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Occurrence and health risk assessment of antimony, arsenic, barium, cadmium, chromium, nickel, and lead in fresh fruits consumed in South Korea

Jian Lee[1,2*], Insun Hwang[1], Ye-Seul Park[1] and Do Yup Lee[2]

Abstract

Although various fruits are consumed as fresh produce in South Korea, information on the concentrations of heavy metals in such fruits remains lacking despite the known toxic effects of the metals. Moreover, the health risks posed by seven potentially toxic metals (As, Ba, Cd, Cr, Ni, Pb, and Sb) ingested through fruit consumption have not been assessed using recent dietary data and occurrence data. Inductively coupled plasma-mass spectrometry was used to quantify these metals in 207 samples of fresh fruits mainly consumed in South Korea. The mean concentrations (mg kg^{-1} fresh weight) of the metals in all fruit samples were as follows: As < 0.0021, Ba 0.3675, Cd < 0.0022, Cr 0.0307, Ni 0.0815, Pb 0.0236, and Sb < 0.0021. Only Ba showed a significant negative correlation with Pb ($\rho = -0.5385$) in the studied fruits at the 95% confidence level. The non-carcinogenic risk of the seven metals in terms of hazard quotients was Pb (0.0149) > As (0.0086) > Ni (0.0081) > Sb (0.0080) > Ba (0.0031) > Cd (0.0027) > Cr (0.0001), and the hazard index, which is the sum of the hazard quotients, was 0.0275 (less than 1). The carcinogenic risks of As and Pb were 4.62E − 07 and 5.05E − 07, respectively (below 1E − 04). The hazard index of seven metals and carcinogenic risks of As and Pb indicated that no health risks were associated with fruit consumption in the Korean population. However, the hazard quotient and carcinogenic risk of Pb in apples were the highest for children aged 1–2 years, indicating that continuous targeted risk monitoring in this age group is required.

Keywords Fruits, Heavy metals, ICP-MS, Estimated daily intake, Hazard quotient, Cancer risk

Introduction

Fresh fruits are significant in the human diet because they contain essential nutrients such as vitamins, minerals, and dietary fiber, along with beneficial non-nutrient substances, such as flavonoids, plant sterols, and other antioxidants [1–3]. Consuming various kinds of fruits ensures adequate intake of these components, which is beneficial in maintaining better health and preventing and managing a variety of communicable and non-communicable diseases [2–4]. For this reason, the World Health Organization recommends consuming more than 400 g of fruits and vegetables daily to enhance general health and lower the risk for specific non-communicable diseases [3–5].

*Correspondence:
Jian Lee
jeana@korea.kr
[1] Food Contaminants Division, Department of Food Safety Evaluation, Ministry of Food and Drug Safety, National Institute of Food and Drug Safety Evaluation, 187, Osongsaengmyeong 2-Ro, Osong-Eup, Heungdeok-Gu, Cheongju-Si, Chungcheongbuk-Do 28159, Republic of Korea
[2] Department of Agricultural Biotechnology, Center for Food and Bioconvergence, Research Institute for Agricultural and Life Sciences, Interdisciplinary Program in Agricultural Genomics, Seoul National University, Seoul 08826, Republic of Korea

However, fruits can also contain various concentrations of essential minerals and potentially toxic metals [6–8], which will be ingested by the human body. Environmental pollution caused by industrial emissions, climate, the nature of the soil in which the plant is grown, overuse of fertilizers and metal-based pesticides, and the degree of maturity of the plant at the time of harvesting, storage, and transportation are all factors that influence heavy metal concentrations in fruits [9, 10]. Heavy metal toxicity can impair the functions of the blood, brain, kidneys, liver, lungs, and other organs, depending on contaminated food consumption and exposure duration (i.e., acute or chronic). Moreover, long-term exposure to heavy metals from food sources may result in cancer [9, 10]. In this regard, metals, such as As, Cd, Hg, and Pb, have gained much attention owing to their toxicity. Therefore, to protect consumer health, the levels of these metals in food should be monitored regularly. Several studies on the occurrence of heavy metals in fruits have been published in response to the growing concern about heavy metals in food [5, 11–14]. Many studies involving human health risk assessments based on fruit consumption data have also been reported [1, 6, 8, 15–18].

Over the past decades, studies involving the occurrence and health risk assessment of potentially toxic metals in food have primarily focused on As, Cd, Hg, and Pb in South Korea and worldwide. Although different fruits are grown in South Korea throughout the year, information on the occurrence of heavy metals in fruits that are widely consumed in South Korea remains limited. Moreover, no health risk assessment results have been reported based on fruit consumption data that reflect recent changes in Korean dietary habits tracked by the National Health and Nutrition Survey from 2016 to 2020 [19]. The target heavy metals investigated in this study were selected from heavy metals included in the Substance Priority List of the Agency for Toxic Substances and Disease Registry [20] and the Priority Pollutant List of the United States Environmental Protection Agency [21], considering their occurrence, toxicity, and potential for human exposure in South Korea. Hg was not investigated because its presence in fruits is typically low or undetectable; it is more commonly found in aquatic food chains, leading to higher exposure through the consumption of fish and seafood [22].

This study aimed to (1) determine the concentrations of seven potentially toxic metals (As, Ba, Cd, Cr, Ni, Pb, and Sb) in the fresh fruits commonly consumed in South Korea via inductively coupled plasma mass spectrometry (ICP-MS), (2) evaluate the correlation between the metals in the fruits categorized by metal or fruit species using principal component analysis (PCA) and hierarchical cluster analysis (HCA), and (3) perform a non-carcinogenic and carcinogenic health risk assessment of the metals ingested through fruit consumption categorized by fruit species as well as by age and sex in the Korean population. This is the first health risk assessment of dietary exposure to these seven metals through the consumption of fresh fruit in South Korea.

Materials and methods

Chemicals and materials

All solutions were prepared using ultrapure water with a specific resistivity of 18 MΩ obtained from a Milli-Q purification system (MilliporeSigma, Bedford, MA, USA). Additionally, 70% (w/w) HNO_3 (Duksan, Ansan, Korea) and 30% (w/w) H_2O_2 (Wako, Osaka, Japan) of electronic grade were used as digestion solvents. Instrument Calibration Standard 2 (100 μg mL^{-1}, Perkin Elmer, Shelton, CT, USA) was used as the multi-element standard for ICP-MS analysis of As, Ba, Cd, Cr, Ni, Pb, and Sb. Using this standard, standard calibration solutions with concentrations 0.1–25 μg L^{-1} were prepared. The solutions were prepared by diluting the stock solutions with 1.5% (w/w) HNO_3 to enhance their stability. The certified reference material (CRM) used was tomato leaves supplied by the National Institute of Standard Technology (NIST) 1573a (Gaithersburg, MD, USA).

Sample collection

In total, 207 fresh fruit samples were collected randomly from local supermarkets in six different regions (Seoul, Gyeonggi-do, Chungcheong-do, Jeolla-do, Kyungsang-do, and Gangwon-do) of South Korea based on the population census model (2018) in order to ensure the coverage of both urban and rural populations, from April to October 2019. Additional file 1: Table S1 provides detailed information on the scientific name and country of origin of each species of the fruit samples collected. These fruits were selected because they are the most consumed fruits in Korea, according to the Korean National Health and Nutrition Examination Survey (KNHANES) of the Korea Disease Control and Prevention Agency [19]. Each sample was kept in its original packaging and transported to the laboratory in an icebox. Fresh fruit samples were prepared for chemical analysis as if they were ready for consumption (washing, peeling, and removing inedible parts). In addition, the edible portions from each fruit sample were homogenized in a food processor (Faciclic 7000, Tefal, Veenendaal, Netherlands) and stored in a freezer (−20 °C) until further analysis.

Microwave-assisted digestion and ICP-MS analysis

The samples were digested using a single-reaction chamber microwave (UltraWAVE™, Milestone, Sorisole, Italy) equipped with 15 polytetrafluoroethylene vessels. Sample digestion was performed using the digestion method established in our previous research [23, 24]. Approximately 0.25 g of each homogenized fruit sample (edible part) was weighed directly into a polytetrafluoroethylene vessel (internal volume, 15 mL) mixed with 0.5 mL 70% HNO_3, 2.5 mL 30% H_2O_2 as the oxidizing agent, and 5 mL H_2O [23]. The heating program was performed in three successive steps: (1) ramp the temperature up to 210 ℃ for 10 min, (2) warm-up at 250 ℃ for 20 min, and (3) maintain at 250 ℃ for 20 min. The oven was operated at 1500 W (full power) [23, 24]. After cooling the chamber to 60 ℃, the final digests were transferred to a 10 mL polypropylene volumetric flask with a cap (Eppendorf, Hamburg, Germany) and diluted with deionized water to 10 mL [23, 24]. Each experiment was performed in triplicate. The analytical blanks were also used in triplicate, and all the sample preparation steps mentioned above were performed without samples.

Determination of potentially toxic metals was carried out using an ICP-MS system (JP-7900 model, Agilent Technologies, Santa Clara, CA, USA) under the following operating parameters [25]: radiofrequency power 1550 W, sampling depth 8.0 mm, carrier gas flow rate 0.82 L min^{-1}, make-up gas flow rate 0.33 L min^{-1}, nebulizer gas flow rate 0.85 L min^{-1}, and He gas flow rate 5.0 L min^{-1} in kinetic energy discrimination mode. The following peaks (*m/z*) were monitored: ^{75}As, ^{137}Ba, ^{111}Cd, ^{52}Cr, ^{60}Ni, ^{208}Pb, and ^{121}Sb. Ar (99.999%, Samsung Oxygen, Cheongju, Korea) was used as the plasma, auxiliary, and nebulizer gas, and He (99.9999%, Danil Gaschem, Eumseong-gun, Korea) was used during the kinetic energy discrimination mode. Polyatomic interferences were verified by monitoring various elemental isotopes and measuring isotopic ratios in the final digests. Furthermore, deionized water blanks were routinely analyzed alongside the digested samples to identify analyte loss or cross-contamination. Calibration standards were also regularly examined as samples to detect drift during ICP-MS.

Analytical quality control

Method validation was performed by following the Association of Official Agricultural Chemists guidelines [26]. Linearity was obtained from the external calibration curves of the elements using least-squares linear regression analysis. Tomato leaves (NIST 1573a) were used as the CRM when evaluating the accuracy and precision of the proposed method. The accuracy (recovery, %) was calculated by comparing the determined values with the certified values of CRM three times a day, and the precision (repeatability) was obtained from the relative standard deviation (%) of triplicate CRM experiments per day. The accuracy and precision of the two metals (Ba and Pb) not reported in the CRM analysis were also examined in the recovery and relative standard deviation of triplicates using grape samples spiked with standards. These tests were carried out in each grape sample at three concentrations (0.4, 2.0, and 4.0 mg kg^{-1}). The limits of detection (LOD) and quantification (LOQ) were determined to be three and ten times the standard deviation of the method blank analytical signal, respectively (n = 7).

Health risk assessment

To assess the non-carcinogenic health risks, the EDI (mg kg^{-1} body weight [b.w.] day^{-1}) and hazard quotient (HQ) of the metals were calculated from Eq. (1) and (2), respectively [10, 27, 28].

$$EDI = \frac{C \times IR \times ED \times EF}{AT \times 10^3} \tag{1}$$

$$HQ = \frac{EDI}{RfD} \tag{2}$$

In these equations, C is the concentration of each metal in fresh fruit samples (mg kg^{-1} fresh weight [f.w.]); IR is the ingestion rate measured as the daily fruit consumption per b.w. (g kg^{-1} b.w. day^{-1}); ED is the duration of exposure (12 years for children aged less than 19 years; 70 years for adults); EF is the exposure frequency (365 days $year^{-1}$); AT is the average duration of exposure (365 days $year^{-1} \times ED$); and RfD is the oral reference dose of each metal (mg kg^{-1} b.w. day^{-1}).

With regard to IR, fresh fruit consumption data were collected from the 24-h food intake records of approximately 32407 KNHANES participants from the South Korean population from 2016 to 2020 [19]. The mean fruit consumption (g f.w. day^{-1}) by fruit species, age, and sex (males and females) in the Korean population, as well as the mean b.w. (kg b.w.) by age and sex, are presented in Figs. 1b and 2a and Additional file 1: Table S2.

RfD values for inorganic As, Ba, Cd, Cr (III), Ni, and Sb are 0.0003, 0.2, 0.001, 1.5, 0.02, and 0.0004 mg kg^{-1} b.w. day^{-1}, respectively [29, 35]. The United States Environmental Protection Agency does not provide the *RfD* of Pb. In a recent study, however, the *RfD* was considered 0.004 mg kg^{-1} b.w. day^{-1} [36, 37], and risk assessment was carried out using this *RfD* of Pb. In the present study, to determine the appropriate *RfD* for HQ calculation, it was assumed that all chromium ions in the fruits were trivalent (non-carcinogenic) and all As was inorganic.

In addition, to estimate the total risk of non-carcinogenic effects from more than one heavy metal, the hazard index (HI) was calculated from the sum of the HQs using Eq. (3) [10, 27, 28]:

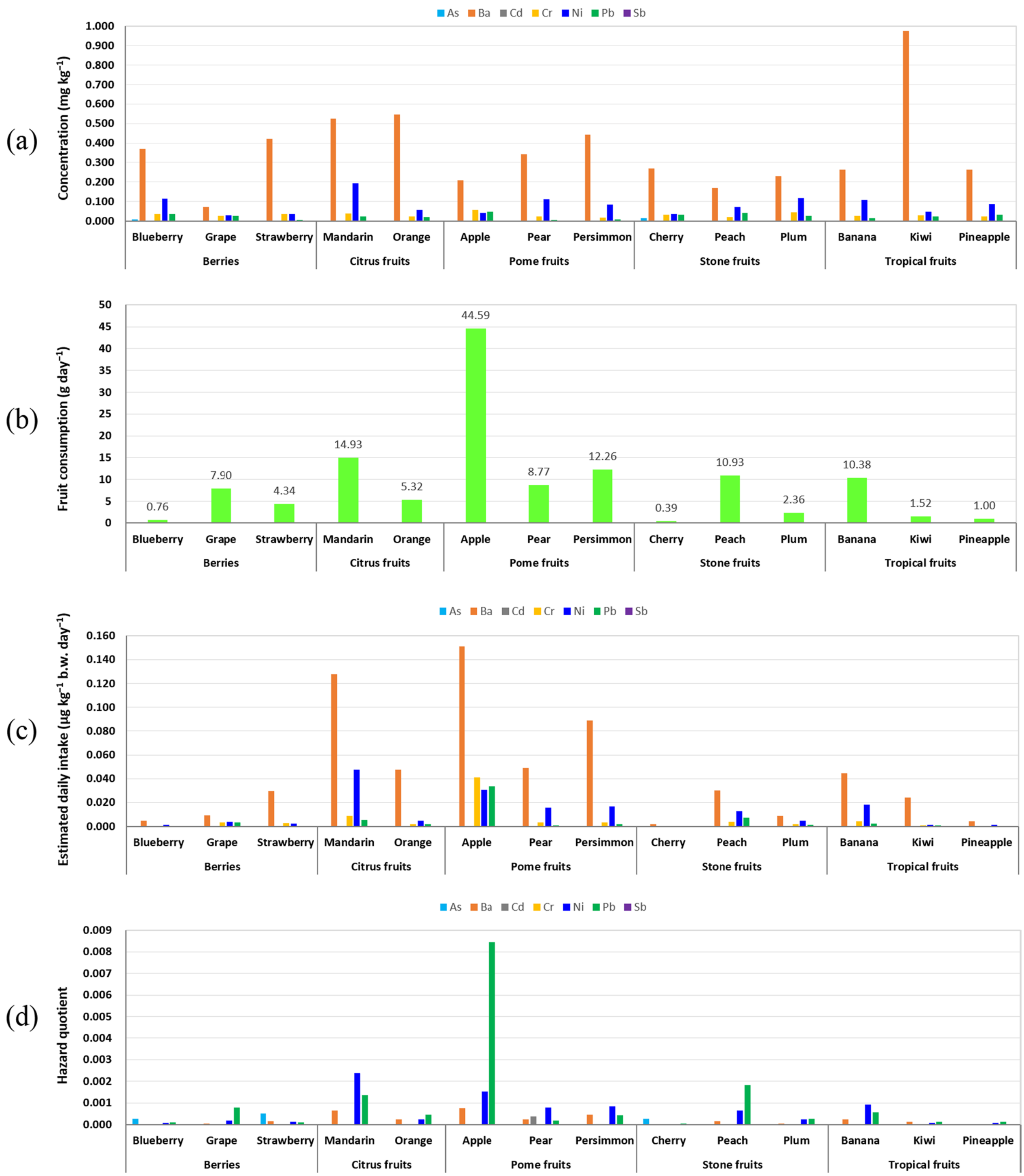


Fig. 1 Occurrence of seven metals by fruit species (**a**), fruit consumption (**b**), estimated daily intakes (**c**), and hazard quotients (**d**) of the metals by fruit species in the Korean population of all ages

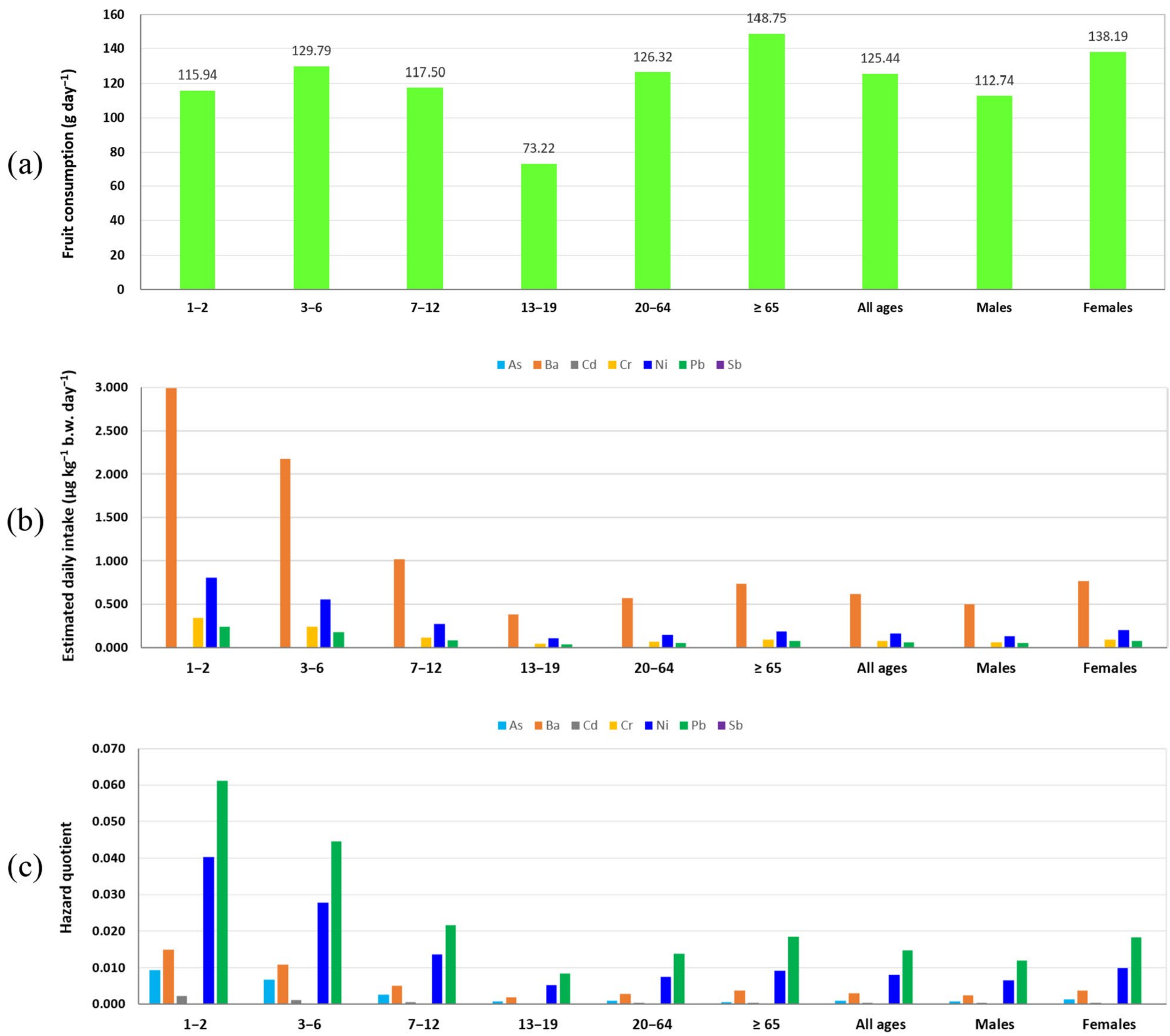


Fig. 2 Fruit consumption (**a**), estimated daily intakes (**b**), and hazard quotients (**c**) by age and sex in the Korean population

$$HI = HQ_{As} + HQ_{Ba} + HQ_{Cd} + HQ_{Cr} + HQ_{Ni} + HQ_{Pb} + HQ_{Sb} \quad (3)$$

To assess the carcinogenic health risk, cancer risk (CR) was calculated using Eq. (4) [10, 27, 28]:

$$CR = EDI \times CSF \quad (4)$$

where CSF is the carcinogenic slope factor for a particular heavy metal. The CSF for inorganic As is 1.5 (mg kg^{-1} b.w. day^{-1})$^{-1}$ [29], but the CSF for Pb is not available, according to the United States Environmental Protection Agency. Recently, the CSF of Pb was determined to be 0.0085 (mg kg^{-1} b.w. day^{-1})$^{-1}$ [28, 36].

Statistical analysis

Descriptive statistics and risk assessments were performed using Microsoft Office Excel 2022 (Microsoft, Redmond, WA, USA); GraphPad Prism version 9.4.1, 2022 (GraphPad Software, San Diego, CA, USA); and SPSS version 28, 2022 (IBM, Armonk, NY, USA). Student's t-test ($p < 0.05$) was used to determine whether there were significant differences between the certified and determined values of a CRM. The quantification data of each fruit group were not normalized, nor was homoscedasticity detected when performing the Shapiro–Wilk and Levene's tests. Therefore, the occurrence data of heavy metals in each fruit group were tested using a non-parametric analysis of variance (i.e., Kruskal–Wallis test) and post-hoc Dunn's test. The Spearman correlation test was performed to examine the correlation between the metals in fruit species. PCA and HCA were employed to classify the fruit species according to their metal concentrations. Eigenvalue criteria > 1.00 were used to determine the number of factors. HCA was applied using Ward's method, and Euclidean distance was used as a distance measure. The fruit consumption and b.w. data of the KNHANES [19] were obtained using Rstudio (version 4.2.2). The R packages "survey," "reshape2," and "dplyr" were used to calculate mean fruit consumption and mean b.w. in the Korean population.

Results and discussion

Analytical performance

The analytical parameters are listed in Table 1. Regarding linearity, the coefficients of determination (R^2) for all the external calibration curves were greater than 0.9996 (Ba). The accuracy of the analytical method was confirmed using tomato leaves (NIST 1573a) as the CRM. The results obtained for As, Cd, Cr, and Ni demonstrated no significant differences between the certified and determined values (t-test; 95% confidence level). Spiking experiments were further evaluated at concentrations 0.4, 2.0, and 4.0 mg kg^{-1} for Ba and 0.1, 0.3, and 0.6 mg kg^{-1} for Pb. The recoveries ranged from 99.8% (Pb) to 114.1% (Ba), with a precision of < 3.2% (Ba). These results are valid according to the criteria proposed by the Association of Official Agricultural Chemists [26]. The LOD and LOQ of the metals ranged from 2.098 (Sb) to 3.009 μg kg^{-1} (Ba) and 6.923 (Sb) to 9.930 μg kg^{-1} (Ba), respectively.

Fruit consumption

According to the KNHANES (2016 – 2020), 14 fruit species account for approximately 95% of the daily fruit consumption by Koreans. Population data were categorized into six age groups (1 – 2, 3 – 6, 7 – 12, 13 – 19, 20 – 64, and ≥ 65 years old) based on the human developmental life cycle [7]. The mean daily consumption of the 14 fruit species by Koreans of all ages was 125.44 g f.w. day^{-1}. Among the fruit species, the pome fruit apples (44.59 g f.w. day^{-1}) were the most consumed fruit, and the stone fruit cherries (0.39 g f.w. day^{-1}) were the least (Fig. 1b). Among age groups, seniors above 65 years of age had the highest mean daily consumption (148.75 g f.w. day^{-1}), whereas children aged 13–19 years had the lowest (73.22 g f.w. day^{-1}). Between sex groups, females (138.19 g f.w. day^{-1}) exhibited greater mean daily consumption than males (112.74 g f.w. day^{-1}) (Fig. 2a).

Occurrence of potentially toxic metals in fruits

The mean concentrations and ranges of the seven metals in the 207 fruit samples examined are presented in Table 2. A comparison of mean metal concentrations

Table 1 Analytical performance obtained from the method in this study

Metal	R^2	LOD (μg kg^{-1})[a]	LOQ (μg kg^{-1})[a]	CRM NIST 1573a Tomato leaves		
				Certified values (mg kg^{-1})	Determined values (mg kg^{-1})[b]	RSD (%)
As	0.9997	2.141	7.065	0.1126 ± 0.0024	0.1085 ± 0.0048	4.4
Cd	0.9999	2.239	7.389	1.517 ± 0.027	1.232 ± 0.004	0.3
Cr	1.0000	2.348	7.748	1.988 ± 0.034	1.765 ± 0.021	1.2
Ni	0.9999	2.679	8.841	1.582 ± 0.041	1.428 ± 0.021	1.4
Sb	1.0000	2.098	6.923	0.0619 ± 0.0032	0.0591 ± 0.0005	0.8
Metal	**R^2**	**LOD (μg kg^{-1})[a]**	**LOQ (μg kg^{-1})[a]**	**Recovery sets at three concentration levels**		
				Fortification levels (mg kg^{-1})[c]	**Rec (%)**	**RSD (%)**
Ba	0.9996	3.009	9.930	0.4	112.1	3.2
				2.0	103.6	0.7
				4.0	114.1	1.2
Pb	0.9999	2.732	9.016	0.1	105.6	1.1
				0.3	104.1	2.2
				0.6	99.8	1.9

R^2 coefficient of determination, *LOD* limit of detection, *LOQ* limit of quantification, *Rec* Recovery, *RSD* relative standard deviation

[a] In the calculation of LOD and LOQ, the sample mass (0.25 g) and the final volume (10 mL) were taken into account. Thus, LOD (μg kg^{-1}) = 40 × LOD (μg L^{-1})

[b] Mean ± standard deviation (n = 3)

[c] Noncertified value

Table 2 Concentrations and ranges of seven metals analyzed in 207 fresh fruit samples

Fruit group	Species	N	Mean ± SD (min – max) (mg kg^{-1} f.w.)						
			As	Ba	Cd	Cr	Ni	Pb	Sb
Total		**207**	**< 0.0021 (< 0.0021 – 0.0400)**	**0.3675 ± 0.3173 (0.0209 – 1.8392)**	**< 0.0022 (< 0.0022 – 0.0098)**	**0.0307 ± 0.0199 (0.0094 – 0.1262)**	**0.0815 ± 0.0769 (0.0062 – 0.4779)**	**0.0236 ± 0.0225 (< 0.0027 – 0.0968)**	**< 0.0021 (< 0.0021 – 0.0089)**
Berries		**48**	**0.0030 ± 0.0047^{ab} (< 0.0021 – 0.0271)**	**0.2945 ± 0.2222^{a} (0.0309 – 0.7485)**	**< 0.0022 (< 0.0022 – 0.0098)**	**0.0326 ± 0.0187^{ab} (0.0100 – 0.0869)**	**0.0590 ± 0.0665^{a} (0.0063 – 0.3298)**	**0.0212 ± 0.0109 (< 0.0027 – 0.0834)**	**< 0.0021 (< 0.0021 – 0.0089)**
	Blueberry	16	0.0062 ± 0.0066 (< 0.0021 – 0.0271)	0.3704 ± 0.1503 (0.1368 – 0.6157)	< 0.0022 (< 0.0022 – 0.0047)	0.0357 ± 0.0187 (0.0137 – 0.0727)	0.1133 ± 0.0893 (0.0267 – 0.3298)	0.0343 ± 0.0208 (0.0091 – 0.0834)	< 0.0021 (< 0.0021 – 0.0089)
	Grape	15	< 0.0021 (< 0.0021 – 0.0041)	0.0710 ± 0.0355 (0.0309 – 0.1805)	< 0.0022 (< 0.0022 – 0.0028)	0.0253 ± 0.0131 (0.0100 – 0.0634)	0.0287 ± 0.0211 (0.0063 – 0.0844)	0.0246 ± 0.0154 (0.0051 – 0.0589)	< 0.0021 (< 0.0021 – 0.0070)
	Strawberry	17	0.0021 (< 0.0021 – 0.0088)	0.4204 ± 0.2275 (0.1086 – 0.7485)	< 0.0022 (< 0.0022 – 0.0098)	0.0360 ± 0.0216 (0.0112 – 0.0869)	0.0347 ± 0.0200 (0.0116 – 0.0791)	0.0060 ± 0.0028 (< 0.0027 – 0.0139)	< 0.0021 (< 0.0021 – 0.0050)
Citrus fruits		**30**	**< 0.0021**	**0.5357 ± 0.2835^{b} (0.1136 – 1.1670)**	**< 0.0022**	**0.0300 ± 0.0133^{ab} (0.0110 – 0.0662)**	**0.1255 ± 0.1141^{b} (0.0174 – 0.4779)**	**0.0215 ± 0.0157 (< 0.0027 – 0.0956)**	**< 0.0021 (< 0.0021 – 0.0040)**
	Mandarin	15	< 0.0021	0.5239 ± 0.2782 (0.1136 – 0.9161)	< 0.0022	0.0369 ± 0.0126 (0.0155 – 0.0662)	0.1947 ± 0.1256 (0.0595 – 0.4779)	0.0221 ± 0.0040 (0.0096 – 0.0298)	< 0.0021 (< 0.0021 – 0.0027)
	Orange	15	< 0.0021	0.5475 ± 0.2882 (0.1142 – 1.1670)	< 0.0022	0.0231 ± 0.0101 (0.0110 – 0.0466)	0.0563 ± 0.0264 (0.0174 – 0.1250)	0.0209 ± 0.0218 (< 0.0027 – 0.0956)	< 0.0021 (< 0.0021 – 0.0040)
Pome fruits		**45**	**< 0.0021^{ab}**	**0.3315 ± 0.2018^{a} (0.0209 – 0.9659)**	**< 0.0022 (< 0.0021 – 0.0093)**	**0.0319 ± 0.0284^{a} (0.0094 – 0.1262)**	**0.0785 ± 0.0518^{ab} (0.0062 – 0.2115)**	**0.0200 ± 0.0231 (< 0.0027 – 0.0831)**	**< 0.0021 (< 0.0021 – 0.0051)**
	Apple	15	< 0.0021	0.2076 ± 0.1296 (0.0209 – 0.5034)	< 0.0022	0.0567 ± 0.0364 (0.0167 – 0.1262)	0.0422 ± 0.0373 (0.0062 – 0.1542)	0.0464 ± 0.0217 (0.0110 – 0.0833)	< 0.0021 (< 0.0021 – 0.0051)
	Pear	15	< 0.0021	0.3424 ± 0.2174 (0.1040 – 0.9659)	0.0026 ± 0.0026 (< 0.0022 – 0.0093)	0.0236 ± 0.0110 (0.0147 – 0.0543)	0.1103 ± 0.0542 (0.0412 – 0.2115)	0.0052 ± 0.0047 (< 0.0027 – 0.0198)	< 0.0021 (< 0.0021 – 0.0028)
	Persimmon	15	< 0.0021	0.4444 ± 0.1732 (0.2319 – 0.8422)	< 0.0022	0.0155 ± 0.0053 (0.0094 – 0.0318)	0.0831 ± 0.0372 (0.0226 – 0.1344)	0.0084 ± 0.0073 (< 0.0027 – 0.0257)	< 0.0021
Stone fruits		**39**	**0.0029 ± 0.0084^{a} (< 0.0021 – 0.040)**	**0.2161 ± 0.1524^{a} (0.0413 – 0.7538)**	**< 0.0022 (< 0.0022 – 0.0051)**	**0.0330 ± 0.0180^{b} (0.0101 – 0.0941)**	**0.0805 ± 0.0577^{ab} (0.0074 – 0.2910)**	**0.0337 ± 0.0268 (< 0.0027 – 0.0931)**	**< 0.0021 (< 0.0021 – 0.0044)**
	Cherry	9	0.0126 ± 0.0136 (< 0.0021 – 0.0400)	0.2690 ± 0.1210 (0.0763 – 0.5053)	< 0.0022	0.0330 ± 0.0127 (0.0101 – 0.0535)	0.0338 ± 0.0227 (0.0074 – 0.0825)	0.0320 ± 0.0190 (0.0058 – 0.0735)	< 0.0021 (< 0.0021 – 0.0022)
	Peach	15	< 0.0021	0.1692 ± 0.1773 (0.0413 – 0.7538)	< 0.0022 (< 0.0022 – 0.0032)	0.0207 ± 0.0062 (0.0133 – 0.0409)	0.0717 ± 0.0351 (0.0173 – 0.1169)	0.0412 ± 0.0330 (0.0035 – 0.0931)	< 0.0021 (< 0.0021 – 0.0044)
	Plum	15	< 0.0021	0.2313 ± 0.1267 (0.0816 – 0.4981)	< 0.0022 (< 0.0022 – 0.0051)	0.0453 ± 0.0202 (0.0202 – 0.0941)	0.1173 ± 0.0662 (0.0191 – 0.2910)	0.0272 ± 0.0215 (0.0032 – 0.0865)	< 0.0021 (< 0.0021 – 0.0041)
Tropical fruits		**45**	**< 0.0021^{b} (< 0.0021 – 0.0035)**	**0.5005 ± 0.4822^{a} (0.0249 – 1.8392)**	**< 0.0022 (< 0.0022 – 0.0030)**	**0.0258 ± 0.0142^{a} (0.0094 – 0.0831)**	**0.0801 ± 0.0800^{b} (0.0113 – 0.4425)**	**0.0223 ± 0.0227 (< 0.0027 – 0.0968)**	**< 0.0021 (< 0.0021 – 0.0057)**
	Banana	15	< 0.0021 (< 0.0021 – 0.0035)	0.2624 ± 0.3048 (0.0249 – 1.0135)	< 0.0022 (< 0.0022 – 0.0030)	0.0247 ± 0.0132 (0.0094 – 0.0555)	0.1072 ± 0.1089 (0.0113 – 0.4425)	0.0135 ± 0.0150 (< 0.0027 – 0.0604)	< 0.0021 (< 0.0021 – 0.0057)
	Kiwi	15	< 0.0021 (< 0.0021 – 0.0028)	0.9745 ± 0.4812 (0.2285 – 1.8392)	< 0.0022	0.0302 ± 0.0176 (0.0123 – 0.0831)	0.0465 ± 0.0264 (0.0161 – 0.1208)	0.0217 ± 0.0176 (0.0046 – 0.0767)	< 0.0021 (< 0.0021 – 0.0034)
	Pineapple	15	< 0.0021 (< 0.0021 – 0.0028)	0.2646 ± 0.1897 (0.0534 – 0.7434)	< 0.0022	0.0224 ± 0.0095 (0.0100 – 0.0427)	0.0866 ± 0.0689 (0.0152 – 0.3002)	0.0316 ± 0.0290 (< 0.0027 – 0.0968)	< 0.0021 (< 0.0021 – 0.0036)

N number of samples, *Mean* arithmetic mean, *SD* standard deviation, *mg kg^{-1} f.w.* concentration calculated on a fresh weight basis, < = less than the LOD

In each column, lowercase superscripts (a, b, c, etc.) indicate statistical variations between different fruit groups. Values with the same superscripts in each column are not significantly different from each other, but those with different superscripts are significantly different ($p < 0.05$)

In each row, bold text indicates statistical results for the total of all fruit species and subtotals of fruit groups by fruit species is shown in Fig. 1a. In descending order, the mean concentrations (mg kg^{-1} f.w.) of the metals in all fruit samples were Ba (0.3675) > Ni (0.0815) > Cr (0.0307) > Pb (0.0236) > As, Cd, and Sb (< LODs). The mean As concentrations (mg kg^{-1} f.w.) among fruit species were < 0.0021, except in blueberries (0.0062), cherries (0.0126), and strawberries (0.0021); the mean Cd concentrations (mg kg^{-1} f.w.) were < 0.0022, except in pears (0.0026); and the mean Sb concentrations were < 0.0021 mg kg^{-1} f.w. in all fruit samples (Table 2). The highest concentrations (mg kg^{-1} f.w.) of As, Ba, Cd, Cr, Ni, Pb, and Sb were found in cherry (0.040), kiwi fruit (1.8392), strawberry (0.0098), apple (0.1262), mandarin (0.4779), pineapple (0.0968), and blueberry (0.0089) samples, respectively.

Currently, the Codex Alimentarius Commission [38] has established a maximum level (ML) of only Pb (0.1 mg kg^{-1} f.w.) for berries and other small fruits, except for cranberries, raisins, and elderberries. The Korean Ministry of Food and Drug Safety [39] established MLs for only Cd (0.05 mg kg^{-1} f.w.) and Pb (0.1 mg kg^{-1} f.w.) in fruits. The European Union has set MLs for Cd and Pb in various fruits under Commission Regulation (EC) No. 1881/2006 [40]. The MLs of Cd are 0.020 mg kg^{-1} f.w. (citrus fruits, pome fruits, stone fruits, table olives, kiwi fruits, bananas, mangoes, papayas, and pineapples), 0.030 mg kg^{-1} f.w. (berries and small fruits, except raspberries), 0.040 mg kg^{-1} f.w. (raspberries), and 0.050 mg kg^{-1} f.w. (fruits, except those listed previously), whereas the MLs of Pb are 0.10 mg kg^{-1} f.w. (fruits, excluding cranberries, currants, elderberries, and strawberry tree fruits) and 0.20 mg kg^{-1} f.w. (cranberries, currants, elderberries, and strawberry tree fruits). Additionally, the Japanese Ministry of Health, Labor, and Welfare has established MLs as follows: 3.5 mg kg^{-1} f.w. (apples and Japanese pears) and 1 mg kg^{-1} f.w. (peaches, strawberries, and grapes) for As (trivalent) and 5 mg kg^{-1} f.w. (apples and Japanese pears) and 1 mg kg^{-1} f.w. (peaches, strawberries, and grapes) for Pb [41]. The concentrations of As, Cd, and Pb in the fruits obtained in this study ranged from 0.0021 to 0.04 mg kg^{-1} f.w., 0.0022 to 0.0098 mg kg^{-1} f.w., and 0.0027 to 0.0968 mg kg^{-1} f.w., respectively, all of which were below the MLs.

Among the five fruit groups, berries (n = 48), citrus fruits (n = 30), pome fruits (n = 45), stone fruits (n = 39), and tropical fruits (n = 45) showed no significant differences in Cd, Pb, and Sb concentrations ($p > 0.05$). However, the fruit groups differed significantly in terms of As, Ba, Cr, and Ni content ($p < 0.05$).

As is one of the most toxic heavy metals in the food chain and occurs in many environments. Inorganic As intake at high concentrations can result in serious health consequences such as bladder, kidney, liver, lung, prostate, and skin cancers [10, 28, 42]. As was found in 36 of the 207 fruit samples, demonstrating a detection rate of 17.4%. Among the fruit groups, stone fruits differed significantly from tropical fruits in terms of mean As concentration ($p = 0.031$). The mean As concentrations (mg kg^{-1} f.w.) among fruit species were < 0.0021, except for blueberries (0.0062), cherries (0.0126), and strawberries (0.0126) (Table 2). Previous studies have reported that the mean As concentration (mg kg^{-1} f.w.) is 0.104 in bananas [16], 0.119 in citrus fruits [43], and < 0.4 in bananas [8]. The mean As concentrations reported in previous studies are higher than those in the present study.

Ba is a nonessential element in organisms and is toxic to animals and plants. Ba exposure can result in various adverse health effects in animals, including metabolic, neurological, and mental disorders, cardiovascular and renal diseases, and even death [44]. In this study, Ba was observed in 207 of the 207 fruit samples, demonstrating a 100.0% detection rate. Among the seven heavy metals, Ba was the most abundant in all fruit species (Fig. 1a and Table 2). The mean Ba concentration (mg kg^{-1} f.w.) in all the fruit samples was 0.3675, with citrus fruits (0.5357) ranking first, followed by tropical fruits (0.5005), pome fruits (0.3315), berries (0.2945), and stone fruits (0.2161). Among the fruit groups, berries, stone fruits, and tropical fruits differed significantly from citrus fruits in terms of mean Ba concentration ($p = 0.002$, 0.007, and 0.007, respectively). Among the fruit species, the mean Ba concentrations (mg kg^{-1} f.w.) were the highest in kiwi fruits (0.9745) and lowest in grapes (0.0710). However, previous studies have not quantified Ba in fruits, making comparisons impossible.

Cd is among the most toxic industrial and environmental heavy metals and has a long half-life [9, 10]. Cd accumulation in the human body may cause multifaceted adverse effects, including carcinogenic, hepatotoxic, nephrotoxic, reproductive, skeletal, and teratogenic effects [10, 28]. In this study, Cd was identified in 23 of the 207 fruit samples, resulting in a detection rate of 11.1%. The mean Cd concentrations in all 14 fruit species were < 0.0022 mg kg^{-1} f.w. Previous studies reported that the mean Cd concentrations (mg kg^{-1} f.w.) were 0.057 and < 0.06 in bananas [8, 16], 0.009 in citrus fruits [43], 0.00062 and 0.300 in mandarins [13, 45], 0.00049 and 0.300 in oranges [13, 43], 0.0018 and 0.001 in apples [11, 14], 0.0024 and 0.018 in strawberries [11, 14], 0.001 in grapes [14], and 0.004 in pears [14]. Therefore, the mean Cd concentrations reported in previous studies are comparable to or somewhat higher than those in our study.

Cr is an essential mineral because it is involved in insulin function and the metabolism and storage of carbohydrates, fat, and protein [10, 28]. However, depending on its oxidation state, Cr can either be potentially toxic as Cr(VI) or vital to the human body as Cr(III). The latter form is commonly found in food and is required in humans at trace levels. Most ingested Cr(VI) is thought to be reduced in the stomach to Cr(III), which is weakly bioavailable and has low cell penetrability [10, 46]. In this study, Cr was detected in 207 of the 207 fruit samples demonstrating a 100.0% detection rate. The mean Cr concentration (mg kg^{-1} f.w.) in the fruit samples was 0.0307, with stone fruits (0.0330) ranking first, followed by berries (0.0326), pome fruits (0.0319), citrus fruits (0.0300), and tropical fruits (0.0258). Among the fruit groups, pome and tropical fruits differed significantly from stone fruits in terms of the mean Cr concentration ($p=0.001$ and 0.040, respectively). The mean Cr concentrations (mg kg^{-1} f.w.) were the highest in apples (0.0567) and lowest in persimmons (0.0155). Prior research reported mean Cr concentrations (mg kg^{-1} f.w.) as 0.010 in grapes, 0.016 in peaches [6], and 0.317 in bananas [8]. The mean Cr concentrations reported in previous studies are comparable to those in our study.

Ni, an essential component of hematopoietic processes, plays an important role in the physiological processes and development of the human immune system [1]. However, high concentrations of Ni can cause allergies, cardiovascular diseases, kidney diseases, lung inflammation, lung fibrosis, and lung and nasal cancers [1, 10]. In this study, Ni was identified in 207 of 207 fruit samples, representing a 100.0% detection rate. Ni was the most common metal in all fruit species after Ba (Fig. 1a). The mean Ni concentration (mg kg^{-1} f.w.) in the fruit samples was 0.0815, with citrus fruits (0.1255) having the highest concentration, followed by stone fruits (0.0805), tropical fruits (0.0801), pome fruits (0.0785), and berries (0.0590). Among the fruit groups, berries differed significantly from citrus and tropical fruits in terms of the mean Ni concentration ($p=0.001$ and 0.013, respectively). Moreover, the mean Ni concentrations (mg kg^{-1} f.w.) were highest in mandarins (0.1947) and lowest in grapes (0.0287). Previous studies have reported that the mean Ni concentrations (mg kg^{-1} f.w.) were 0.018 in grapes, 0.036 in peaches [6], and 0.037 in bananas [8]. The Ni values in the aforementioned studies are lower than those in the present study.

Pb is the most prevalent industrial metal in the atmosphere, water, soil, and food. Cumulative Pb intake can result in irreversible neurological damage, renal dysfunction, hypertension, cognitive decline, and behavioral disturbances [37]. This study detected Pb in 193 of the 207 fruit samples, demonstrating a 93.2% detection rate. The mean Pb concentration (mg kg^{-1} f.w.) in all fruit samples was 0.0236 and was significantly higher in apples (0.0464) and lower in strawberries (0.0060) (Fig. 1a and Table 1). Previous studies reported mean Pb concentrations (mg kg^{-1} f.w.) as follows: 0.010 in Jamaican bananas [16], 0.152 in citrus fruits [43], 0.0202 in mandarins, 0.0102 in oranges [13], 0.0233 in apples, 0.0272 in strawberries [11], 0.009 in apples, 0.005 in grapes, 0.008 in pears, 0.009 in strawberries [14], and 0.003 in bananas [8]. The mean Pb concentrations in all the fruit varieties in the present study are similar to or lower than those previously reported.

Sb, a nonessential toxic trace metal, is hazardous to human health. Although Sb is widely distributed on the Earth, its contamination is primarily caused by anthropogenic activity. Exposure to Sb can lead to blood, gastrointestinal, and neurological disorders, especially in children [47]. In this study, Sb was detected in 42 of the 207 fruit samples at a detection rate of 20.3%. However, the mean Sb concentration in all fruit varieties was < 0.0021 mg kg^{-1} f.w. No previous studies have been conducted on Sb concentrations in fruits, which makes comparisons of our results with existing literature difficult.

The results of Spearman's non-parametric correlation analysis of the seven hazardous metals in the fruit samples are presented in Additional file 1: Table S3. This analysis was conducted to determine the rank correlations between the metal concentrations. The intermetal correlations in fruit samples provide essential information on the sources of heavy metals. All metal pairs had positive low-to-moderate correlations (except for the As-Ba, As-Ni, As-Pb, Ba-Cd, Ba-Cr, Ba-Pb, Ba-Sb, Cd–Pb, Ni-Pb, and Ni-Sb pairs). At a 95% confidence level, Ba demonstrated significant negative correlations with Pb ($\rho=-0.5385$) but no correlations with other metal pairs. The correlations between Ba and Cr, Cr and Ni, and Ni and Pb were close to zero, indicating no or weak relationships. These findings are consistent with the following PCA and HCA interpretations.

Multivariate analysis of potentially toxic metals in fruits

Principal component analysis

An important step in PCA is determining the number of factors that must be extracted, that is the number of factors that better describe the relationship pattern between the variables that will be observed. Alternatively, the minimum number of factors that maximizes the total variance can be identified. Although there is no consensus on the number of factors to be extracted, we used the eigenvalue rule in conjunction with the Kaiser criterion [48], which only uses principal components (PCs) with eigenvalues greater than 1. Variable and factor-loading correlations with absolute values between 0.75 and 1.0 are considered strong, those between 0.5 and 0.75 are

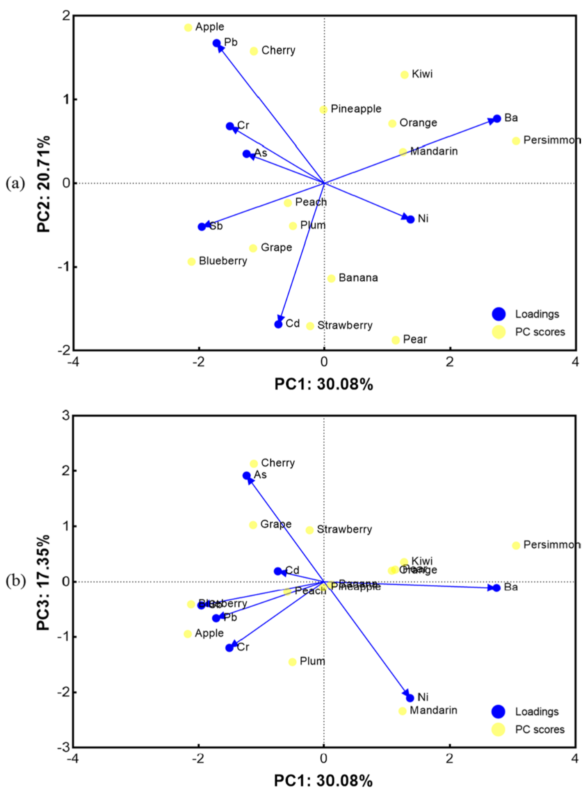


Fig. 3 PCA score and loading plots of the first principal component (PC1) versus the second principal component (PC2) (**a**) and the third principal component (PC3) (**b**) for hazardous metals in fruit species

considered moderate, and those between 0.5 and 0.3 are considered weak [49].

Additional file 1: Table S4 shows the loadings for the original variables in the three PCs and the variances explained by each component. The three PCs had eigenvalues greater than 1, and these three components explained 68.14% of data variability. Figure 3a shows a plot of the first principal component (PC1) versus the second principal component (PC2) for the fruits. For PC1, the dominant variables were Ba, Cr, Pb, and Sb, which represented 30.08% of the total variability and were directly related to the relatively higher contents of Ba in kiwi fruits and oranges, Pb in apples, and Sb in blueberries (Fig. 3a). PC2 accounted for 20.71% of the total variance, with Cd and Pb concentrations being the dominant variables directly related to the higher Pb content of apples and Cd content of strawberries and pears. A plot of PC1 versus the third principal component (PC3) for fruits is shown in Fig. 3b. PC3 explained 17.35% of the data variability, with As and Ni concentrations as the main variables, which were closely related to the relatively higher As and Ni contents of cherries and mandarins, respectively.

Hierarchical cluster analysis

HCA was conducted using the mean values of seven metals in the fruit species. Ward's method, also referred to as the least variance method, was used to group the data to achieve a minor internal error between the vectors that comprised each group and the mean vector of the group. This was equivalent to determining the minimum standard deviation among the data from each group. This technique relies primarily on the concept of analysis of variance linked to the Euclidean distance. Consequently, it is possible to confirm that the similarity of the compared elements increases with decreasing Euclidean distance, which can help identify similarities between samples [48].

Ward's method with Euclidean distances was used to create a dendrogram. The dendrogram at the cleavage of linkage distance 5 in Fig. 4a shows that Ba and Ni formed distinct groups, whereas As, Cd, Cr, Pb, and Sb formed a large group. This indicates that Ba was the most different metal, followed by Ni. As shown in Fig. 4b, when the dendrogram was cut at a linkage distance of eight, the first group comprised bananas, pineapples, plums, cherries, apples, peaches, and grapes. The second group included blueberries, pears, strawberries, persimmons, mandarins, and oranges. The third group comprised kiwi fruits. This demonstrates that kiwi fruits are distinct from the other fruit species. These findings are consistent with those obtained using PCA in Fig. 3.

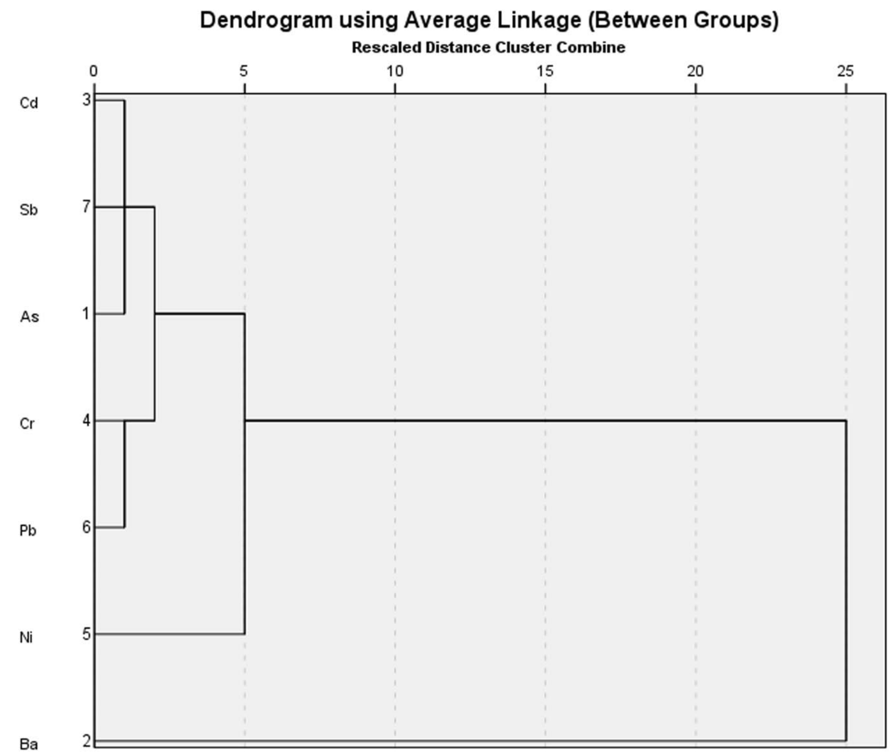
Risk assessment of potentially toxic metals in fruits

Exposure assessment

The EDIs (μg kg^{-1} b.w. day^{-1}) of potentially toxic metals were estimated using the mean concentration (mg kg^{-1} f.w.) of each metal in the 14 fresh fruit species, mean fruit consumption of each fresh fruit (g f.w. day^{-1}), and mean body weight (kg^{-1} b.w.). The EDIs of the seven metals ingested via fruit consumption in the Korean population are presented in Figs. 1c and 2b and Table 3. Additional file 1: Table S5 provides more detailed information on the fruit variety, age, and sex. Left-censored occurrence data (below the LODs) of the metals in 14 fresh fruit species were not available (NA) to calculate the exposure assessment.

The EDI of As through the consumption of the studied fruits by the Korean population of all ages was 0.0003 μg kg^{-1} b.w. day^{-1}, and the EDI of As (0.0002 μg kg^{-1} b.w. day^{-1}) through strawberry consumption was the highest at 49.4% (Fig. 1c and Table 3). Antoine et al. [16] reported that the EDI of As was

(a)



(b)

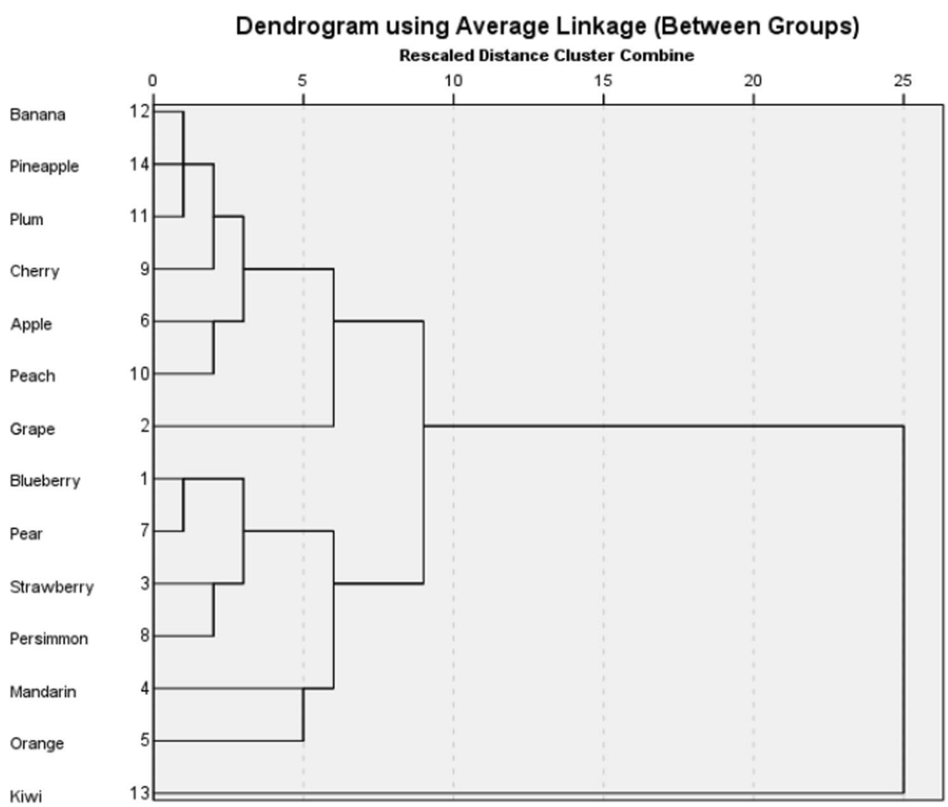


Fig. 4 Hierarchical dendrograms by metal (**a**) and by fruit species (**b**) using Ward's method (the distances reflect the degree of correlation between different metals)

Table 3 Estimated daily intakes (EDIs), hazard quotients (HQs), and hazard index (HI) of seven metals through fruit consumption in the Korean population of all ages

Fruit group	Species	EDI (μg kg^{-1} b.w. day^{-1})							HQ							HI[a]
		As	Ba	Cd	Cr	Ni	Pb	Sb	As	Ba	Cd	Cr	Ni	Pb	Sb	
Total		0.0003	0.6213	0.0004	0.0759	0.1612	0.0594	–[c]	0.0010	0.0031	0.0004	0.0001	0.0081	0.0148	–	0.0275
Berries	Subtotal	0.0002	0.0435	–	0.0063	0.0076	0.0040	–	0.0008	0.0002	–	0.0000[b]	0.0004	0.0010	–	0.0024
	Blueberry	0.0001	0.0046	–	0.0004	0.0014	0.0004	–	0.0003	0.0000	–	0.0000	0.0001	0.0001	–	0.0005
	Grape	–	0.0092	–	0.0033	0.0037	0.0032	–	–	0.0000	–	0.0000	0.0002	0.0008	–	0.0010
	Strawberry	0.0002	0.0298	–	0.0026	0.0025	0.0004	–	0.0005	0.0001	–	0.0000	0.0001	0.0001	–	0.0009
Citrus fruits	Subtotal	–	0.1751	–	0.0110	0.0523	0.0072	–	–	0.0009	–	0.0000	0.0026	0.0018	–	0.0053
	Mandarin	–	0.1276	–	0.0090	0.0474	0.0054	–	–	0.0006	–	0.0000	0.0024	0.0013	–	0.0044
	Orange	–	0.0475	–	0.0020	0.0049	0.0018	–	–	0.0002	–	0.0000	0.0002	0.0005	–	0.0009
Pome fruits	Subtotal	–	0.2889	0.0004	0.0477	0.0631	0.0362	–	–	0.0014	0.0004	0.0000	0.0032	0.0090	–	0.0140
	Apple	–	0.1511	–	0.0412	0.0307	0.0338	–	–	0.0008	–	0.0000	0.0015	0.0084	–	0.0108
	Pear	–	0.0490	0.0004	0.0034	0.0158	0.0007	–	–	0.0002	0.0004	0.0000	0.0008	0.0002	–	0.0016
	Persimmon	–	0.0889	–	0.0031	0.0166	0.0017	–	–	0.0004	–	0.0000	0.0008	0.0004	–	0.0017
Stone fruits	Subtotal	0.0001	0.0408	–	0.0056	0.0175	0.0086	–	0.0003	0.0002	–	0.0000	0.0009	0.0022	–	0.0035
	Cherry	0.0001	0.0017	–	0.0002	0.0002	0.0002	–	0.0003	0.0000	–	0.0000	0.0000	0.0001	–	0.0003
	Peach	–	0.0302	–	0.0037	0.0128	0.0074	–	–	0.0002	–	0.0000	0.0006	0.0018	–	0.0026
	Plum	–	0.0089	–	0.0017	0.0045	0.0010	–	–	0.0000	–	0.0000	0.0002	0.0003	–	0.0005
Tropical fruits	Subtotal	–	0.0730	–	0.0053	0.0207	0.0033	–	–	0.0004	–	0.0000	0.0010	0.0008	–	0.0022
	Banana	–	0.0445	–	0.0042	0.0182	0.0023	–	–	0.0002	–	0.0000	0.0009	0.0006	–	0.0017
	Kiwi	–	0.0242	–	0.0008	0.0012	0.0005	–	–	0.0001	–	0.0000	0.0001	0.0001	–	0.0003
	Pineapple	–	0.0043	–	0.0004	0.0014	0.0005	–	–	0.0000	–	0.0000	0.0001	0.0001	–	0.0002

[a] HI = sum of HQs of seven metals

[b] 0.0000 = value under 0.00005

[c] "-" = not available (NA) because the mean concentration of the corresponding metal in each fruit was below the LOD

0.051 μg kg^{-1} b.w. day^{-1} in bananas grown in Jamaica, which was higher than our result with bananas. Furthermore, the EDIs (μg kg^{-1} b.w. day^{-1}) of As were highest in children aged 1–2 years (0.0028) and lowest in those aged 13–19 years and older than 65 years (0.0002), with females (0.0004) showing a greater EDI of As than males (0.0002) (Additional file 1: Table S5).

The EDI of Ba through the consumption of the studied fruits by the Korean population of all ages was 0.6213 μg kg^{-1} b.w. day^{-1}. The EDIs (μg kg^{-1} b.w. day^{-1}) of Ba through the consumption of apples (0.1511) and mandarins (0.1276) were the first and second highest, with contributions of 24.3% and 20.5%, respectively (Fig. 1c and Table 3). However, the absence of previous research on the EDI of Ba via fruit consumption has made these comparisons difficult. Furthermore, the lowest and highest EDIs (μg kg^{-1} b.w. day^{-1}) of Ba were observed in children aged 13–19 years (0.3827) and children aged 1–2 years (2.9931), respectively, and females (0.7707) exhibited a greater EDI of Ba than males (0.5004) (Fig. 2b and Additional file 1: Table S5).

The EDI of Cd through the consumption of the studied fruits by the Korean population of all ages was 0.0004 μg kg^{-1} b.w. day^{-1}; only pears contributed to this value (Table 3). Antoine et al. [16] reported an EDI of Cd (0.028 μg kg^{-1} b.w. day^{-1}) in Jamaican bananas, which is higher than our result. Furthermore, the EDIs (μg kg^{-1} b.w. day^{-1}) of Cd were highest in children aged 1–2 years (0.0023), lowest in children aged 13–19 years (0.0003), and greater in females (0.0005) than in males (0.0004) (Additional file 1: Table S5).

The EDI of Cr through the consumption of the studied fruits by the Korean population of all ages was 0.0759 μg kg^{-1} b.w. day^{-1}, and the EDI through apple consumption (0.0412 μg kg^{-1} b.w. day^{-1}) contributed the most at 54.3% (Fig. 1c and Table 3). Previous research has indicated that the EDIs (μg kg^{-1} b.w. day^{-1}) of Cr were 0.0018 in strawberries consumed in Algeria [15], 0.00068 in grapes, and 0.00509 in peaches [6], which were all lower than our findings: 0.0026 for strawberries, 0.0033 for grapes, and 0.0037 for peaches. Another study discovered an EDI of Cr (0.2 μg day^{-1} person^{-1}) through banana consumption [8]. Furthermore, the EDIs (μg kg^{-1} b.w. day^{-1}) of Cr were highest in children aged 1–2 years (0.3407), lowest in children aged 13–19 years (0.0436), and higher in females (0.0933) than in males (0.0618) (Fig. 2b and Additional file 1: Table S5).

The EDI of Ni through the consumption of the studied fruits by the Korean population of all ages was 0.1612 μg kg^{-1} b.w. day^{-1}. The EDI of Ni through mandarin consumption (0.0474 μg kg^{-1} b.w. day^{-1}) had the highest contribution at 29.4% (Fig. 1c and Table 3), owing to the highest mean Ni content of mandarins (0.1947 mg kg^{-1}) (Table 2). Esposito et al. [6] found that the EDIs (μg kg^{-1} b.w. day^{-1}) of Ni were 0.0013 with grapes and 0.0113 with peaches, which are slightly lower than our findings of 0.0037 in grapes and 0.0128 in peaches. Shaheen et al. [8] discovered an EDI of Ni (0.028 μg day^{-1} $person^{-1}$) with bananas. Furthermore, the EDIs (μg kg^{-1} b.w. day^{-1}) of Ni were highest in children aged 1–2 years (0.8062), lowest in children aged 13–19 years (0.1062), and higher in females (0.1980) than in males (0.1314) (Fig. 2b and Additional file 1: Table S5).

The EDI of Pb through the consumption of studied fruits by the Korean population of all ages was 0.0594 μg kg^{-1} b.w. day^{-1}, with apple consumption (0.0338 μg kg^{-1} b.w. day^{-1}) contributing the most at 56.9% (Fig. 1c and Table 3) as the mean Pb content of apples (0.0464 mg kg^{-1}) and apple intake were the highest according to our study (Table 2 and Additional file 1: Table S6). Prior investigations have revealed the EDIs (μg kg^{-1} b.w. day^{-1}) of Pb of 0.005 with Jamaican bananas [16] and 0.0018 with strawberries consumed in Algeria [15], which are higher than our values of 0.0023 with bananas and 0.0004 with strawberries. Shaheen et al. [8] reported an EDI of Pb (0.0022 μg day^{-1} $person^{-1}$) through banana consumption. In contrast, the EDIs (μg kg^{-1} b.w. day^{-1}) of Pb were the highest in children aged 1–2 years (0.2444) and lowest in children aged 13–19 years (0.0337), with females (0.0731) exhibiting a greater EDI of Pb than males (0.0482) (Fig. 2b and Additional file 1: Table S5).

The EDIs of Sb through the consumption of the studied fruits by the Korean population stratified by age and sex were NA because all the mean Sb contents of the fruit species were less than the LODs (Fig. 1c and Table 3). However, no previous research on the EDI of Sb through fruit consumption has been conducted, making comparisons difficult.

Overall, the EDIs (μg kg^{-1} b.w. day^{-1}) of the metals obtained through the consumption of the studied fruits by Korean population of all ages were Ba (0.6213) > Ni (0.1612) > Cr (0.0759) > Pb (0.0594) > Cd (0.0004) > As (0.0003) > Sb (NA) (Table 3). Apples (44.59 g day^{-1} f.w.) were the most consumed fruit species by Koreans (Fig. 1b). Consequently, the EDI of Ba from apple consumption (0.1511 μg kg^{-1} b.w. day^{-1}) was the highest among the 14 fruit species studied (Fig. 1c and Table 3). Although seniors over 65 years had the greatest daily fruit consumption (148.75 g f.w. day^{-1}) (Fig. 2a), the EDI of Ba in children aged 1 − 2 years was the highest because they had the lowest body weight (11.94 kg b.w.) among all age groups (Fig. 2b and Additional file 1: Table S5).

Risk characterization

The non-carcinogenic health risks of the seven metals from fruit consumption were evaluated in the Korean population using HQ, which is the ratio of the metal's EDI to the *RfD*. The HQ and HI of the seven metals are presented in Figs. 1, 2, and 3, and Table 3; more details are included in Additional file 1: Table S6. When the HQ > 1, adverse effects are likely to occur; if HQ ≤ 1, adverse effects are not likely to occur [27, 28].

The HQ of As due to fruit consumption in the Korean population of all ages is 0.0010. Strawberries ranked at the top (0.0005), followed by blueberries and cherries (0.0003), while all other fruits were NA (Fig. 1d and Table 3). Furthermore, the HQs of As were highest in children aged 1–2 years (0.0094) and lowest in children aged 13–19 years (0.0008), with females (0.0013) displaying greater HQ than males (0.0008) (Fig. 2c and Additional file 1: Table S6).

The HQ of Ba ingested by consuming the fruits studied in the Korean population of all ages was 0.0031. Apples ranked first (0.0008), followed by mandarins (0.0006) and the other fruits (less than 0.0004) (Fig. 1d and Table 3). Moreover, the HQs of Ba were highest in children aged 1–2 years (0.0150), lowest in children aged 13–19 years (0.0019), and higher in females (0.0039) than in males (0.0025) (Fig. 2c and Additional file 1: Table S6).

The HQ of Cd calculated by evaluating the fruit consumption of the Korean population of all ages was 0.0004; the HQ of Cd ingested through pear consumption (0.0004) was the highest, followed by all the other fruits (NA) (Fig. 1d and Table 3). Additionally, the HQs of Cd were highest in children aged 1–2 years (0.0023) and lowest in children aged 13–19 years (0.0003). Females (0.0004) displayed higher Cd HQs than males (0.0003) (Fig. 2c and Additional file 1: Table S6).

The HQ of Cr calculated by examining the fruit consumption of the Korean population of all ages was 0.0001, and the HQs of Cr for each fruit species were close to 0.0000 (Fig. 1d and Table 3). The HQs of Cr were highest in children aged 1 − 2 years and 3–6 years (0.0002) and lowest in children aged 13 − 19 years and adults aged 20 − 64 years (0.0001). Females (0.0001) displayed greater HQ of Cr than males (0.0000) (Fig. 2c and Additional file 1: Table S6). This was due to the extremely low Cr exposure from fruit consumption compared with the high Cr *RfD*, indicating no risk to human health.

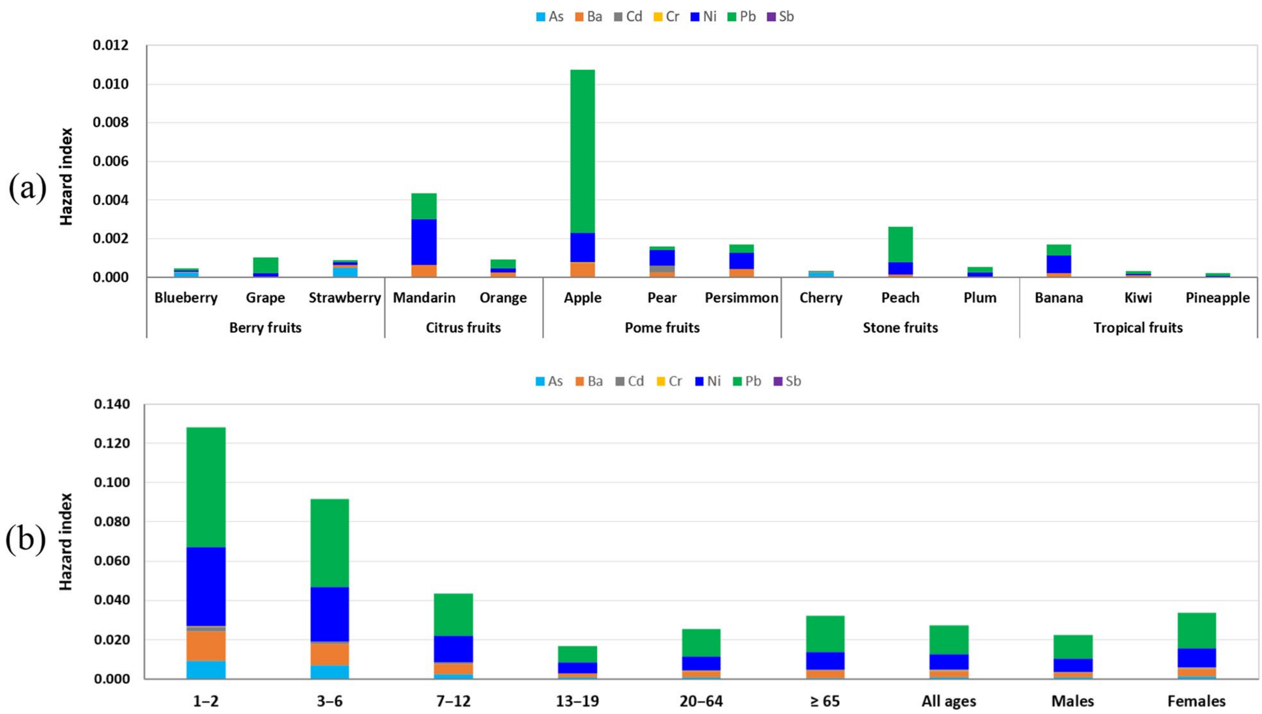


Fig. 5 Hazard indexes by fruit species (**a**) and by age and sex (**b**) in the Korean population

The HQ of Ni obtained by examining the fruit consumption of the Korean population of all ages was 0.0081. Consumption of mandarins resulted in the highest HQ (0.0024), followed by that of apples (0.0015) and the remaining fruits (below 0.001) (Fig. 1d and Table 3). In addition, the HQs of Ni were highest in children aged 1–2 years (0.0403) and lowest in children aged 13–19 years (0.0053), with the HQ in females (0.0099) surpassing that in males (0.0066) (Fig. 2c and Additional file 1: Table S6).

The HQ of Pb ingested through fruit consumption in the Korean population of all ages was 0.0148. Apples rated the highest (0.0084), whereas the rest rated below 0.002 (Fig. 1d and Table 3). Moreover, the HQs of Pb were highest in children aged 1–2 years (0.0611) and lowest in children aged 13–19 years (0.0084), with the HQ in females (0.0183) exceeding that in males (0.0121) (Fig. 2c and Additional file 1: Table S6).

The HQ of Sb calculated from the consumption of the examined fruits in the Korean population of all ages was NA, as was the EDI of Sb, because the mean Sb concentrations in all fruit species were lower than the LOD (Tables 2 and 3).

The HQs of the seven metals ingested via the consumption of the fruits studied in the Korean population of all ages were < 1 for all fruit species. The metals arranged in descending order of HQs were as follows: Pb (0.0148) > Ni (0.0081) > Ba (0.0031) > As (0.0010) > Cd (0.0004) > Cr (0.0001) > Sb (NA) (Table 3). In addition, HI, which is the sum of the HQs, was 0.0275, and the metals arranged in descending order of their percentage contribution to the HI were Pb (54.0) > Ni (29.4) > Ba (11.3) > As (3.7) > Cd (1.4) > Cr (0.2) > Sb (NA) (Table 3). HIs were the highest in apples (0.0108) and lowest in pineapples (0.0002) (Fig. 5a and Table 3). Regarding age and sex, the HIs were highest in children aged 1–2 years (0.1283) and lowest in those aged 13–19 years (0.0167); the HI was higher in females (0.0338) than in males (0.0223) (Fig. 5b and Additional file 1: Table S6).

The carcinogenic health risks of As and Pb ingested via fruit consumption were assessed because these metals have both non-carcinogenic and carcinogenic effects, depending on the exposure dose. Figure 6 shows the CRs of As and Pb according to the fruit species consumed as well as the age and sex groups; more details are given in Additional file 1: Table S7. The calculated CRs are interpreted as follows: a CR of 1E − 06 to 1E − 04 represents no carcinogenic risk for consumers, while CR > 1E − 04 indicates a relatively significant risk [27, 36].

The CR of As ingested via consumption of the studied fruits in the Korean population of all ages was 4.62E − 07; when stratified by fruit species, strawberries exhibited the highest CR (2.28E − 07), followed by cherries and blueberries (1.20E − 07 and 1.14E − 07, respectively). In contrast, all other fruits exhibited negligible CR (Fig. 6a and Additional file 1: Table S7). Furthermore, the CRs of

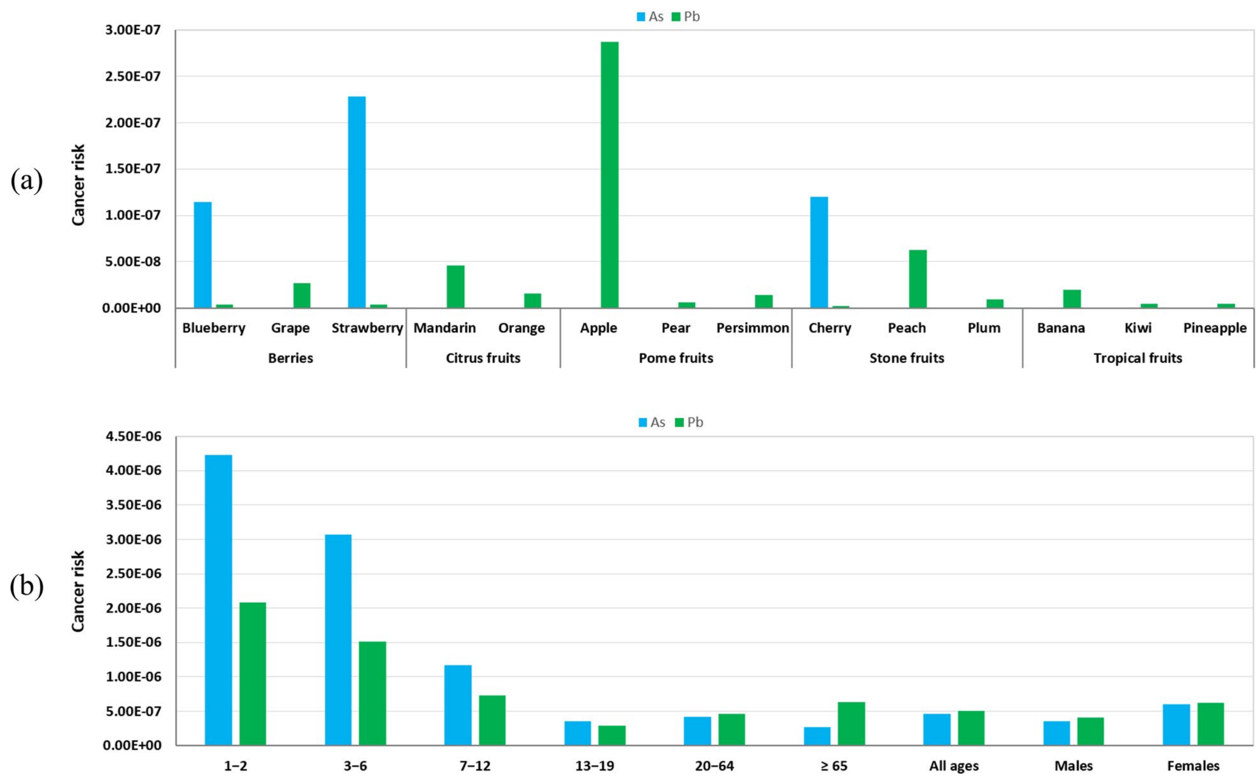


Fig. 6 Cancer risks by fruit species in the Korean population of all ages (**a**) and by age and sex for the studied fruits in the Korean population (**b**)

As were highest in children aged 1 − 2 years (4.23E − 06) and lowest in adults aged above 65 years (2.71E − 07); the CRs of As were higher in females (5.99E − 07) than in males (3.51E − 07) (Fig. 6b and Additional file 1: Table S7).

The CR of Pb ingested via consumption of the studied fruits in the Korean population of all ages was 5.05E − 07, with apples displaying the highest CR of Pb (2.87E − 07) and cherries displaying the lowest CR (1.71E − 09) (Fig. 6a and Additional file 1: Table S7). The CRs of Pb were highest in children aged 1 − 2 years (2.08E − 06) and lowest in children aged 13 − 19 years (2.87E − 07), with females (6.21E − 07) displaying greater CR than males (4.10E − 07) (Fig. 6b and Additional file 1: Table S7).

Taken together, among the HIs of the 14 fruit species, the HI of apples (0.0108) contributed the most, accounting for 39.2% of the overall HI (0.0275) (Fig. 5a and Table 3). In particular, the HQ of Pb (0.0084) from apple intake contributed to 77.8% of the HI in apples, and the CR of Pb (2.87E − 07) due to apple intake contributed to 56.9% of the total CR of Pb (5.05E − 07) (Fig. 6a and Additional file 1: Table S7). Moreover, the HQ of As (0.0005) from strawberry intake contributed 57.2% to the HI in strawberries (0.0009), and the CR of As (2.28E − 07) due to strawberry intake contributed 49.4% to the total CR of As (4.62E − 07) (Fig. 6a and Additional file 1: Table S7). Therefore, the reduction of As and Pb in agricultural environments during the production stage of apple and strawberry orchards is required to minimize the health risks posed by these metals.

Concerning age groups, the HQ of Pb (0.0611) in children aged 1–2 years contributed to 47.6% of the HI in children aged 1–2 years (0.1283) (Fig. 5b and Additional file 1: Table S6). Specifically, the HQ (0.0307) and CR (1.05E − 06) of Pb due to apple intake contributed to 50.3% of the total HQ (0.0611) and total CR (2.08E − 06) of Pb, respectively, in children aged 1–2 years who consumed the 14 fruit species (Fig. 6b and Additional file 1: Table S7). In contrast, the HQ (0.0307) and CR (2.57E − 06) of As ingested via strawberry intake contributed to 60.7% of the total HQ (0.0611) and total CR (4.23E − 06) of As, respectively, in children aged 1 − 2 years who consumed the 14 fruit species (Fig. 6b and Additional file 1: Table S7). Consequently, from a consumer's perspective, proper washing or peeling methods may be advisable before consuming apples and strawberries to reduce the health risks of As and Pb in children aged 1–2 years.

Abbreviations

Ar	Argon
As	Arsenic
Ba	Barium
b.w.	Body weight
Cd	Cadmium
CSF	Carcinogenic slope factor
CR	Cancer risk
Cr	Chromium
CRM	Certified reference material
EDI	Estimated daily intake
f.w.	Fresh weight
HCA	Hierarchical cluster analysis
He	Helium
HI	Hazard index
HQ	Hazard quotient
ICP-MS	Inductively coupled plasma mass spectrometry
KNHANES	Korean National Health and Nutrition Examination Survey
LOD	Limit of detection
LOQ	Limit of quantification
ML	Maximum level
Ni	Nickel
NIST	National Institute of Standard Technology
Pb	Lead
PCA	Principal component analysis
RfD	Oral reference dose
RSD	Relative standard deviation
Sb	Antimony
SD	Standard deviation

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s13765-023-00799-x.

Additional file 1: Table S1. Information of fruit species samples collected in this study. **Table S2.** Fruit consumption and body weights by age and sex in the Korean population. **Table S3.** Spearman correlation coefficients (ρ) between occurrences of each metal. **Table S4.** Loadings of the variables for the three principal components. **Table S5.** Estimated daily intakes (EDIs) of seven metals via fruit consumption by age and sex in the Korean population. **Table S6.** Hazard quotients (HQs) and hazard indexes (HIs) of seven metals via fruit consumption by age and sex in the Korean population. **Table S7.** Cancer risks (CRs) of As and Pb via fruit consumption by age and sex in the Korean population.

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Author contributions
JL: conceptualization, investigation, data curation, methodology, validation, software, visualization, writing—original draft, and writing—review & editing. IH: methodology, investigation, data curation, software, and visualization. YSP: formal analysis, validation, investigation, and data curation. DYL: methodology, writing—review & editing, supervision. All authors read and approved the final manuscript.

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Availability of data and materials
The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing interests
The authors declare that they have no competing interests.

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