-1} and Cr: 1.5 mg kg^{-1} day^{-1} (Caicedo-Rivas et al., 2022; Lee et al., 2023; Onuoha et al., 2016; Ullah et al., 2017; Wong et al., 2022). The MRL value of Cd was Cd: 0.1 μg kg^{-1} day^{-1} (ATSDR, 2024).

The Hazard Index (HI) was calculated assuming a cumulative health risk posed by multiple TTEs in a mixture having synergistic, additive and interactive effects (E.q. 7) (Ullah et al., 2017).

$$EDI = (C \times RCR_{Day})/BW \tag{3}$$

EDI = Estimated Daily Intake (mg kg^{-1} bw^{-1} $person^{-1}$)
RCR_{Day} = Cooked rice consumption per day per person (kg $person^{-1}$)
BW = Body weight (50 – 70 kg weight range respectively)

$$HQ_{RfD} = (EF \times ED \times RCR_{Day} \times C)/(RfD_{Oral} \times BW \times AT) \tag{4}$$

$$HQ_{MRL} = EDI \ / \ MRL \tag{5}$$

HQ = Hazard Quotient
EF = Exposure frequency (in days, considered at daily rice consumption thereby, 365 days per year)
ED = Exposure duration (in years, considered for 70 years' life span for an average adult)
RfD_{Oral} = Oral Reference Dose (considered for each element per mg $kg^{-1}d^{-1}$)
AT = Average time for non-carcinogens (365 days for life expectancy or number of exposure years equivalent to 25,550 days)
MRL = Minimum Risk Level

$$HI = \sum_{i}^{n} HQ \tag{6}$$

HI = Hazed Index
HQ = Hazard Quotient

2.5.3. Carcinogenic risk assessment

The carcinogenic risk assessment was conducted by modelling Incremental Lifetime Cancer Risk (ILCR) facilitated by EDI and USEPA Cancer slope factors for TTEs (Eq. 7).

$$ILCR = EDI \times CSF \tag{7}$$

ILCR = Incremental Lifetime Cancer Risk
EDI = Estimated Daily Intake
CSF = Cancer Slope Factor

The cancer slope factors (CSFs) of TTEs used for the carcinogenic risk assessment were Cd: 15 per mg $kg^{-1}d^{-1}$, As: 1.5 per mg $kg^{-1}d^{-1}$ and Pb: 8.5×10^{-3} per mg $kg^{-1}d^{-1}$ and respectively (Lee et al., 2023; Oni et al., 2022; Sarwar et al., 2020; Taghizadeh et al., 2022; USEPA, 1995). Since Cr(III) was not considered as human carcinogen it has been eliminated from the carcinogenic risk assessment (Health Security Agency- United Kingdom, 2016).

2.6. Data and statistical analysis

Elemental concentrations were reported as mean (± SD) and median (IQR) on milligrams per kilogram dry weight basis (mg kg^{-1} dw) of lyophilized grain powders. After extrapolating with the percentage moisture content, the elemental content was reported on a wet weight basis (mg kg^{-1} ww) as needed for separate analyses. The moisture content at the washed and cooked stages was used to determine the elemental contribution of cooking water, thus reduced appropriately from grain levels for a more precise estimation of the changes in rice grain elements.

Statistical analysis was conducted using statistical software IBM© SPSS 25.0 for Windows®. The graphical representation of information was supported by data visualization software GraphPad Prism© 10.0 for Windows and MS- Excel (Microsoft Corporation© – 2022). Samples that resulted in values lower than the limit of detection (< LOD) were allocated with half of the LOD before proceeding to statistics. After normality testing (Kolmogorov – Simirnov test statistic, $p < 0.05$), outliers above or below three standard deviations for all variables were excluded ($n = 1$) from further statistical comparisons. Data were not transformed. Accordingly, non-parametric test statistics (Mann-Whiteny (U), Kruskal Wallis (H), Wilcoxon signed – rank (W), Kendal's Tau correlation (τ_b)) were used to compare distributions and assess correlations between groups. A P-value of $p \leq 0.05$ was considered statistically significant.

3. Results & discussion

3.1. Elemental profile & quality control

The LOD for Cd, As, Pb and Cr were; 0.005, 0.033, 0.037 and 0.045 mg kg^{-1}, while the LOQ for Cd, As, Pb, and Cr were; 0.045, 0.329, 0.372 and 0.447 mg kg^{-1} respectively. The standard calibration for Cd, As, Pb and Cr showed excellent linearity ($r^2 \geq 0.998$) over 0.5 – 1000 ppb with intra-run precision (<10% RSD) suitable for trace element analysis in rice digests (Table S2). The reported values for Cd, As and Pb for the external quality control material (matrix matched Certified Reference Material: European Commission Institute for Reference Materials and Measurements (IRMM) – 804) were; 1.55 (± 0.04), 0.047 (± 0.004) and 0.40 (± 0.03) mg kg^{-1} dw respectively with 96.3, 95.7 and 94.5% recoveries which were within the certified levels (*k2*, 95% CI) that confirmed the reliability of data.

3.2. Distributions TTEs different rice varieties, correlations and their transition during the cooking process

The distribution and transition of TTEs of the entire cohort at each processing stage are illustrated by truncated violin plots in Fig. 2.

3.2.1. Distribution of TTEs in raw rice grains

The Median (IQR) values for Cd, As, Pb and Cr were; 0.032 (0.062 – 0.008), 0.049 (0.063 – 0.035), 0.095 (0.170 – 0.047) and 0.376 (0.532 – 0.166) mg kg^{-1} ww basis respectively (Fig. 2). From the total samples 12% were <LOD for Cd, 16% for Pb and 8% for Cr respectively. None of the samples were below the LOD for As.

The Cd concentrations measured in the present study (0.032 (0.062 – 0.008) mg kg^{-1}) was consistent with findings reported in literature. The previously reported Cd concentrations in Sri Lankan rice ranged from non-detected to 0.567 mg kg^{-1} (Bandara et al., 2008; Chandrajith et al., 2011; Chandrasiri et al., 2022; Diyabalanage et al., 2016; Jayatilake et al., 2013; Kulathunga et al., 2022; Lockwood et al., 2024; Navarathna et al., 2021; Nyachoti et al., 2022). Particularly, Cd concentrations exceeding 0.5 mg kg^{-1}, in Sri Lankan rice samples have been highlighted by several researchers, both in global sampling cohorts (Meharg et al., 2013; Shi et al., 2020) and in locally conducted studies (Bandara et al., 2008; Diyabalanage et al., 2016), suggesting that elevated Cd levels are not an isolated occurrence in this context.

In the present study, the median (IQR) As concentration in raw rice grains was 0.049 (0.063 – 0.035) mg kg^{-1} (Fig. 2), which falls within the lower end compared to previously reported range; < 0.05–0.293 mg kg^{-1} for Sri Lankan rice (Chandrajith et al., 2011; Chandrasiri et al., 2022; Diyabalanage et al., 2016; Kulathunga et al., 2022; Lockwood et al., 2024; Nyachoti et al., 2022). Similarly, the median (IQR) Pb concentration measured in the present study (0.095 (0.170 – 0.047) mg kg^{-1}) (Fig. 2), falls within the lower end of the

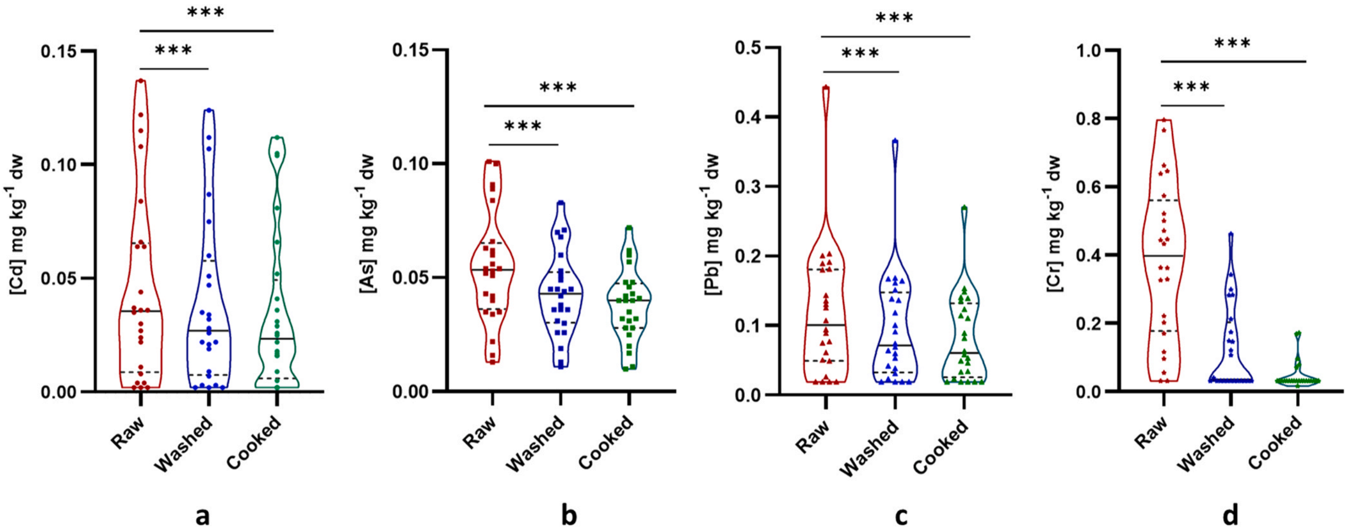

Fig. 2. Transition of toxic trace elements (TTEs) during domestic cooking process, concentrations expressed as milligrams per kilogram dry weight basis (mg kg^{-1} dw): 2.a: Cd, 2.b: As, 2.c: Pb and 2.d: Cr. Lines represent the significant differences between the processing stages, ***significance $p < 0.001$, two-tailed.

previously reported values for Sri Lankan rice, which ranged from 0.06 to 1.47 mg kg^{-1} (Bandara et al., 2008; Chandrajith et al., 2011; Chandrasiri et al., 2022; Diyabalanage et al., 2016; Kulathunga et al., 2022; Lockwood et al., 2024; Nyachoti et al., 2022). The median (IQR) Cr concentration in the present study was 0.0376 (0.532 – 0.166) mg kg^{-1} (Fig. 2), falling within the mid-range of the limited data available in the published literature for Sri Lankan rice, which ranged from < 0.07–1.11 mg kg^{-1}(Chandrasiri et al., 2022; Diyabalanage et al., 2016; Lockwood et al., 2024; Navarathna et al., 2021). The differences observed between the TTE levels of the present study and those previously reported are likely attributable to a combination of factors, including geographical origin, sourcing practices, rice variety and cultivar, and post-harvest processing methods. Agronomic practices and growth conditions specific to the cultivation areas are among the factors that may contribute to these variations.

Excessive agrochemical use in Sri Lankan paddy farming has been widely reported (Jayasumana et al., 2014, 2015), where application frequency estimated to have doubled over the past decade (Dissanayake et al., 2022). Adulteration of agrochemicals with TTEs (Priyashantha & Mahendranathan, 2019) can directly contaminate paddy soils, facilitating TTE uptake by the rice plant. Many paddy farmers in Sri Lanka have been found to deviate from recommended agrochemical compositions and application practices, with heavy reliance on moderately hazardous products not recommended by the WHO (Dissanayake et al., 2022). Elevated levels of Cd, Pb and other trace elements have further been detected in the water and sediments of reservoirs within Sri Lanka's dry agricultural zones (Bandara et al., 2008), which serve as the primary irrigation source for the rice cultivation in these regions during dry periods. In combination, these conditions present plausible pathways for TTE accumulation in rice grains cultivated across Sri Lanka, and are consistent with the varietal and spatial variations observed in the present study.

3.2.1.1. Distribution of TTEs according to pericarp colour. In the current study, the Cd and As concentrations were higher in red pericarp grains compared to white pericarp grains, although this difference was statistically non-significant ($U_{Cd,\ As}$, $p > 0.05$) (Fig. S1). Conversely, the Pb and Cr concentrations were higher in white pericarp grains compared to red, again without a statistical difference ($U_{Pb,Cr}$, $p > 0.05$) (Fig. S1). The cumulative TTE concentrations were also higher in red pericarp grains, though this difference did not reach a statistical significance ($U_{\Sigma TTE}$, $p > 0.05$). The trend of elevated Cd and As levels in red rice varieties is consistent with previously reported findings for Sri Lankan rice (Chandrasiri et al., 2022; Diyabalanage et al., 2016), and may be attributed to the higher affinity of TTEs to accumulate within the rice bran (Jo & Todorov, 2019), which is largely retained during the milling and polishing process of red rice varieties.

3.2.1.2. Distribution of TTEs according to rice categories. The TTE distribution is the present study among the three rice categories investigated; Traditional, Improved and Imported, did not show a significant inter- categorical variation ($H_{Cd,As,Pb,Cr}$ $p > 0.05$) (Fig. S2). Among categories, traditional varieties accumulated higher concentrations of Cd and Pb, while Imported varieties showed elevated As levels and Improved varieties showed higher Cr concentrations, although none of these differences showed a statistical significance ($p > 0.05$). Within the Traditional category specifically, the highest Cd concentration was observed in the red pericarp *Pachchaperumal (Siyapath-el)* variety, followed by *Suwandel* and *Kaluheenati.*

These findings are compatible with previously reported trends in literature. According to Lockwood et al. (2024) higher Cd accumulation in the Traditional variety *Suwandel* which is reflective of the increased Cd speciation within the edible endosperm upon uptake (Gu et al., 2020). Similarly, Chandrasiri et al. (2022) reported higher levels of all TTEs in Traditional rice compared to Improved and Imported varieties, while Diyabalanage et al. (2016) reported higher As in Improved rice and higher Pb in Traditional rice, collectively highlighting the varietal differences in the distribution patterns of each TTE.

The higher TTE accumulation observed in the Traditional varieties in the present study may be attributed to several factors. First, the majority of the Traditional rice varieties investigated were red rice, which, is associated with higher TTE accumulation due to intact bran as previously discussed. Second, Traditional varieties grown in Sri Lanka have considerably longer growth cycles (5 – 6.5 months) compared to Improved varieties (3 – 3.5 months), resulting in a prolonged soil – plant transmission period that may facilitate a greater TTE uptake. Additionally, organic cultivation practices commonly associated with Traditional rice farming, including the use of potentially contaminated organic manure and fertilizes which may further contribute to TTE release in to the paddy soil (Navarathna et al., 2021).

3.2.1.3. Distribution of TTEs in parboiled and non – parboiled rice. In the present study, parboiled rice showed higher TTE concentrations compared to non-parboiled rice, although the differences were not statistically significant ($H_{Cd,As,Pb,Cr}$, $p > 0.05$) (Fig. S3). A similar trend was observed within the Improved rice category, where parboiled *Samba* and

Nadu varieties showed higher TTE levels compared to non – parboiled *Kekulu* varieties, again without statistical significance ($U_{\sum TTE}$, $p > 0.05$) (Fig. S4). These findings are consistent with Chandrasiri et al. (2022), who reported higher As and Pb levels in parboiled rice, including parboiled imported Basmati and white Samba varieties.

The higher TTE concentrations observed in parboiled rice may be attributed to parboiling process itself. As a post – harvest modification process, parboiling is primarily employed to minimize grain breakage during milling and polishing (Bhattacharya, 1996; Muchlisyiyah et al., 2023) and to protect the grains from pests and mould formations (Elbert et al., 2001). The core process involves soaking, heat treatment through direct boiling and/ or steaming followed by drying that has been shown to promote migration of elements and nutrients from the rice bran in to the endosperm (Akhter, 2023). This phenomenon forms the basis for the generally recognized nutritional superiority of parboiled rice over non – parboiled rice (Akhter, 2023; Thennakoon & Ekanayake, 2022). Moreover, the use of contaminated water during the soaking and hydration steps of the parboiling process itself represents a further potential pathway for TTE introduction into the rice endosperm.

According to consumption surveys of Sri Lanka, the rural populations predominantly tend to consume non-parboiled rice (commonly referred to as; *Kekulu*/ country rice), while urban populations are more likely to consume parboiled rice. (Department of Census & Statistics, Ministry of Economic Policies & Plan Implementation, 2019; Department of Census & Statistics, Ministry of National Policies & Economic Affairs Sri Lanka, 2016; Department of Statistics & Ministry of Finance & Planning Sri Lanka, 2010). Given that parboiled rice was found in the present study exhibiting higher TTE accumulation compared to non – parboiled rice (although statistically non – significant), urban consumers may be at a comparatively greater risk of dietary TTE exposure than their rural counterparts, solely on the basis of habitual rice consumption patterns.

3.2.2. Correlations of TTEs in raw rice grains

The correlations between TTEs were analyzed according to Kendal's Tau correlation statistic (Table S6). The distribution of Cd in raw rice grains in the current study showed a weak, statistically non – significant, negative correlation with that of As levels ($\tau_{b\ \mathrm{Cd:As}} = -0.228$, $p > 0.05$). However, the Cd levels showed a moderate positive correlation to that of Pb values which was statistically significant ($\tau_{b\ \mathrm{Cd:Pb}} = 0.370$, $p < 0.05$) and a weak, non-significant, positive correlation to that of Cr levels respectively ($\tau_{b\ \mathrm{Cd:Cr}} = 0.092$, $p > 0.05$).

The observed negative correlation between Cd and As levels in the present study ($\tau_{b\ \mathrm{Cd:As}} = -0.228$, $p > 0.05$) (Table S6) can be attributed to contrasting effects of paddy soil water management on the bioavailability and translocation of these two elements. Flooding conditions alter the soil redox potential in a manner that simultaneously increases As bioavailability facilitated through the reduction of As(V) to more mobile As(III) and dissolution of iron oxides, the primary soil absorbent of As (Nickson et al., 2000; Ohtsuka et al., 2013; Zhao et al., 2010). This process simultaneously suppresses the Cd uptake through the formation of insoluble sulfide and carbonate compounds (De Livera et al., 2011; Khaokaew et al., 2011; Kosolsaksakul et al., 2014). Such trade-off relationships have been reported in both Japanese (Arao et al., 2009; Honma et al., 2016) and Chinese rice (Duan et al., 2017; Hu et al., 2013), and are further supported by the genotypic variations in root – to – grain translocation mobilities of Cd and As (Duan et al., 2017; Limmer & Seyfferth, 2022; Luo et al., 2022). This further complicates the breeding strategies aimed simultaneously reducing the accumulation of both elements. Notably, OsNRMP5 (gene that encodes for a natural resistance-associated macrophage protein, a key transporter of divalent metal ions into rice plants) mutants grown under aerobic conditions have shown promise in reducing grain accumulation of both Cd and As simultaneously (Ishikawa et al., 2012), highlighting the potential of integrated water management and breeding strategies as mitigation approaches.

The observed significant positive correlation between the elements Cd and Pb ($\tau_{b\ \mathrm{Cd:Pb}} = 0.370$, $p < 0.05$) (Table S6) was compatible with works by previous scholars (Liu et al., 2020; Shakerian et al., 2012). This trend can be attributed to their shared geochemical origin, similar soil – to – plant transfer mechanisms and overlapping translocation pathways. Cd and Pb has a common occurrence in earth's crust (Li et al., 2024). Multivariate analyses of paddy soils have demonstrated strong positive correlations between the two elements that also reflected similarly in the grain levels, further suggesting synergy between their uptake mechanisms (Tariq & Rashid, 2013). A large – scale field study encompassing over 260 rice accessions confirmed a statistically significant positive correlation between grain Cd and Pb, while also identifying environmental factors and genetic predispositions of rice subpopulations as key determinants of TTE accumulation patterns (Liu et al., 2019). Notably, the *Indica* subpopulation, which broadly represents Sri Lankan rice has been associated with elevated levels of Cd, Pb and As attributed to distinctive quantitative genetic loci that influence both soil – plant TTE transition and speciation within endosperm (Liu et al., 2019). It has been discovered that there are two primary determinants of Cd and Pb accumulation in rice grains; factors governing root – level accumulation and those controlling root – to – grain translocation (Li et al., 2024), both of which are amenable to targeted intervention through hybridization strategies aimed at developing rice varieties with lower TTE affinities.

3.2.3. Elemental concentrations in washing and cooking water

The concentrations of TTEs in Colombo Municipal Council water ($n = 11$) that was used for the washing and cooking of raw rice grains were; Cd: <LOD, As: <LOD, Pb: 0.005 and Cr: 0.001 mg L^{-1} respectively. The levels of TTEs in water were below the guideline limits for drinking water standards (US EPA, 2015). The impact of cooking water on the variation and distribution of TTEs was therefore considered negligible.

3.2.4. Distribution and transition of TTEs during the washing and cooking stages

The concentrations of TTEs; Cd, As, Pb and Cr at each stage of the cooking processes are depicted in Fig. 2. There was a significant reduction of all TTE levels, during raw to wash and raw to cook transformations ($W_{\mathrm{Cd,\ As,\ Pb,\ Cr}}$, $p < 0.001$). Following washing, 12% of rice samples were < LOD for Cd, 16% for Pb, and 52% for Cr. All samples retained detectable levels of As. After cooking, the proportions <LOD increased to 20% for Cd and Pb, and 72% for Cr, with As remaining detectable in all samples.

The observed significant reduction of all TTEs in this study during the domestic cooking processes (Fig. 2) suggest that household processing methods play a determining role in the reduction of dietary TTE exposures from rice consumption.

Many previous studies have demonstrated the impact of washing and rinsing of rice on decreasing the levels of TTEs in cooked rice grains (Fakhri et al., 2018; Khan et al., 2010; Sharafi et al., 2019; Shariatifar et al., 2020), particularly in polished rice (Gray et al., 2015). Washing rice until water runs clear was sufficient to decrease As levels, but not Cd (Khan et al., 2010). Washing rice for five times have shown to remove maximum amount of TTEs in rice (Shahriar et al., 2022).

Fernando et al. (2020) reported reductions in Cd, Cr, and Pb in Sri Lankan rice during cooking and are further supported by studies conducted on other rice varieties globally (Abu-Almaaly, 2020; Liu et al., 2018; Menon et al., 2021; Sharafi et al., 2019). However, data specifically examining the TTE changes during cooking of Sri Lankan rice varieties remain scarce in published literature. The extent of TTE reduction during cooking has been shown to be influenced by the water: rice ratio employed. Cooking with a water: rice ratio of ~2:2, similar to what was used in the present study has been reported to results in no significant change in Cd and As, while cooking with excess water ratios (6:1 or 10:1) has been shown to moderately reduce Cd (by 10–15%) and As (by 30 – 40%) (Gray et al., 2015). It is noteworthy, however, that the use of contaminated water during cooking could potentially offset these reductions through a net addition of TTEs to the cooked grain, underlying the importance of water quality as a compounding factor in dietary TTE exposure assessments.

3.2.4.1. Reductions of TTEs between pericarp colours. The % Reduction of Cd and Cr during washing step in this study was higher in red pericarp varieties, while As and Pb reductions were greater in white pericarp grains (Fig. 3). None of these differences however, were statistically significant ($p > 0.05$). During cooking, TTE retention was higher in red rice compared to white rice investigated in this study for all elements except As, with a statistically significant difference observed only for Pb (U_{Pb}, $p < 0.05$) (Fig. 3.c). The element with highest reduction, irrespective of the pericarp colour was Cr (Fig. 3.d).

These pericarp - dependent differences in TTE reduction observed here can be partly explained by the spatial distribution of elements within the rice grain. As and Pb are predominantly concentrated in the outer bran and germ layers, while Cd is more uniformly distributed throughout the grain (Lockwood et al., 2024). In white rice, the bran is largely removed during polishing and further dislodged during washing, which accounts for the greater percentage reduction of As and Pb observed in white pericarp varieties in the present study. In contrast, red rice retains its bran layer due to lower polishing degrees, which on one hand limits the removal of As and Pb during washing, but on the other hand results in higher overall TTE retention through the cooking process. This effect is further compounded in the present study by the fact that the majority of red pericarp varieties were parboiled, Traditional and Improved types, in which the parboiling process promotes migration of elements into the endosperm, making them less susceptible to removal during subsequent washing and cooking steps.

3.2.4.2. Reduction of TTEs between different rice categories. Present study findings reveal that the median % Reduction of Cd, during the washing step decreased in the order of Traditional rice > Improved rice > Imported rice (H_{Cd}, $p > 0.05$) (Fig. 4.a). However, the % Reduction of As was higher in Imported rice followed by Improved rice and Traditional rice with a significant inter-categorical variation (H_{As}, $p < 0.05$) (Fig. 4.b). The % Reduction of Pb decreased in the order of Traditional rice > Imported rice > Improved rice (H_{Pb}, p > 0.05) (Fig. 4.c). The Cr % Reduction was higher in the Imported rice (H_{Cr}, $p > 0.05$) (Fig. 4.d). Overall, the Traditional varieties showed the highest potential for cumulative TTE retention during the domestic cooking process followed by Improved and Imported varieties ($H_{\Sigma TTE}$, $p > 0.05$).

The observed differences on TTEs reduction between the rice categories could be attributed to the processing of raw rice grain including polishing (Lockwood et al., 2024). Majority of traditional varieties consumed in Sri Lanka comprise of intact bran and germ that can be dislodged during the washing step resulting a higher loss of TTEs from the raw rice grains. On the other hand, the rice that belongs to the Improved category usually undergoes various degrees of polishing that may lead to leaching of TTEs that were accumulated in the endosperm.

3.2.4.3. Reductions of TTEs between parboiled and non – parboiled rice. Parboiled varieties retained higher amounts of TTEs during the domestic cooking process compared to non-parboiled rice ($U_{\Sigma TTE}$, $p > 0.05$) (Fig. 5). The % Reductions of Cd and As specifically were significantly higher in non-parboiled rice ($U_{Cd,As}$, $p < 0.01$) during the processing stages (Fig. 5.a and 5.b). Although the % Reductions of Pb and Cr were also higher in non – parboiled, the differences were not statistically significant ($U_{Pb,Cr}$, $p < 0.05$) (Fig. 5.C and 5.d).

Since the parboiling treatment promotes the migration of elements from bran to the endosperm (Akhter, 2023), the TTEs are not easily removed during the washing stage (Muchlisyiyah et al., 2023). Moreover, the parboiling process induce structural changes including gelatinization (Muchlisyiyah et al., 2023) which may lead to reduce leaching of elements from the matrix.

This trend is also visible within the Improved rice category; where Parboiled *Samba* and *Nadu* rice showing significantly lower % Reductions of Cd and As (Fig. 6.a and 6.b) compared with Non – parboiled *Kekulu* rice ($U_{Cd,As}$, $p < 0.01$). However, the % Reduction of Pb during the cooking process was similar between both *Kekulu* and *Samba* rice, which was ~ twice that of *Nadu* rice (H_{Pb}, $p > 0.05$) (Fig. 6.c). Over 90% reduction of Cr was observed for *Samba* and *Nadu* respectively (Fig. 6.d), which was slightly higher than the *Kekulu* rice (H_{Cr}, $p > 0.05$).

In a recent study (David et al., 2020), an introduction of a parboiling process immediately before cooking of rice grains have shown a significant increase in selected trace elements that included Cr, Aluminum (Al) and Nickle (Ni) etc. while significantly increased the TTEs of Cd, As and Pb compared to non – parboiled control. On the contrary, Goh et al. (2024) reported that parboiling before cooking of rice has reduced the availability of TTEs (>77%), specifically reducing Cd, As and Pb levels below the maximum toxicological safe limits/ permissible values (Goh et al., 2024). These differences may be attributed to several factors, including Goh et al. (2024)'s method of parboiling of pre-husked/polished rice in excess deionized water (1:4 ratio, 100°C, discarded) promoting leaching via high temperature and osmosis, differing polishing rates, varietal differences between Malaysian and Sri Lankan rice. Moreover, Sri Lankan parboiling process use husk-intact grains and non-deionized water prior to milling; usually by immersing raw paddy in boiling water until the husk is slightly split open, followed by sun drying before hulling and polishing (Thennakoon & Ekanayake, 2022).

3.3. The role of average cooking time on the TTE transition during wash to cook transformation

The average cooking time (for a standard 100 g portion of raw rice) in this study showed statistically non – significant negative correlation with the % reduction of TTEs; Cd ($Ʈ_b = -0.171$, $p > 0.05$), As ($Ʈ_b = -0.160$, $p > 0.05$) and a moderate significant negative correlation with that of Pb ($Ʈ_b = -0.317$, $p < 0.05$) while showing a statistically non – significant positive correlation with that of Cr ($Ʈ_b = 0.044$, $p > 0.05$).

The negative correlation observed between cooking time and Pb reduction may partly explain the lower percentage reduction of Pb observed in parboiled rice varieties, which generally require longer cooking times than their non-parboiled counterparts (Meresa et al., 2020).

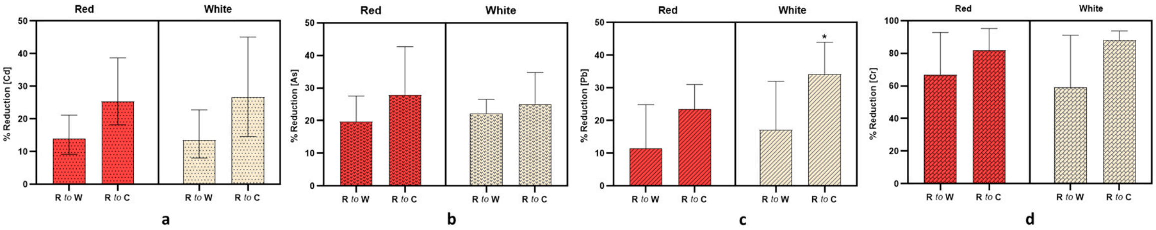

Fig. 3. The median percentage reductions (% Reduction) of toxic trace element (TTE) concentrations in the pericarp colours; Red and White during the processing stages; R *to* W: Raw to Wash and R *to* C: Raw to Cook: 3.a: Cd, 3.b: As, 3.c: Pb and 3.d: Cr., *significance $p < 0.05$, two-tailed.

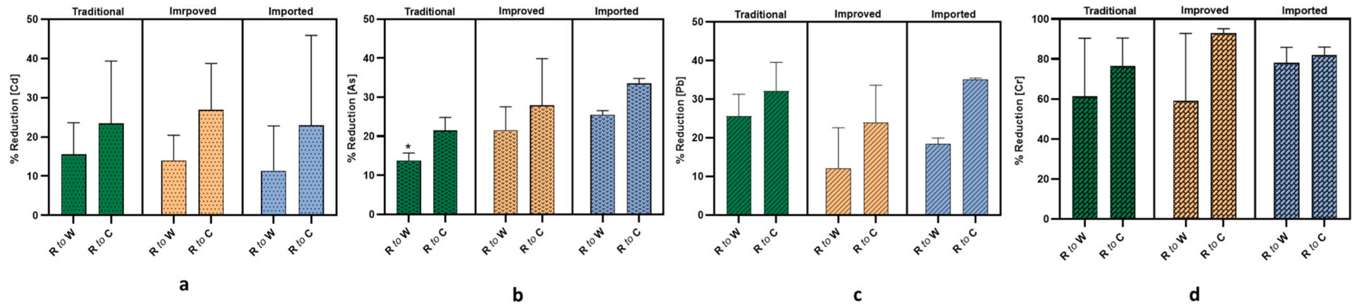

Fig. 4. The median percentage reductions (% Reduction) of toxic trace element (TTE) concentrations across different rice categories; Traditional, Improved and Imported during processing stages; R *to* W: Raw to Wash and R *to* C: Raw to Cook: 4.a: Cd, 4.b: As, 4.c: Pb and 4.d: Cr, *significance $p < 0.05$, two-tailed.

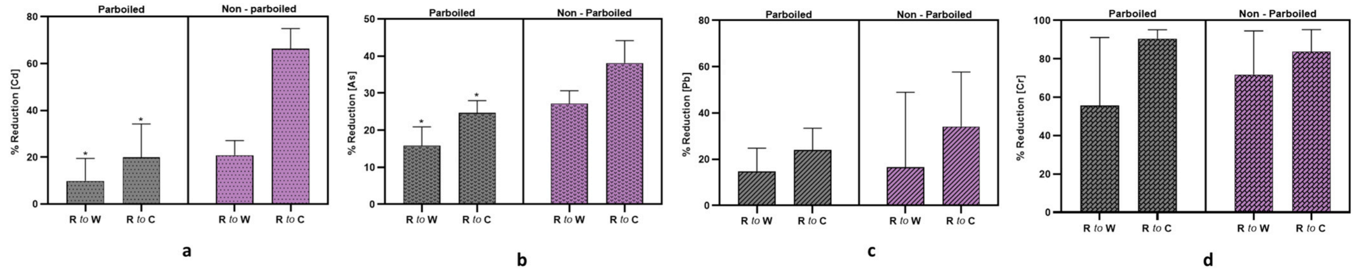

Fig. 5. The median percentage reductions (% Reduction) of toxic trace element (TTE) concentrations according to parboiling treatment of rice; parboiled and non-parboiled, during processing stages; R *to* W: Raw to Wash and R *to* C: Raw to Cook: 5.a: Cd, 5.b: As, 5.c: Pb and 5.d: Cr, *significance $p < 0.05$, two-tailed.

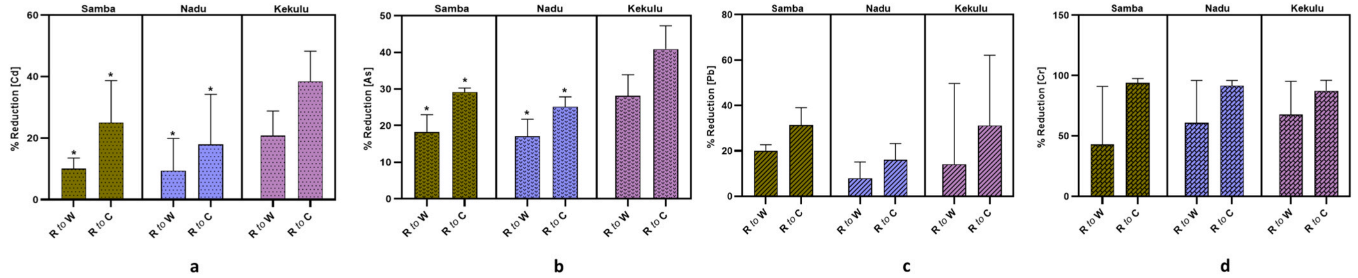

Fig. 6. The median percentage reductions (% Reduction) of toxic trace element (TTE) concentrations across Improved category rice types; Samba, Nadu and Kekulu during processing stages; R *to* W: Raw to Wash and R *to* C: Raw to Cook: 6.a: Cd, 6.b: As, 6.c: Pb and 6.d: Cr, *significance p < 0.05, two-tailed.

Comparable data on the relationship between cooking time and TTE reduction in rice are scarce in the published literature, likely reflecting the considerable variability in cooking methods, thermal processes (cooked directly using pots, use of rice cookers, pressure cooking and the use of a microwave etc.) and vessel materials (aluminum, steel, clay etc.) employed across studies, which complicates direct comparison. It is also noteworthy that the majority of TTEs are thermally stable and therefore, remain largely within the cooked grain regardless of the cooking duration.

3.4. Risk assessment for TTE intake by Sri Lankan consumers

3.4.1. Rice consumption rates of Sri Lankan adults

The derived mean cooked rice consumption was 0.918 (± 0.092) kg with a lower bound intake (10^{th} percentile) of 0.800 kg and an upper bound intake (90^{th} percentile) of 1.124 kg, considered uniformly across 50, 55, 61.4 and 70 kg weight categories. Therefore, per weekly and monthly cooked rice consumption was estimated to be 6.426 (5.600 – 7.868) kg $person^{-1}$ and 27.540 (24.000 – 33.720) kg $person^{-1}$ respectively.

The recommended daily intake of starch-based foods, including cooked rice, for an average Sri Lankan adult is 0.520 – 0.845 kg (Ministry of Health Sri Lanka, 2021), suggesting that the actual rice consumption of a typical Sri Lankan adult exceeds the recommended dietary guidelines. This pattern of high consumption is well established; it has been reported that an average Sri Lankan adult male has historically consumed approximately 0.9 kg of cooked rice per day, equivalent to an annual intake of 329 kg of cooked rice or approximately 110 kg of raw rice (Jayatissa et al., 2014). The current per capita raw rice consumption of a typical Sri Lankan adult is estimated at 110 – 120 kg per year, placing Sri Lanka among the ten highest rice-consuming nations globally. In context, the daily rice consumption of Sri Lankans is approximately twice the global average (Kadupitiya et al., 2022; Xu et al., 2020) and twenty-two times that of the average European consumer (Xu et al., 2020), underscoring the particular significance of rice as a vehicle for dietary TTE exposure in the Sri Lankan population.

3.4.2. Toxicological guideline limits for commercial commodity – raw rice

3.4.2.1. Maximum permissible values (JECFA – MLs) and country-specific thresholds. The MLs established by the WHO/JECFA are available for Cd (rice-whole commodity), As (rice-polished) and Pb (cereal grains) at 0.4, 0.35 and 0.2 mg kg^{-1} respectively. No ML has yet been established for Cr.

Japan and India currently follow the Codex standard for Cd, whereas China and New Zealand have adapted a stricter limit of 0.1 mg kg^{-1}. While China has not set a maximum limit for total As species, it enforces a limit of 0.2 mg kg^{-1} for inorganic As, which is considerably lower than the codex maximum level. India has not specified a limit for As species in rice, however regulates all food allowed at a maximum limit of 1.1 mg kg^{-1}. On the contrary, New Zealand maintains a higher threshold level at 1.0 mg kg^{-1}. While adapting the same ML for Pb, China has also introduced a threshold for Cr at 1.0 mg kg^{-1}. The New Zealand threshold for Pb stands at 0.2 mg kg^{-1} and applies to all cereals while India allows a much higher margin of 2.5 mg kg^{-1} for food items not specified.

None of the raw rice grain samples from the present study exceeded the WHO/JECFA MLs for Cd or As. However, 20% exceeded the country specific thresholds by China and New Zealand 0.1 mg kg^{-1} highlighting the potential for chronic dietary exposure to Cd. Moreover, one sample exceeded the WHO/JECFA Pb ML at 0.2 mg kg^{-1}. None of the samples exceeded the country specific Cr allowable limit of 1.0 mg kg^{-1} by China.

Many studies have highlighted that the current permissible threshold values are not appropriate to protect high consuming populations from health effects (Samarajeewa, 2022; Satarug et al., 2010; Satarug, 2018; Wei et al., 2023; Yang et al., 2023). A recent study in China reported that the current health limits are, not only inadequate to safeguard its consumers from exposure to TTEs through rice consumption, but also could induce health effects that are non-negligible (Wei et al., 2023). For instance, prolonged/ chronic low grade exposure to Cd, As, and Pb have been implicated in many chronic diseases, including hypertension and neurological disorders (Balali-Mood et al., 2021; Mitra et al., 2022).

Certain food crops have higher affinities to particular TTEs, known as 'hyper-accumulators', which pose an increased exposure threat (Meharg et al., 2013; Satarug, 2018). According to the U.S. National Health and Nutrition Examination Survey (NHANES) 2011–2012, several Asian subpopulations were highlighted for the highest levels of blood Cd attributed to the consumption of high TTE accumulating food, frequently in larger quantities (Awata et al., 2017).

3.4.3. Toxicological guideline limits for consumption profile – cooked rice

3.4.3.1. Estimated daily intakes (EDIs), provisional tolerable weekly intakes (JECFA - PTWI), tolerable weekly intakes (EFSA-TWI), provisional tolerable monthly intakes (JECFA - PTMI). The EDI values of Cd, As, Pb and Cr calculated based on the cooked rice consumption patterns by adults under different weight categories are summarized in Table 1. Sensitivity analyses validated ½ LOD substitution (against two alternative substitution scenarios; zero and full LODs) respectively for Cd (20% non-detects, CV=1.27%), Pb (16%, CV=3.72%), and Cr (72%, CV=38.5%) for the EDI calculations for exposure estimation purposes (Table S4, S5). All CVs fall within acceptability thresholds (<100% for >70% and <80% censored data) (Antweiler & Taylor, 2008; EFSA, 2010). The substitution with ½ LOD, EFSA's recommended approach (EFSA et al., 2018), confirmed stable central tendency measures across censoring levels.

The WHO/JECFA – PTMI value for Cd is 25 μg kg^{-1} per body weight (bw) is set considering 70 kg adult. EFSA has established a more rigorous TWI limit at 2.5 μg kg^{-1} bw^{-1} for Cd. The previously established PTMI for Pb at 25 μg kg^{-1} bw^{-1} has been withdrawn without further recommendations. Moreover, the WHO/JECFA – TWI of 15 μg kg^{-1} bw^{-1} (equivalent to 2.1 μg kg^{-1} bw^{-1} day^{-1}) for inorganic As has also been withdrawn as new epidemiological evidence suggests cancer risks at a lower benchmark dose level, thus concluding the existing PTMI to be non – health protective.

The daily, weekly and monthly dietary Cd exposure between different weight categories and rice intake patterns are depicted in Fig. 7; a. b and c respectively. With current Cd concentration in cooked rice grains and average rice intake levels, none of the samples in investigated in the current study had the potential to exceed the JECFA PTMI of 25 μg kg^{-1} bw^{-1} for 70 kg adult (Fig. 7.c). However, 8.3% of samples exceed the limit under 61.4 kg weight category and 12.5% under 50 kg and 55 kg weight categories. At a higher rice intake, the number of samples exceeding the PTMI increased to 14.4% for the 50 and 55 kg categories and up to 12.5% for the 61. 4 kg and 70 kg weight categories respectively.

Table 1
Estimated Daily Intake (EDI) values for TTEs from rice consumption (whole cohort) with different intake patterns and weight categories.

Weight category (kg)	EDIs		
	Rice Intake: mean (± SD)		
	Cd (μg kg^{-1})		
	Low	**Average**	**High**
50	0.290 (± 0.298)	0.332 (±.342)	0.407 (± 0.419)
55	0.263 (± 0.271)	0.302 (± 0.311)	0.370 (± 0.381)
61.4	0.236 (± 0.243)	0.271 (± 0.279)	0.331 (± 0.341)
70	0.207 (± 0.213)	0.237 (± 0.245)	0.291 (± 0.300)
	As (μg kg^{-1})		
50	0.320 (± 0.137)	0.365 (± 0.157)	0.448 (± 0.193)
55	0.289 (± 0.123)	0.330 (± 0.143)	0.406 (± 0.174)
61.4	0.259 (± 0.111)	0.297 (± 0.126)	0.365 (± 0.157)
70	0.226 (± 0.097)	0.260 (± 0.111)	0.320 (± 0.137)
	Pb (μg kg^{-1})		
50	0.689 (± 0.535)	0.790 (± 0.616)	0.969 (± 0.753)
55	0.626 (± 0.487)	0.718 (± 0.560)	0.879 (± 0.682)
61.4	0.561 (± 0.436)	0.643 (± 0.500)	0.787 (± 0.614)
70	0.492 (± 0.382)	0.565 (± 0.439)	0.693 (± 0.538)
	Cr (μg kg^{-1})		
50	0.405 (± 0.357)	0.466 (± 0.410)	0.570 (± 0.501)
55	0.369 (± 0.325)	0.423 (± 0.372)	0.518 (± 0.457)
61.4	0.331 (± 0.291)	0.379 (± 0.333)	0.465 (± 0.407)
70	0.290 (± 0.255)	0.333 (± 0.292)	0.407 (± 0.358)

The weekly Cd intake upon consumption of the studied rice samples is illustrated in Fig. 7.b. At an average rice consumption, 25% of the samples showed a potential to exceed the EFSA – TWI of 2.5 μg kg^{-1} bw^{-1} for 70 and 61.4 kg adult weights and 29.2% and 33.3% for 55 and 50 kg weight categories respectively. A low rice intake did not lower the risk of exposure for 61.4 kg adults, but reduced the exposure risk by 7.1% for 55 and 70 kg weight categories and by 14.3% for the 50 kg weight category. On the contrary, a higher rice consumption yielded a 5.9%, 6.7% and 14.3% increase in samples with the potential to exceed EFSA-TWI. These results demonstrate that people with lower body weights who consume rice in higher quantities are at a higher risk of dietary intoxication with Cd.

3.4.4. Non – carcinogenic exposure risk

3.4.4.1. Tolerable daily intakes (TDIs), interim reference levels (IRLs) and toxicological reference values (TRVs). A recommended daily intake level < 0.06 mg for Cd for 70 kg adult which is equivalent to; 0.886 μg kg^{-1} bw^{-1} day^{-1} has been shown to be kidney protective (Satarug et al., 2017; Ullah et al., 2017). In the current study, 12.5% of the samples had the potential to exceed this limit at average rice intake by an adult representing either 50 or 55 weight categories, while no threat was apparent for at 61.4 kg and 70 kg weight categories. At a lower rice intake, 12.5% of the samples analyzed showed and EDI above the threshold at 50 kg category. With a higher rice intake, 16.7% of the samples showed exceeding EDI values for both 50 and 55 kg weight categories while 12.5% showed exceeding EDIs under 61.4 and 70 kg respectively.

The epidemiological evidence stresses that a daily Cd intake of 16 – 32 mg day^{-1} by an average adult aged 54 yrs with a body weight of ~67.24 kg has been shown to be associated with a 25 – 94% increased risk of breast cancer (Julin et al., 2012; Satarug et al., 2017). This level is equivalent to 0.357 μg kg^{-1} bw^{-1} day^{-1}, reflective of EFSA guidelines

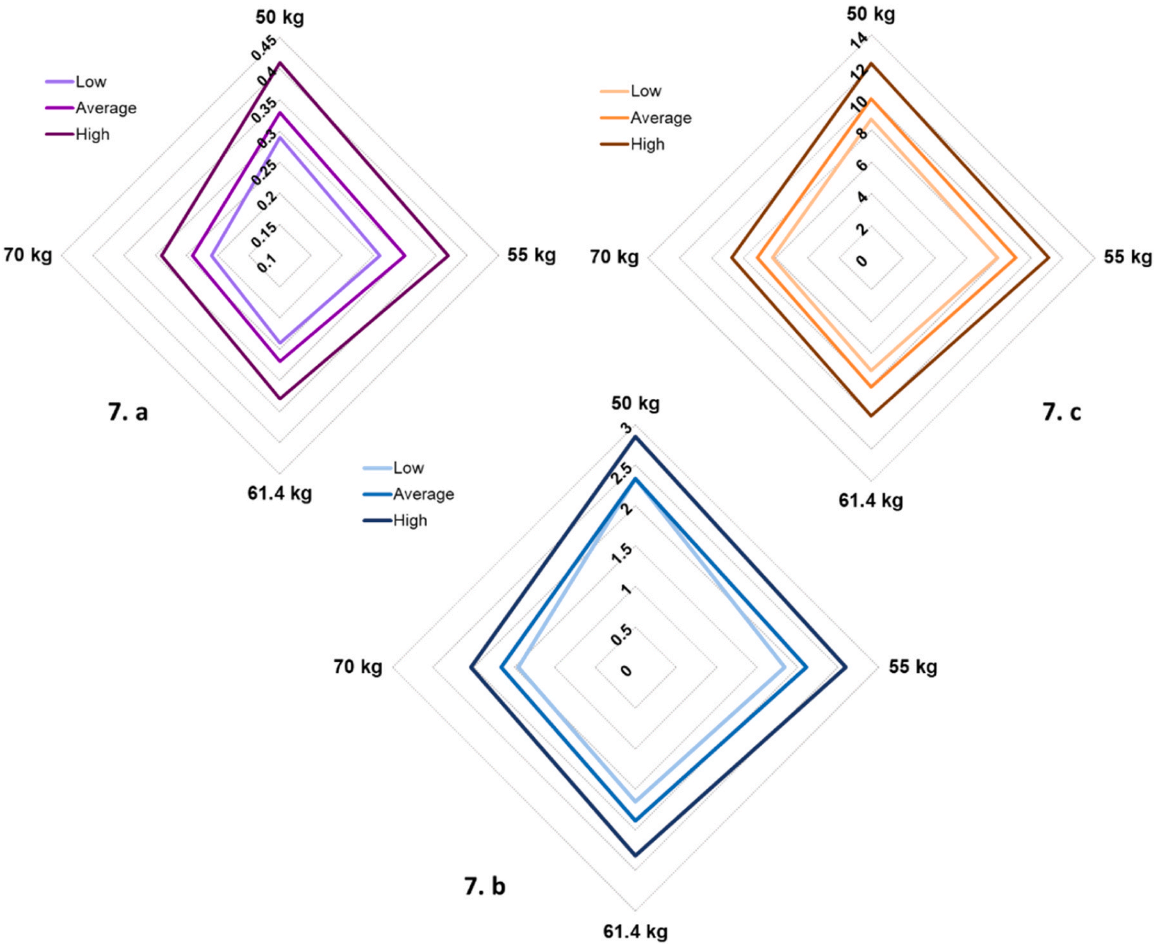

Fig. 7. Daily, weekly and monthly mean Cd exposure estimations micrograms per kilogram of bodyweight (μg kg^{-1} bw) for different body weight categories; 50, 55, 61.4 and 70 kg (axes) and upon rice consumption rates; Low, Average and High (coloured lines): 7.a: estimated daily Cd intake, 7.b: weekly Cd intake, 7.c: monthly Cd intake.

thus, reinforces the need of much lower levels of toxicological limits to fully safeguard the consumers against exposure related adverse health effects.

The EFSA Tolerable Daily Intake (TDI) for Cd is 0.36 μg kg^{-1} bw^{-1} day^{-1} (derived from EFSA –TWI)(EFSA, 2012), which is exceeded by 25% of samples under 61.4 kg and 70 kg weight categories at an average rice consumption (Fig. 7.a). A higher rice intake did not increase the risk for a 70 kg adult. However, having a body weight of 61.4 kg showed a potential exceedance by an additional 4%.

The U.S Food and Drug Administration (FDA) has established a Toxicological Reference Value (TRV) for dietary Cd exposure at 0.21 – 0.36 μg kg^{-1} bw^{-1} day^{-1} (Schaefer et al., 2023), having an upper cut off limit reflective of the EFSA-TDI. A total of 25.0% and 33.3% of the samples in the current study resulted in exposures that exceeded the lower TRV under 70 kg and 61.4 kg weight categories respectively, considered unsafe for consumption and a violation of FDA food safety regulations. Having a higher rice intake resulted in an additional 4.2% and 8.3% increase in the samples with the potential to exceed the TRV.

The EDI for As upon consumption of the investigated rice samples is summarized in Table 1. The TDI - As is 2–7 μg kg $^{-1}$ bw^{-1} (WHO, 2019), which is equivalent to ~ 0.32 mg kg^{-1} for 70 kg adult. However, some studies have suggested much lower allowable limits; 0.13 mg kg $^{-1}$ per person (Ullah et al., 2017). None of the samples from the current study exceed these thresholds, thereby demonstrating that the consuming population is not at risk of dietary exposure to As. Currently there is no established TRL value by FDA for As to comprehend a further evaluation. The FDA has established a specific code of practice in focus of rice producing countries to avoid and minimize the presence of As in rice to safeguard consumers (JECFA, 2017). The EFSA has recently updated the previously established reference value of 3 – 8 μg kg $^{-1}$ bw^{-1} to represent a much lower value at 0.06 μg kg $^{-1}$ bw^{-1} (EFSA-CONTAM et al., 2024). All samples from the current study either matched or exceeded this threshold. However, it should be noted that the above reference value is set specifically for inorganic As, which was the lowest dose to prevent the incidence of skin cancer. Therefore, the current study treats this toxicological limit as too conservative a guideline to evaluate non – cancerous health effects evaluation for exposure to total As without speciation.

There is no established recommended limit for the daily intake of Pb by JECFA. The previously established PTWI value of 25 μg kg^{-1} bw^{-1} which was equivalent to ~3.6 μg kg $^{-1}$ bw^{-1} day^{-1} was withdrawn due to a reduction in intelligence quotients in children and an increase of systolic blood pressure in adults, surfaced from new epidemiological studies (JECFA, 2023). The EU guidelines offer TDI value of 0.2 mg kg^{-1} for a 70 kg adult (EU directorate - General Health and Consumer Protection, 2004; Ullah et al., 2017) which is equivalent to 2.9 μg kg $^{-1}$ bw^{-1}. None of the samples from the current study had the potential to exceed these limits at average rice consumption. However, at a higher rice intake, the EDI projected by one sample (4.2%) exceeded the TDI limit for lower weight categories of 50 and 55 kg. Overall, the Pb levels in Sri Lankan commercial rice is therefore, unlikely to cause adverse non – carcinogenic health risks based on average consumption.

Currently, the JECFA Codex Alimentarius does not provide a Provisional Tolerable Daily Intake (PTDI) value for Cr. The U.S. Institute of the National Academy of Sciences has determined a daily adequacy range for Cr(III) from 0.020 to 0.045 mg for adults and adolescents (ATSDR, 2012). However, the ASTDR has not established any TDI values for Cr. The EFSA currently maintains a TDI of 0.3 mg kg $^{-1}$ bw^{-1} for adults that is not associated with any adverse health effects (EFSA Panel on Dietetic Products, Nutrition and Allergies (NDA), 2014). However, some studies have suggested lower values; 0.2 mg kg $^{-1}$ bw^{-1} (Ullah et al., 2017) as a safer dietary threshold. None of the samples from the current study exceeded these toxicological limits indicating no apparent health risk for consumers.

3.4.4.2. Hazard quotients (HQs). The HQ is an estimation of non – carcinogenic health risk posed to a population upon contaminant exposure using a set of standard health / regulatory guidelines i.e. RfD and MRL. A HQ level ≥ 1 indicates a susceptibility of the exposed population to non – carcinogenic health hazards while HQ < 1 indicate that there is no health hazard. However, a HQ > 0.5 could highlight a higher risk potential (Mandour et al., 2021).

The calculated HQ_{RfD} for Cd in the present study did not exceed 1 at average rice consumption across 55 kg, 61.4 kg and 70 kg weight categories, while 12.5% of samples at 50 kg exceeded HQ level of 1. Moreover, 16.7%, 20.8% and 25% samples for 70 kg, for both 55 and 61.4 kg and 50 kg weight categories respectively resulted in HQ_{RfD} Cd > 0.5 indicating that consumers may still be vulnerable to health effects during chronic consumption. At a higher rice intake, 12.5% of samples across 50 – 61.4 kg weight range resulted HQ_{RfD} Cd > 1, while 25% of samples showed HQ_{RfD} Cd > 0.5. At 70 kg category, no samples exceed 1, while 20.8% showed HQ_{RfD} Cd > 0.5.

When the HQ_{MRL} for Cd was calculated at an average rice intake, 68% of the samples exceeded 1 across all weight categories. Therefore, at a more sensitive exposure threshold, the majority of rice samples posed a higher risk of chronic dietary intoxication which may lead to non – carcinogenic, adverse health effects. A lower rice intake decreased the risk of exceedance by 3%, only for the 70 kg category. On the other hand, a higher rice intake did not produce an additional risk of HQ_{MRL} for any weight category.

The MRL of Cd (0.1 μg kg^{-1} day^{-1}) (ATSDR, 2024) is ten times more sensitive than the RfD. The MRL suggests a level of a given chemical compound that a human can ingest or inhale without considerable problem to their health. However, the lack of complete epidemiological studies makes it difficult to link MRL levels with definite disease etiologies. Exceeding the MRL therefore, does not substantiate adverse health effects. Instead, it can be used as an early surveillance tool to monitor sites and routes that may lead to possible health effects during chronic/ prolonged exposures.

All the samples investigated resulted in HQ_{RfD} As > 1 across all weight categories upon all rice intake levels. Results leaved that 66.7%, 58.3%, 50.0% and 37.5% of samples from 50, 55, 61.4 and 70 kg weight categories respectively resulted in EDIs that exceed the HQ_{RfD} As of 1 at an average rice intake. When the rice intake was low, the percentage of samples with EDI resulting HQ_{RfD}As > 1 dropped by ~8.4% for 50 and 55 kg categories, 12.5% for 61.4 kg category followed by 16.7% 70 kg category. On the contrary, a higher rice intake elevated the risk of exceedance to 79.2% for 50 kg category, 66.7% across 55 kg and 61.4 kg categories and 50% for 70 kg weight category.

The HQ_{RfD} Pb > 1 was not exceeded by rice samples consumed at any consumption pattern. At a high intake level, only one sample had resulted EDIs that exceeded HQ_{RfD} Pb > 0.5. The calculated HQ_{RfD} values for Cr were below 0.001 level and posed no threat to consumers. The elevated threshold RfD of Cr, compared to other elements is reflective of the role it plays in human metabolism (ATSDR, 2012).

3.4.4.3. Hazard indices (HIs). An HI ≤ 1 indicates safe levels, while an HI $\geq$ 1suggests a potential for adverse health effects (Price, 2023).

A calculated HI level greater than 1 was observed for 25% for 50 kg, 16.7% for 55 – 61.4 kg and 8.3% for 70 kg weight categories respectively under the average rice intake level. These results indicated a greater risk potential associated with the mixture of four TTEs studies in this study to cause adverse health effects to consumers. At a lower rice intake, the risk of exceedance was reduced by 20% over 50 – 61.4 kg weight range and 33.3% over 70 kg weight category. However, a higher rice intake posed an additional 16% risk of exceeding the HI of 1 for adults in 50 kg category and 33.3% risk for adults who belong to 61.4 – 70 kg weight range but did not add an additional risk for 55 kg weight category.

The % contribution of Cd, As, Pb and Cr for the HI were 51.0%, 18.6%, 30.3% and 0.1%. The observed strong positive correlation between Cd and Pb may be a contributing factor for resulting the higher % contribution to increase the HIs of rice samples.

3.4.5. *Carcinogenic exposure risk: incremental lifetime cancer risk (ILCR)*

The International Agency for Research on Cancer (IARC) has documented Cd and its compounds, As and its inorganic compounds, as group – 1 human carcinogens while Pb and its inorganic compounds as group – 2 A probable human carcinogens that could induce various cancers upon prolong exposures (IARC., 2024).

The calculated ILCR values based on EDI of Cd ranged from 4.98×10^{-3} – 3.56×10^{-3} at an average rice intake between 50 – 70 kg weight range. There was a $\sim 1.12 \times 10^{-3}$ – 0.8×10^{-3} increase in ILCR when the rice intake was higher. The ILCR – As at average rice consumption ranged from 5.47×10^{-4} – 3.91×10^{-4} respectively from 50 to 70 kg and increased to 6.70×10^{-4} – 4.78×10^{-4} when the rice intake was high. The Pb levels in the rice samples analyzed resulted ILCR values ranged from 6.72×10^{-6} – 4.80×10^{-6} across the studied weight range. At a higher rice intake, these values were elevated by 1.5×10^{-6} – 1.07×10^{-6}.

The ILCR values $\leq \times 10^{-6}$ is generally considered as a level with negligible carcinogenic outcome and a value $> \times 10^{-4}$ considered a carcinogenic exposure risk level. In the present cohort, at the exception of Pb, both the Cd and As levels in rice posed a carcinogenic risk resulting ILCR values that were greater than $\times 10^{-4}$. Considering the higher percentage of samples that exceeded this particular threshold, more stringent protocols to monitor the levels of Cd and As in rice are required.

3.5. *Strengths & limitations of the study*

The study employed established Total Diet Study (TDS) methodologies, utilizing a pooling approach to generate representative population-level exposure estimates while maximizing analytical efficiency. To ensure statistical reliability despite a modest sample size ($n = 25$), the research applied non – parametric methods, which are resilient towards skewed distributions and outliers often found in TTE datasets. A primary strength was the evaluation of '*food as consumed*' (cooked rice), addressing a critical gap in literature that predominantly assesses raw commercial grains. Furthermore, the study utilized localized consumption data (~300 g person^{-1} day^{-1}) synthesized from published Sri Lankan dietary surveys. This allowed a more accurate risk assessment than global averages (~128 g person^{-1} day^{-1}) typically used by international regulatory bodies. The research also provided a comprehensive analysis of grain variables, including traditional versus improved varieties, pericarp color, and parboiling treatments, to identify how these factors influence elemental distribution. Finally, by tracking elemental transitions across raw, washed, and cooked fractions, the study offered precise insights into how domestic preparation modifies the toxicological profile of the final meal.

Pooling reduces variability arising from individual sample heterogeneity and supports multi-stage processing (raw, washed, cooked) within practical resource limits. Therefore, it remains a widely accepted practice in dietary exposure assessment and provides a robust estimate of mean contaminant concentrations. The pooling of food items, while efficient for population – level estimates (Lee et al., 2015), can mask within – sample variability and potentially obscure extreme/high - end contaminant values. Additionally, the study did not perform elemental speciation analysis, but considered regional geochemical evidence that total As represents As (III) and total Chromium represents Cr (III). While this is a common practice for estimating bioavailability, it may underestimate the presence of more toxic forms such as hexavalent Chromium (Cr (VI)). The reliance on a synthesized mean for consumption is another limitation, as it may not fully capture high – end consumers or recent shifts toward wheat – based diets in urban populations. Furthermore, as a preliminary attempt, the risk assessment in the current study was focused on adult consumers, which may not accurately reflect the heightened sensitivity and specific risks faced by vulnerable groups such as children. Lastly, the study focused on rice as the single primary vector, potentially overlooking other cumulative exposure routes such as contaminated drinking water or secondary food groups.

4. Conclusions

The findings of this study summarized the availability of TTEs; Cd, As, Pb and Cr in a cohort of highly consumed Sri Lankan rice varieties in both raw and cooked fractions. The results revealed that the levels of Cd and As in raw rice grains were below the JECFA – MLs while one sample exceeded the levels for Pb. However, 20% of the samples exceeded 0.1 mg kg^{-1} ww for Cd, a stringent country specific guideline maintained by several rice consuming countries. Except for As, the reduction of other TTEs during the cooking process were higher in red pericarp rice than white. Traditional/heirloom varieties showed the highest retention of TTEs compared to Improved and Imported varieties and parboiled rice retained higher amounts of TTEs compared to non – parboiled varieties. Although the domestic cooking process significantly reduced the availability of the TTEs in cooked grains, due to high rice consumption, 25% of the samples were found exceeding the EFSA- TDI- Cd while 8.3% for JECFA – PTMI - Cd. The lower body weights and higher rice intakes posed an additional risk of exceedance. The HI > 1 was observed for 16.7% of the samples for a Sri Lankan adult of 61.4 kg, thus indicating the potential of cumulative exposure risk of TTE mixtures. Except for Pb, both Cd and As levels showed carcinogenic risk for average consumers. The higher consumption rates of Sri Lankans need to be considered in order to establish country specific cut off limits for monitoring TTEs in Sri Lankan rice varieties, and to safeguard consumers from potential dietary exposures and consequent adverse health effects.

CRediT authorship contribution statement

Jayani Wathsala Gunawardana: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **Inoka Chinthana Perera:** Investigation, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – review & editing. **Nekadage Don Amal Wageesha:** Conceptualization, Methodology, Resources, Supervision, Visualization, Writing – review & editing. **Chamindri Witharana:** Methodology, Resources, Supervision, Validation, Visualization, Writing – review & editing. **Sameera Anuruddha Gunawardena:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Writing – review & editing.

Funding

This study was funded by University of Colombo Research grants (AP/3/2/2018/SG/18).

Declaration of Competing Interest

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

Acknowledgements

Authors acknowledge the assistance provided by the Rice Research and Development Institute (RRDI)- Bathalegoda - Sri Lanka, National Aquatic Resources Research and Development Agency (NARA) – Sri Lanka and Sri Lanka Institute of Nanotechnology (SLINTEC).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.foohum.2026.101200.

Data availability

All analyzed data have been included in the manuscript. Additional information available from corresponding author at a reasonable request.

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