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ORIGINAL ARTICLE

Quantification of heavy metals and health risk assessment in processed fruits' products

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Abstract Present study was intendant to assess heavy metals (HMs) concentration and associated health risk in processed fruits' products sold in the local markets of North Pakistan. In total seven metals viz. cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb) and zinc (Zn) were quantified in 345 samples of different brands categorized into eight groups (Sauces, Ketchup, Juices, Jams, canned fruits, tomato paste, marmalades and pickles). On the comparative basis, Fe was dominating with highest concentration in pickles, canned fruits and sauces at 143.3 ± 43.2, 83.64 ± 23.19 and 50.17 ± 15.1 mg/kg, respectively), followed by Cd in sauces (22.94 ± 6.91 mg/kg), Cr in juices (12.97 ± 3.91 mg/kg) and Pb in pickles (12.53 ± 3.77 mg/kg). Measured levels of these metals varied significantly and were relatively higher than their permissible limits. Univariate and multivariate analysis depicted strong association among Cr, Co, Pb and Fe and confirmed HMs contamination through natural and anthropogenic sources in processed foods. Health risk index (HRI) for Cd, Cr and Pb was greater than unity (<1.0), particularly in sauces, jams and canned fruits. Target hazard quotient (THQ) and hazard index (HI) of Cd, Cr and Pb were relatively high. But target cancer risk (TCR) assessment indicates that these metals were within the acceptable limit, except for Cd concentration in sauces, jams and canned fruits that may cause cancer to consumers.

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1. Introduction

Heavy metals (HMs) are among the major food chain contaminants triggering serious health effects even consumed in least concentration (Fathabad et al., 2018a). Natural processes (i.e. volcanic eruptions, leaching from soil and rocks etc.), and anthropogenic activities including mining, industrialization and urbanization are the main contributors of HMs adulteration in food chain (Ali et al., 2019; Tchounwou et al., 2012), and is the main concern in food safety, human health and quality assurance (Shaheen et al., 2016). Disproportionate use of chemical fertilizers, pesticides and polluted water for irrigation are the main factors responsible for HMs contamination, predominantly in fruits and vegetables (Amer et al., 2019).

Estimated, 90% of the HMs exposure to consumers occurs through contaminated food (Martorell et al., 2011), which contributes up to 30% of cancer in human along with other health disorders (Mansour et al., 2009). Foods either fresh or processed are contaminated with HMs (Hajeb et al., 2014), that become noxious when ingested above the tolerable limit (Dghaim et al., 2015). HMs possess various adverse health effect owing to inadequate mechanism of elimination from human body, non-biodegradable nature, long biological half-lives and potential to accumulate in different body parts (Shaheen et al., 2016). Therefore, bioaccumulation of HMs in human leads to mutagenesis, carcinogenesis, teratogenesis and heart, nervous system, liver, kidney, lungs, bone and spleen disorders (Parkar and Rakesh, 2018).

Cd concentration in food above 0.0004 mg/kg/day causes renal dysfunction, memory loss, cardiovascular disorders, cancer and even death (Fathabad et al., 2018a; Barone et al., 2018; Dghaim et al., 2015; Agoramoorthy et al, 2008). This metal mainly effects the renal system, causing irreversible damage to the renal tubules involved in the mechanisms of nutrient reabsorption (Rubio et al., 2018). Though, Cr involves in the synthesis of nucleic acids (DNA & RNA) and proper functioning of the immune system (Manore et al., 2009), but Cr^{6+} is lethal and even in a very low concentration (150 µg/g for adults and 0.2–5.5 µg/g for children) causes diabetes, heart attack and cancer (Kabatha-Pendias, 2001).

Elevated level of Co in human body results in vomiting, heart problems, and affects functioning of thyroid, causes nausea and over production of red blood cells (Parveen et al., 2020). Likewise, above the threshold level viz. 35 mg/day, Cu causes hair loss, skin infections and respiratory diseases (Khan et al., 2010). Nevertheless, Fe is important for red blood cells, enzymes activity and for appropriate immune functioning (Leung and Furness, 1999), but above its recommended level may cause diarrhea, dizziness, vomiting, nausea, joint pain, cardiovascular disorders and disturbs metabolic functions (Dghaim et al., 2015; Hashemi et al., 2017). Likewise, above permissible limits Pb damages the central nervous system (Hsu et al., 2006), exclusively in developing children and fetuses and cause nephropathies, alterations of the gastrointestinal tract and Alzheimer's disease (Paz et al, 2019; Fathabad et al., 2018a). Recommended daily intake of Zn is Zn in excessive amount viz. 21 mg/day (USEPA, 2002; Parveen et al., 2020), reduces immune function, causes obesity, diarrhea, kidney and liver failure and anemia (Singh et al., 2010), damages reproductive system (Nolan, 2003) and effects blood lipoprotein and copper level (Dghaim et al., 2015).

The presence of antioxidants and other biologically active ingredients in fruits makes them effective in the treatment of numerous diseases (Roba et al, 2016; Ji-yun et al, 2016). The use of processed foods and method of food preservation goes back to the time when human beings learned how to cook and store the food. Refrigeration, freezing, dehydration, acidification, irradiation, extrusion, extraction, filtering and packaging techniques involved in food preservation were invented in 20thcentury (Eicher-Miller et al., 2012). Although, food processing and preservation techniques are significant to meet the demand of growing human population. However, nature of food, harvesting time, mode of transportation, use of chemical preservatives, packaging and storage cause HMs contamination in processed foods specifically in developing countries where monitoring and managerial issues are ineffective and disorganized. Main populace in these countries use plants and animals based processed foods, which are one of the major sources of HMs exposure. In this context, present study was intended with the aim to estimate health risk assessment to consumers, predominantly associated with HMs concentration in the processed fruit-based food products available in the local markets of North Pakistan.

2. Materials and methods

2.1. Sampling

Based on availability, processed fruits-based products (n = 345) of different national and international brands viz. National, Shan, Shangrilla, Mitchell's, Fruit Tree and Nestle were collected from the local markets of Abbottabad, Haripur and Mansehra districts of Khyber Pakhtunkhwa. Pakistan. These samples were collected in triplicates and categorized in to different groups including "juices", "jam", "canned fruits", "sauces", "ketchup", "pickles", marmalades and tomato paste (102, 81, 39, 33, 33, 33, 15 and 9 samples, respectively) on the basis of product types.

2.2. Digestion and instrumentation

Processed fruit product samples were digested following the protocol as explained earlier by Parveen et al. (2020). Briefly, powder sample (~2g) was added into digestion tubes containing digestion mixture (HNO_3 and $HClO_4$) at 2:1. The mixture was heated on digester Pelican Kelplus- KES 20LRAL-TS at 120 °C until clear solution was obtained. Afterwards, final volume was adjusted up to 50 mL with deionized water. Along with a batch of 6 samples a blank was also prepared in the same way. Quantification of selected metals: Cd, Cr, Co, Cu, Fe, Pb and Zn was done using the flame atomic absorption spectrometer (Perkin Elmer S#80156060702) under optimum analytical conditions. Working standards for tested metals were prepared from stock solutions (1000 mg/L). Optimum analytical conditions maintained on atomic absorption spectrophotometer 'AAS' to selected metals using air-acetylene flame (Perkin Elmer S#80156060702) are mentioned in Table S1.

2.3. Statistical analysis

Analytical data were evaluated by univeriate and multivariate approaches using STATISTICA (StatSoft Inc, 1999) and SPSS Statistics V21.0 (IBM, Chicago USA). Cluster analysis (CA) and principle component analysis (PCA) were performed using Ward's and Varimex rotation methods to find compositional pattern of data set and sources of contamination. Graphical representations of data were done by using GraphPad Prism 8.0 (GraphPad Software, Inc. USA) and Sigma Plot 12.1 (Systat Software Inc. San Jose, California). Data were presented as mean ± SD for triplicate analysis.

2.4. Health risk assessment

Filed survey was conducted to document the information regarding the consumption of processed fruit-based products among the local communities of three districts: Haripur, Abbottabad and Mansehra of KP province of Pakistan. In total, 100 informants were participated in the field survey (Table S2). Data were collected using semi structured interviews and questionnaire comprising multiple choice questions. Information were gathered on family members, types of product consumed, and quantity consumed etc. (Table S3).

The health risk was calculated by finding the daily intake (mg/day/person) of selected metals through consumption of the processed fruit-based products and then relate them to the reference oral dose (Singh et al., 2010).

Estimated daily intake (EDI) was calculated following the procedure reported previously (Cherfi et al., 2014) using equation:

$$EDI = C \times I$$

where; 'C' represents the metal concentration in fruit products (mg/kg), and 'I' denotes daily intake of the product.

Health risk index (HRI) is a proportion between the estimated daily intake (EDI) of the metal and reference oral dose (RfD) for each metal and body weight (BW) of the consumers (Cui et al., 2004) and the HRI less than 1 for any metal consider safe for consumer (Parveen et al., 2020). The HRI was calculated using following equation:

$$HRI = \sum \frac{n(Cn \times Dn)}{RfD \times Bw}$$

where Cn is the mean metal concentration in specific fruit product on fresh weight basis (mg/kg); Dn is the average daily intake rate of a specific fruit product in a whole year; RfD showed safe level of exposure by oral intake for lifetime; and Bw is the average body weight.

Target hazard quotient (THQ) and *Hazard index* (HI) are used to evaluate the non-carcinogenic health risk to human (Yang et al., 2011). THQ was calculated using equation reported previously (USEPA, 2015).

$$THQ = \frac{C \times I \times 10^{-3} \times EFr \times EDtot}{RfD \times Bw \times ATn}$$

where C represents mean metals level in fruit product (mg/kg); I is the ingestion rate (g/day/person); Efr is the exposure frequency (days/year); EDtot is the total exposure duration (years); Bw is the average body weight adult (kg); and ATn is the averaging time, non-carcinogens (EDtot × 365 days/year).

Hazard index (HI) is the sum of hazard quotients for trace metals and was calculated by formula as reported by earlier (USEPA, 2006; USEPA, 2015).

$$HI = THQ_1 + THQ_2 + \ldots + THQ$$

where THQ1 − n is Target hazard quotients for 1 − n metals.

Target cancer risk (TCR) is intended to determine the cancer risk to consumers due to food consumption. The TCR in processed fruit products was calculated following the method reported previously (USEPA, 2006; USEPA, 2015)

$$TCR = \frac{Cb \times 1 \times 10^{-3} \times CPS \times \times EFr \times EDtot}{BWa \times ATc}$$

where ATc is averaging time carcinogens; carcinogens potency slope oral (µg/g/day) is CPS; Efr is the exposure frequency (days/year); EDtot is the total exposure duration (years); BWa is the average body weight; and Cb is metal concentration.

3. Results and discussion

Heavy metals including Cd, Cr, Co, Cu, Fe Pb and Zn were quantified in 345 samples of processed fruit based products categorized in to eight groups viz. sauces, ketchup and pickles (*n = 11 brands of each*), juices (*n = 34 brands*), jams (*n = 27 brands*), canned fruits (*n = 13 brands*), tomato paste and marmalades (*n = 3 and 5 brands, respectively*).

3.1. Comparative assessment of HMs in fruits' products

On the whole Fe was dominating in term of highest concentration in almost all brands of processed fruit-based products followed by Cd, Cr and Pb. Whereas, Co and Cu were usually at lowest levels (Table 1). Likewise, in many samples measured levels of HMs were below the detection limit.

As mentioned in Fig. 1a, in different brands of sauces Fe, Cd and Zn were highest in concentration (50.17 ± 15.1, 22.94 ± 6.92 and 4.100 ± 1.24 mg/kg, respectively). Relatively average concentration of Cd, Cu, Fe, Pb and Zn, in our samples were higher than previous reports from Bahrain (Musaiger, 2008) and Bangladesh (Haque et al., 2018). In ketchup samples, Fe, Cr and Pb were dominating in term of mean concentration levels (10.43 ± 3.5.725 ± 1.72 and 4.438 ± 1.33 mg/kg, respectively). Measured levels of all metals in ketchup were more than reported in tomato sauce from Nigerian (Adegbola, 2013) and in different brands of ketchup consumed by the inhabitants of Ghana (Boadi et al., 2012), Nigeria (Iwegbue et al., 2012) and Romania (David et al., 2008). In addition, average concentration of Pb (4.438 ± 1.33 mg/kg), was also higher than the recommended standard of Codex (2016) for ketchup (1.0 mg/kg). However, Fe concentration in our samples was relatively lower than reported earlier by Harmanescu et al., (2007) and David et al., (2008) from Romania.

In juice samples of 34 brands average levels of HMs concentration in different brands of juice was in subsequent order: Fe > Pb > Co > Cr > Zn > Cd > Cu. Comparatively, lower mean concentrations of Fe, Zn, Cu, Co, and Cr were reported in the fruit juice from Saudi Arabia (Farid and Enani, 2010),

Table 1 Measured levels of HMs (mg/Kg) in different fruit-based products (*n = 345*).

Samples	Sample Name	Company	Cd	Cr	Co	Cu	Fe	Pb	Zn
Sauces									
S1	Chilli garlic sauce	Mitchelle's	35.06 ± 0.57				41.41 ± 1.48	2.833 ± 0.41	0.958 ± 0.09
S2	Chilli garlic sauce	National	14.83 ± 0.72	9.704 ± 0.49			50.69 ± 1.16		
S3	Chilli garlic sauce	Shangrilla	18.93 ± 0.59	2.909 ± 0.92			70.27 ± 1.06	2.353 ± 0.30	0.256 ± 0.08
S4	Chilli garlic sauce	Saucy				0.091 ± 0.01			0.678 ± 0.03
S5	Chilli garlic sauce	Kinza		0.312 ± 0.15	0.159 ± 0.02	0.060 ± 0.00	37.87 ± 1.54		2.728 ± 0.40
S6	Chilli garlic sauce	Knorr		0.375 ± 0.03		1.155 ± 0.08	29.19 ± 1.05		12.91 ± 0.92
S7	Chilli garlic sauce	Shezan		3.907 ± 0.27		1.189 ± 0.25	20.48 ± 0.78	6.147 ± 0.23	10.11 ± 0.29
S8	Khatti methi imli	National					140.9 ± 6.70	1.935 ± 0.24	8.591 ± 0.53
S9	Khatti methi imli	Mitchelle's		0.141 ± 0.03	0.126 ± 0.03				1.371 ± 0.39
S10	Khatti methi imli	Shangrilla		0.718 ± 0.09	0.047 ± 0.02		3.280 ± 0.96		1.658 ± 0.14
S11	Khatti methi imli	Kinza					57.47 ± 2.49	0.224 ± 0.14	1.732 ± 0.08
	Max.		35.06	9.704	0.159	1.189	140.87	6.147	12.91
	Min.		14.83	0.141	0.047	0.060	3.279	0.224	0.256
	Mean		22.94	2.581	0.111	0.623	50.169	2.698	4.100
	SE		6.917	0.778	0.033	0.188	15.127	0.814	1.236
Ketchup									
K1	Tomato Ketchup	Shangrilla	10.76 ± 0.84	9.490 ± 0.54			30.14 ± 1.10	4.856 ± 0.66	0.160 ± 0.06
K2	Tomato Ketchup	National	5.609 ± 0.57	13.32 ± 0.48			27.93 ± 0.52	0.331 ± 0.09	5.969 ± 0.09
K3	Tomato Ketchup	Kinza	1.763 ± 0.31	0.528 ± 0.05			4.397 ± 1.00		1.435 ± 0.10
K4	Tomato Ketchup	Bake Parlor			0.140 ± 0.03		3.070 ± 0.69		1.520 ± 0.19
K5	Tomato Ketchup	Saucy				0.007 ± 0.00	0.888 ± 0.13		5.694 ± 0.45
K6	Tomato Ketchup	Mitchelle's			0.839 ± 0.03		0.297 ± 0.03		0.816 ± 0.04
K7	Tomato Ketchup	Knorr	0.815 ± 0.07			0.057 ± 0.01	5.761 ± 0.36		0.880 ± 0.09
K8	Tomato Ketchup	Best food		0.575 ± 0.04	0.032 ± 0.00	1.155 ± 0.23			
K9	Tomato Ketchup	Bake Parlor			0.171 ± 0.04		23.30 ± 1.07		
K10	Tomato Ketchup	Shezan		4.704 ± 0.44		1.590 ± 0.08	1.963 ± 0.29	10.66 ± 0.80	8.533 ± 0.68
K11	Tomato Ketchup	Open	1.478 ± 0.40	5.734 ± 0.33	0.639 ± 0.67	1.007 ± 0.07	6.595 ± 0.87	1.903 ± 0.16	0.211 ± 0.13
	Max.		10.76	13.32	0.839	1.590	30.14	10.66	8.533
	Min.		0.071	0.037	0.004	0.002	0.034	0.093	0.036
	Mean		4.086	5.725	0.364	0.763	10.43	4.439	2.802
	SE		1.232	1.726	0.109	0.230	3.146	1.338	0.845
Juices									
JU1	Mango Juice	Fruitavitals			0.449 ± 0.04			1.443 ± 0.13	1.668 ± 0.17
JU2	Mango Juice	Fruitien		2.040 ± 0.17	4.147 ± 0.24	1.283 ± 0.22	4.156 ± 0.11	2.918 ± 0.14	
JU3	Mango Juice	Nesfruta		1.917 ± 0.17		2.164 ± 0.25			
JU4	Mango Juice	Slice		1.678 ± 0.17		0.130 ± 0.01			
JU5	Mango Juice	Shezan		2.706 ± 0.08	6.476 ± 0.08	1.107 ± 0.04	16.38 ± 0.18	1.770 ± 0.30	
JU6	Mango Juice	Haleeb		34.96 ± 0.23	4.094 ± 0.15	0.271 ± 0.04	4.868 ± 0.38	1.967 ± 0.07	1.223 ± 0.20
JU7	Apple Juice	Fruitavitals		2.972 ± 0.13			33.95 ± 1.45		1.443 ± 0.19
JU8	Apple Juice	Fruitien		1.331 ± 0.27		0.218 ± 0.03	4.618 ± 0.68		2.028 ± 0.15
JU9	Apple Juice	Nesfruta	0.191 ± 0.01		9.895 ± 0.16		3.833 ± 0.52		0.663 ± 0.17
JU10	Apple Juice	Tops	0.192 ± 0.01	1.163 ± 0.20	0.409 ± 0.02	0.873 ± 0.05		9.318 ± 0.45	2.740 ± 0.37
JU11	Apple Juice	Shezan		1.502 ± 0.17		0.177 ± 0.08	15.13 ± 1.59	8.392 ± 0.24	
JU12	Apple Juice	Tops	0.044 ± 0.01	2.485 ± 0.46	2.533 ± 0.36	5.799 ± 0.31			7.860 ± 0.42
JU13	Apple Juice	Haleeb		3.696 ± 0.05	10.66 ± 0.49	0.495 ± 0.02	5.866 ± 0.38		0.567 ± 0.12

Table 1 (*continued*)

Samples	Sample Name	Company	Cd	Cr	Co	Cu	Fe	Pb	Zn
JU14	Guava Juice	Fruitiens		23.93 ± 0.10		0.828 ± 0.07	1.505 ± 0.26	0.221 ± 0.14	2.173 ± 0.27
JU15	Guava Juice	Fruitavitals			15.28 ± 0.49	1.149 ± 0.23	38.66 ± 0.50	7.318 ± 0.45	2.175 ± 0.39
JU16	Guava Juice	Tops Pouch	0.299 ± 0.00	2.992 ± 0.24			3.083 ± 0.54		0.345 ± 0.06
JU17	Guava Juice	Tops	1.171 ± 0.24	0.623 ± 0.03			77.13 ± 2.16	15.08 ± 0.80	0.495 ± 0.02
JU18	Guava Juice	Shezan	0.118 ± 0.03	0.191 ± 0.01	0.168 ± 0.02	1.057 ± 0.22	14.08 ± 3.22		0.781 ± 0.08
JU19	Red Grapes Juice	Fruitavitals	24.94 ± 0.10		3.486 ± 0.22	0.080 ± 0.02	37.85 ± 2.22	7.331 ± 0.45	11.24 ± 0.58
JU20	Red Grapes Juice	Fruitien		1.842 ± 0.11		0.614 ± 0.07	9.655 ± 0.48		1.324 ± 0.09
JU21	Red Grapes Juice	Tops	0.268 ± 0.04	0.589 ± 0.02				14.87 ± 0.50	0.492 ± 0.02
JU22	Peach Juice	Fruitavitals	0.491 ± 0.01	0.733 ± 0.02	0.040 ± 0.03	0.875 ± 0.14	70.64 ± 1.86	12.88 ± 0.43	3.275 ± 0.53
JU23	Peach Juice	Fruitien	0.159 ± 0.03	2.202 ± 0.23		0.788 ± 0.07	3.688 ± 0.46		
JU24	Peach Juice	Rani float			7.053 ± 0.39	0.195 ± 0.02			3.685 ± 0.46
JU25	Pineapple Juice	Fruitien		5.141 ± 0.31		0.519 ± 0.03	3.593 ± 0.69		4.005 ± 0.49
JU26	Pomegranate Juice	Fruitien	0.441 ± 0.03	2.783 ± 0.29	4.287 ± 0.39	1.946 ± 0.12	0.962 ± 0.09		
JU27	Lychee Juice	Tops		1.506 ± 0.18		0.142 ± 0.03		8.508 ± 0.41	
JU28	Sugarcane Juice	Open		5.076 ± 0.22		3.628 ± 0.55	13.90 ± 0.26	11.29 ± 1.04	6.930 ± 0.13
JU29	Fruit Punch	Shezan		0.271 ± 0.04	4.840 ± 0.40	0.553 ± 0.10	2.422 ± 0.38		5.565 ± 6.60
JU30	Fruit Punch	Shezan 250	1.488 ± 0.19	1.615 ± 0.33		1.191 ± 0.24	3.908 ± 0.15	0.293 ± 0.07	6.833 ± 0.75
JU31	Kinnow	Fruitavitals	0.868 ± 0.12	2.910 ± 0.21		0.552 ± 0.10	7.833 ± 0.26	2.814 ± 0.25	0.368 ± 0.07
JU32	Orange Juice	Fruitien		4.190 ± 0.27	2.148 ± 0.19	1.641 ± 0.20	0.875 ± 0.05	8.856 ± 0.22	1.640 ± 0.20
JU33	Orange Pulpy	Open	0.050 ± 0.01	2.623 ± 0.36	4.378 ± 0.34	0.588 ± 0.02	2.738 ± 0.39		0.381 ± 0.03
JU34	Orange Juice	Shezan		3.667 ± 0.18	5.805 ± 0.62	1.677 ± 0.16	6.190 ± 0.30		2.172 ± 0.27
	Max.		24.94	34.96	15.29	5.799	77.13	15.09	11.24
	Min.		0.044	0.191	0.040	0.080	0.875	0.221	0.345
	Mean		2.194	4.115	4.786	1.091	14.90	6.781	2.772
	SE		0.376	0.706	0.821	0.187	2.556	1.163	0.475
Jams									
JM1	Strawberry Jam	Mitchell's				0.048 ± 0.02		3.881 ± 0.19	7.398 ± 0.36
JM2	Strawberry Jam	National	13.60 ± 0.59	0.402 ± 0.01	5.140 ± 0.29	1.741 ± 0.08		5.904 ± 0.59	
JM3	Strawberry Jam	Mitchell's					26.64 ± 0.55	1.537 ± 0.24	2.384 ± 0.03
JM4	Strawberry Jam	Fruit tree	4.421 ± 0.33	1.308 ± 0.08			4.654 ± 0.72	3.275 ± 0.27	7.318 ± 0.43
JM5	Strawberry Jam	Salman's	12.73 ± 0.94		2.156 ± 0.20		22.20 ± 1.14		4.382 ± 0.26
JM6	Strawberry Jam	Shezan		2.791 ± 0.08				6.295 ± 0.39	1.530 ± 0.62
JM7	Apple Jam	Mitchell's					11.48 ± 0.82	0.700 ± 0.13	4.114 ± 0.48
JM8	Apple Jam	National	74.73 ± 4.45	1.231 ± 0.17	0.163 ± 0.02			5.796 ± 0.31	
JM9	Apple Jam	Fruit tree		2.483 ± 0.42			24.33 ± 1.12	2.427 ± 0.31	1.472 ± 0.23
JM10	Apple Jam	Salman's	8.862 ± 0.22	1.502 ± 0.18	1.169 ± 0.22	0.162 ± 0.06	22.37 ± 1.89		3.895 ± 0.17
JM11	Mixed Fruit Jam	Mitchell's				0.077 ± 0.02		4.919 ± 0.13	3.526 ± 0.23
JM12	Mixed Fruit Jam	National	30.68 ± 0.54		0.162 ± 0.02			3.833 ± 0.19	5.946 ± 0.25
JM13	Mixed Fruit Jam	Mitchell's	19.09 ± 0.26				19.81 ± 1.50	8.730 ± 0.40	1.322 ± 0.06
JM14	Mixed Fruit Jam	Fruit tree		2.789 ± 0.26		0.033 ± 0.01	11.89 ± 1.62	2.669 ± 0.17	4.264 ± 0.13
JM15	Pineapple Jam	Mitchell's	27.60 ± 0.80	1.566 ± 0.10	2.291 ± 0.25		16.09 ± 1.03		41.20 ± 1.15
JM16	Pineapple Jam	Fruit tree		4.105 ± 0.21			43.59 ± 1.02	1.543 ± 0.31	5.180 ± 0.27
JM17	Pineapple Jam	National	2.895 ± 0.16				11.90 ± 1.19	1.530 ± 0.37	8.066 ± 0.70
JM18	Mango Jam	Mitchell's				0.074 ± 0.02		1.672 ± 0.19	3.588 ± 0.09
JM19	Mango Jam	National	37.31 ± 0.48	0.801 ± 0.02	48.42 ± 1.57	16.94 ± 0.38		15.71 ± 0.42	
JM20	Mango Jam	Salman's	3.199 ± 0.32	1.711 ± 0.21		0.188 ± 0.02		2.667 ± 0.05	0.806 ± 0.07

(*continued on next page*)

Table 1 (*continued*)

Samples	Sample Name	Company	Cd	Cr	Co	Cu	Fe	Pb	Zn
JM21	Mango Jam	Ahmad's	5.066 ± 0.33	0.501 ± 0.00	0.412 ± 0.00	0.003 ± 0.00			4.776 ± 0.33
JM22	Blackcurrant	Mitchell's	24.08 ± 0.27		3.192 ± 0.29		19.86 ± 0.33		3.406 ± 0.39
JM23	Blackcurrant	National		7.611 ± 0.22			6.133 ± 0.30	7.807 ± 0.29	1.354 ± 0.30
JM24	Blackcurrant	Fruit tree	8.941 ± 0.45					7.538 ± 0.60	5.461 ± 0.33
JM25	Blackcurrant	Salman's		0.710 ± 0.02	0.697 ± 0.08	0.138 ± 0.05		5.600 ± 6.55	11.29 ± 0.38
JM26	Banana jam	Shezan		1.556 ± 0.15		0.269 ± 0.05		22.65 ± 0.51	1.837 ± 0.25
JM27	Orange jam	Shezan	0.267 ± 0.05	5.514 ± 0.38		0.490 ± 0.03		18.22 ± 1.05	7.334 ± 0.32
	Max.		74.73	7.611	48.42	16.94	43.59	22.65	41.20
	Min.		0.267	0.402	0.162	0.003	4.654	0.700	0.806
	Mean		18.23	2.286	6.380	1.680	18.53	6.132	5.910
	SE		3.509	0.440	1.228	0.323	3.567	1.180	1.137
Canned Fruits									
C1	Fruit Cocktail	California			0.256 ± 0.08	0.073 ± 0.01	23.96 ± 0.18		1.768 ± 0.18
C2	Fruit Cocktail	Italia		1.196 ± 0.24	0.226 ± 0.01	0.796 ± 0.04	28.96 ± 0.55		0.542 ± 0.04
C3	Fruit Cocktail	Farm Fresh		1.770 ± 0.08		0.403 ± 0.01	14.31 ± 1.30		
C4	Fruit Cocktail	Fruitamins			0.083 ± 0.01	0.623 ± 0.08	17.95 ± 1.01		
C5	Pineapple	Fine life	9.577 ± 0.53			1.204 ± 0.16	26.67 ± 0.47	1.744 ± 0.22	
C6	Pineapple	Mitchell's			0.561 ± 0.06		97.29 ± 1.21		19.38 ± 0.38
C7	Pineapple	OK	26.10 ± 0.68		3.320 ± 0.08		60.67 ± 1.19	0.717 ± 0.12	18.02 ± 0.30
C8	Pineapple	California	0.306 ± 0.01		23.17 ± 0.55	1.057 ± 0.11	138.2 ± 3.70	18.78 ± 0.64	8.914 ± 0.14
C9	Pineapple	Pollac		0.161 ± 0.06		1.581 ± 0.28	21.85 ± 2.39	3.280 ± 0.40	4.930 ± 0.27
C10	Strawberry	Italia	0.796 ± 0.01	8.443 ± 0.30	19.93 ± 0.11	0.997 ± 0.01	195.4 ± 8.56	16.78 ± 0.46	60.11 ± 1.27
C11	Cherry	Italia	2.889 ± 0.18		26.26 ± 1.40	0.594 ± 0.04	209.9 ± 9.09	20.81 ± 0.35	11.36 ± 0.92
C12	Pear halves	California	11.88 ± 0.15		12.12 ± 0.19		186.5 ± 4.26	20.38 ± 0.93	14.90 ± 0.46
C13	Peach halves	California		15.07 ± 0.11		1.425 ± 0.33	65.80 ± 4.66	5.750 ± 0.37	1.817 ± 0.21
	Max.		26.10	15.07	26.26	1.58	209.9	20.81	60.11
	Min.		0.306	0.161	0.083	0.073	14.309	0.717	0.542
	Mean		8.593	5.328	9.547	0.875	83.65	11.03	14.17
	SE		2.383	1.478	2.648	0.243	23.20	3.06	3.93
Tomato paste									
TP1	Tomato Paste	Lui	8.043 ± 0.35	2.563 ± 0.16			15.04 ± 0.58		3.736 ± 0.39
TP2	Tomato Paste	Mitchell's	3.480 ± 0.57	0.924 ± 0.08			41.57 ± 0.62	0.517 ± 0.03	0.175 ± 0.04
TP3	Tomato Paste	White Pearl		2.417 ± 0.23		3.907 ± 0.15	9.570 ± 0.93	16.69 ± 0.41	14.46 ± 0.78
		Max.	8.043	2.563		3.907	41.57	16.69	14.46
		Min.	3.480	0.924		3.907	9.570	0.517	0.175
		Mean	5.761	1.968		3.907	22.06	8.603	6.122
		SE	3.326	1.136		2.256	12.737	4.967	3.535
Marmalades									
MM1	Citrus Marmalade	Salman's	3.987 ± 0.04	2.245 ± 0.17		0.045 ± 0.02	12.99 ± 0.35	3.463 ± 0.27	2.941 ± 0.10
MM2	Golden Mist	Mitchelle's			0.695 ± 0.03				2.096 ± 0.22
MM3	Orange Marmalade	National		1.441 ± 0.12	0.119 ± 0.00	0.004 ± 0.00	4.243 ± 0.11		2.446 ± 0.44
MM4	Harar murabba	Open		1.219 ± 0.21		4.952 ± 0.26	9.936 ± 0.41	0.565 ± 0.12	7.944 ± 0.10
MM5	Saib murabba	Open	3.987 ± 0.04	2.245 ± 0.17		0.045 ± 0.02	12.99 ± 0.35	3.463 ± 0.27	2.941 ± 0.10
	Max.		3.987	2.245	0.695	4.952	12.998	3.463	7.944
	Min.		3.987	1.219	0.119	0.004	4.243	0.565	2.096

Table 1 (*continued*)

Samples	Sample Name	Company	Cd	Cr	Co	Cu	Fe	Pb	Zn
	Mean		3.987	1.635	0.407	1.667	9.059	2.014	3.857
	SE		1.783	0.731	0.182	0.745	4.051	0.900	1.725
Pickles									
A1	Mixed Achar	Natoional	8.518 ± 0.39		8.275 ± 0.35		317.9 ± 10.20	26.16 ± 0.27	10.78 ± 0.68
A2	Mango	Natoional	0.869 ± 0.07	19.64 ± 0.53	12.33 ± 0.44	2.936 ± 0.09	213.3 ± 4.27	13.57 ± 0.62	14.77 ± 0.66
A3	Mango	Mitchell's	2.395 ± 0.06	19.31 ± 0.48	2.611 ± 0.24	1.070 ± 0.09	186.7 ± 8.16	8.720 ± 0.22	15.01 ± 0.07
A4	Lime and chilli	Mitchell's	1.350 ± 0.27	19.15 ± 0.50	20.21 ± 0.35	0.004 ± 0.00	233.1 ± 9.33	17.29 ± 0.38	13.07 ± 0.25
A5	Mixed Achar	Ahmad's	1.571 ± 0.27		12.49 ± 0.10	0.510 ± 0.08	158.8 ± 9.19	14.17 ± 0.26	11.29 ± 0.39
A6	Mixed Achar	Shangrilla	3.936 ± 0.10		7.205 ± 0.27	0.487 ± 0.06	158.5 ± 2.61	14.17 ± 0.30	14.05 ± 0.72
A7	Mango kasundi	Mitchell's	1.970 ± 0.13		21.48 ± 0.60	6.073 ± 0.09	182.2 ± 3.75	18.02 ± 0.48	14.69 ± 2.68
A8	Chilli achar	Natoional		17.01 ± 0.44		3.276 ± 0.25	32.70 ± 2.21	0.523 ± 0.06	9.842 ± 0.59
A9	Mixed Achar	Shezan		11.87 ± 0.24	4.399 ± 0.39	0.742 ± 0.08	71.24 ± 1.91	9.870 ± 0.68	10.31 ± 0.82
A10	Phalli Achar	Open	0.091 ± 0.01	2.394 ± 0.35	9.901 ± 0.17	4.832 ± 0.25	6.158 ± 0.027	2.877 ± 0.19	3.233 ± 0.19
A11	Chilli Achar	Open		1.474 ± 0.23	3.691 ± 0.15	6.499 ± 0.40	16.10 ± 2.06		2.000 ± 0.05
	Max.		8.518	19.64	21.48	6.499	317.95	26.16	15.01
	Min.		0.091	1.474	2.611	0.004	6.158	0.523	2.000
	Mean		2.588	12.98	10.26	2.643	143.35	12.54	10.82
	SE		0.780	3.913	3.093	0.797	43.22	3.780	3.263

Alphabetical letters represents significant differences at $p < 0.05$.

but were higher than the fruit juice sold in the markets of Jordan (Massadeh and Al-massaedh, 2018), Pakistan (Anwar et al., 2014), Iraq (Al-Mayaly, 2013), Poland (Szymczycha-Madeja and Welna, 2013), Turkey (Hamurcu et al., 2010) and Italy (Coco, 2006). Threshold levels of Cd, Cu, Pb and Zn in fruit juice are 0.005, 0.01, 0.01 and 0.2 mg/kg, respectively (WHO, 1984). Conversely, measured levels of these metals in our case were far beyond the permissible limits, therefore may cause severe threat to consumers' health. There was no significant difference in Fe and Cd levels in different brands of Jam (Table 1). Relatively, measured levels of Pb, Cd and Cr in our samples were significantly higher than fruit jam consumed in the markets of India and Rwanda (Asema and Parveen, 2018; Poornima et al., 2014; Mukantwali et al., 2014). Estimated levels of Cu, Fe, Pb, Zn and Cd levels in pineapple jam sold in the local markets of Rwanda were 3.29, 2.95, 0.77, 3.55, 0.01 mg/kg, respectively ().

The descending order of HMs quantified in various brands of canned fruits was: Fe > Zn > Pb > Co > Cd > Cr > Cu. Fe was in highest concentration, followed by Zn and Pb (88.64 ± 23.1; 14.73 ± 3.93 and 11.02 ± 3.05 mg/kg, respectively). Source of raw material, mode of transportation, storage and packaging, pH, concentration of oxygen in space between product and can head and quality of coating material used (Iwegbue et al., 2009), and industrial processes i.e. kneading, cutting, rolling, sheeting, and chopping (Jothi and Uddin, 2014), contribute significantly in HMs contamination in canned fruits. Comparatively, measured values of HMs in our samples were higher than reported previously in canned fruits (Divis et al., 2017; Leao et al., 2016; Rafique et al., 2008). Furthermore, average Cd level in our samples was considerably higher than European Union standard "0.05 mg/kg" (EU, 2006b), which is alarming and may leads to serious adverse health effects.

In tomato paste, mean value of Fe was highest (22.06 ± 12.7 mg/kg), followed by Pb and Zn (8.603 ± 4.96 and 6.122 ± 3.53 mg/kg, respectively). Relatively, all HMs in tomato paste were higher than reported from Nigeria (Adegbola, 2013; Hadiani et al., 2014), Iran (Hadiani et al, 2014), Turkey (Kocak et al., 2005), and Romania (David et al., 2008)). Furthermore, mean values of Pb and Cd in tomato paste were above the threshold levels of these metals viz. 1.0 and 0.05 mg/kg, respectively (Codex, 2016). To best of our knowledge, HMs metals have rarely been investigated in marmalades. Present study revealed that Fe, Cd and Zn metals were highest (9.058 ± 4.96, 3.986 ± 1.78 and 3.856 ± 1.72 mg/kg, respectively) in different brands of marmalades, while Co was lowest in concentration (0.406 ± 0.181 mg/kg). In 11 different brands of pickles, Fe was highest with an average of (143.3 ± 43.2 mg/kg), followed by Cr and Pb at 12.97 ± 3.91 and 12.53 ± 3.77 mg/kg. Comparatively, measured levels HMs in our samples were higher than reported previously from Behrain and Turkey (Musaiger, 2008; Tuzen, 2007).

Fig. 1b, revealed that Fe concentration was reasonably higher in all brands of fruits' produced, followed by Cd, Zn, Cr, Pb, Co and Cu. Cd metal ranged from 22.94 ± 6.91 to 2.193 ± 0.376 mg/kg in different brands of sauces and fruit juice, respectively. This indicates that even the lowest level of Cd in our samples was significantly higher than the permissible limit viz. 0.05 mg/kg (Codex, 2016), that may lead to serious health issues in consumers specifically associated with this metal (Baldwin and Marshall 1999). Municipal sewage, chemical industries, Ni-Cd batteries, metallic alloys, tobacco smoking, smelting, plating, and non-recycled Cd products contributing significantly in increasing levels of Cd in soil, water and food chain (Divis et al., 2017; Poornima et al., 2014; Chakraborty et al., 2013; Tiimub and Afua, 2013; ATSDR, 1999). Comparatively, Cr was highest in different brands of fruit juice, jams and ketchup (12.97 ± 3.93, 5.792 ± 1.72, 5.327 ± 1.47 mg/kg, respectively). And these values were relatively higher than the recommended level of Cr in canned or processed foods is 0.4 mg/kg (FAO/WHO, 2003). However, in other products i.e. canned fruits, sauces, tomato paste, marmalades and pickles measured levels of Cr were within safe limit.

Average concentration of Co was highest in pickles, followed by canned fruits, jams, juices, marmalades, ketchup, sauces, and tomato paste. Measured levels of Co in fruit juice were relatively higher than reported previously from Saudi Arabia (Farid and Enani, 2010) and Ghana (Ackah et al., 2014). In processed foods, Co concentration should not be more than 0.05 mg/kg (EU, 2006a), but in except tomato paste this metal was significantly higher than threshold level, which may impose adverse health effects on consumers Use of copper vessels in jams ensures equal heat distribution, lowers cooking time, and preserves fruit colors. In studied samples, Cu concentration was ranged 0.623 ± 0.18 mg/kg (sauces) to 3.907 ± 2.23 mg/kg (tomato paste). Permissible limit for Cu in fruits' juice is 5 mg/kg, whereas in tomato ketchup, sauces and tomato paste is 50 mg/kg (FAO/WHO, 2003). Therefore, in our samples Cu concentration lies within permissible limits.

Maximum permissible limit for Fe in processed foods is 50 mg/kg (Health Ministry, 1988). Pickles, canned fruits and sauces were among the top three groups having the highest levels of Fe at 143.3 ± 43.2, 83.64 ± 23.1 and 50.16 ± 15.1 mg/kg, respectively. Extensive use of Fe in machinery or steel containers in food processing industries, use of iron rich spices in sauces and pickles may increase its concentration processed foods. Likewise, presence of acids in different fruits may cause leeching of iron, particularly if such fruits are stored or packed in iron or steel containers (Ogidi et al., 2017). Pb concentration was significantly higher in pickle samples, canned fruits and tomato paste (12.53 ± 3.77, 11.02 ± 3.05 and 8.602 ± 4.96 mg/kg, respectively) compared to its permissible limits viz. 1 mg/kg for canned fruits, jams and preserved tomatoes and 0.03 mg/kg for juices (Codex, 2016). Pb concentration in our samples was in agreement with previous reports from Bangladesh (Tasnim et al., 2010) and Iran (Fathabada et al., 2018b). High concentration of Pb in canned fruits and paste may be attributed to the soldering process (Divis et al., 2017), while Pb deposition in soil and atmosphere, use of pesticides and fertilizers, harvesting techniques, storage conditions, transportation, processing machinery, water pipes and containers, gasoline and burning paints may involve in Pb contamination in processed foods (Jothi and Uddin, 2014). It has been reported that about 90% daily exposure to Pb in human comes from the food (Krejpcio et al., 2005), which is lethal to human and causes various types of cancer (Shariatifar et al., 2017). Average concentration of Zn in all products ranged 2.771 ± 0.47 (in juices) to 14.17 ± 3.93 mg/kg (in canned fruits). Measured level of Zn was much higher in canned fruits than permissible limit that is 5 mg/kg (FAO/WHO, 2003). Maximum level of zinc in canned fruit might be due to its application in metal alloys used in making cans.

3.2. Correlations

Interrelationship among the measured levels of mean HMs concentration were investigated in terms of correlation varimax. Results of Pearson's correlation coefficient analysis of HMs' concentration in processed fruit-based products are mentioned in Table 2. Except Cd, all metals showed associations with each other. This might be due the fact that Cd has different origin than rest of the metals quantified in our samples. Highly significant positive associations were observed between Co-Pb and Cr-Fe at r = 0.956 and 0.843 ($p < 0.01$), respectively. Whereas, Co-Zn, Pb-Zn, Fe-Zn and Fe-Pb also depicted strong positive relationships with r = 0.841, r = 0.813, r = 0.781 and r = 0.746, respectively ($p < 0.05$). Such a strong association between these metals suggests their common sources of contamination in processed foods such as various activities involve in food processing industries. However, inverse relationships showed depletion or enrichment of specific elements at the cost of others as seen in case of negative correlations of Cd with other metals (Table 2). Furthermore, negative associations suggest that Cd content are not controlled by single source factor and may be synergistic effect of both natural and anthropogenic activities (Suresh et al., 2012). The quartile distribution of HMs in different categories of processed fruit products was assessed as shown in box and whisker plot (Fig. 2). All targeted metals demonstrated broad range spread over various orders of magnitude. Cd, Cr, Pd, Zn and Fe showed broad and symmetrical distribution, whereas Co and Cu depicted broad ranged asymmetrical spread in the quartile values. Narrow range distribution was observed in the case of Mg, Zn, Cu, and Cd.

3.3. Multivariate analysis

Multivariate analysis was performed using cluster analysis (CA) and principal component analysis (PCA) to identify the sources of heavy metals in selected samples. BioVinci software was used for hierarchal cluster analysis of HMs in processed fruit samples (Fig. 3), depicted three main clusters. Based on average distribution pattern of HMs, two main clusters were identified for heavy metals. Among HMs, Cd-Zn were placed in first cluster, whereas Fe, Cr, Pb, Co and Cu were grouped in second cluster. The second cluster comprises two subgroups viz. Fe-Cr and Pb-Co, while Cu was in separate group. Likewise, based on distribution of HMs, different categories of processed fruit-based products were also in two main clusters, which were composed of sub-clusters. In first cluster, pickles and canned fruits were closely associated, accompany by tomato paste. Marmalades, ketchup, juice and jams were grouped in second cluster, whereby ketchup and juice were closely linked along with marmalades and jams. However, sauces were placed in separate group. Hierarchal cluster analysis specifies close association of HMs metals, which might be due to similarities in distribution pattern and source of contamination in the studied samples. Though, Cd is a carcinogenic metal, but was bunched with Zn that is an essential metal and within the permissible limit.

Principle component analysis (PCA) is extensively used to estimate the contribution of natural and anthropogenic

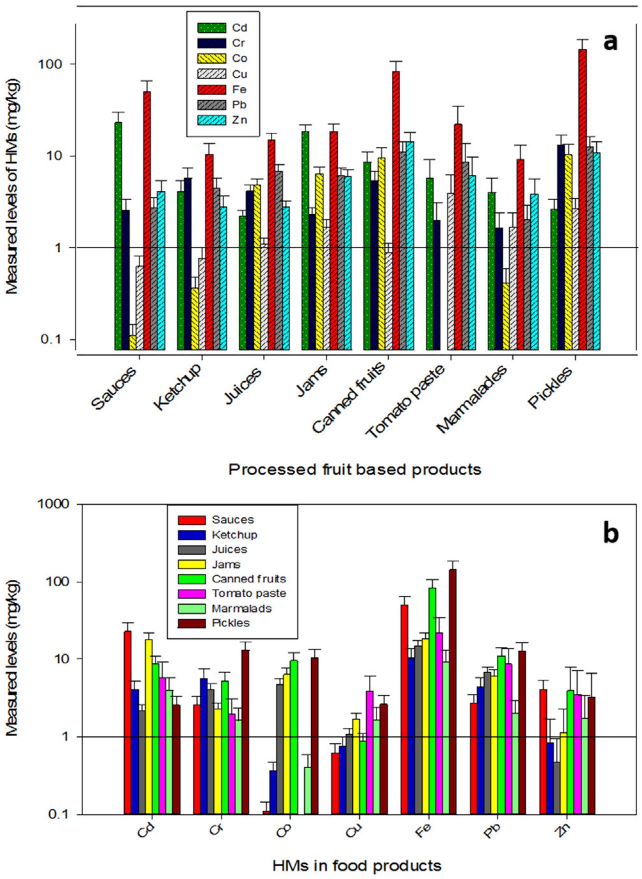

Fig. 1 Comparative assessment of heavy metals in processed fruits' products.

sources of HMs contamination in water, soil, air and plant samples (Liu et al., 2015). Mean values of HMs in processed fruits' products were treated by PCA using Varimax rotation with Kaiser Normalization to find out their compositional pattern and sources of contamination. Two principle components viz. PCA1 and PCA2 were extracted based on eigenvalue (>1) and accumulated variance explained 81.88% of the total variations (Table 3). First component contributes 57.96% variation of the total variation with maximum loading of four metals (Pb, Fe, Cr and Co), with percentage variance of 0.937, 0.929, 0.896 and 0.877, respectively. The contribution rate of these metals in PC1 revealed that main sources of contamination of these metals are related to each other. Such as natural phenomenon including mineral weathering, biogenic and forest fires, erosion and volcanic activities, and anthropogenic activities like industrial processing, use of chemical fertilizers and pesticides, municipal water and mining contributing considerably Co, Cr, Fe and Pb contamination in food chain and processed foods as well (Singh et al., 2018). The contribution of second component was 23.92% of the cumulative variation and had maximum loading of Zn (0.836%) and Cd (0.811%). This indicates that Zn and Cd had common origin in processed fruits' products. Fig. 4a, indicates that Pb, Fe, Cr and Co metals shares come sources of contamination in processed fruits' products shares, while Cd and Zn come from other sources. In Fig. 4b, solid symbols represent the mean concentration of HMs in each food type. The position of black circles in a plot relative to the direction of green lines approximates correlations between food and the gradient of element concentrations. The lengths of green lines indicate the overall contribution of each food to the analysis. The directions of the green lines indicate the correlation with each axis i.e. vector lines parallel to an axis are highly correlated with that axis, while angles between the vector lines show correlations between food types.

3.4. Health risk assessment

Health risk assessment associated with HMs' contamination in processed fruits' products was calculated by estimating health risk index (HRI), target hazard quotient (THQ), hazard index (HI) and target cancer risk (TCR). Heat map of HRI associated with Fe, Zn, Cu, Co, Cr, Cd and Pd concentration in 345 samples of processed fruits' products categorized into eight different groups was generated using GraphPad Prism version 8.0 (Fig. 5a&b). For each group, HMs concentration was presented in each rectangle of the heat map, whereby red colors represent high metal concentrations and blue color indicates low concentrations. Measured levels of HRI for Co, Cu, Fe and Zn (Fig. 5a), in all categories of processed fruits' products were within the safe limit (<1.0). However, HRI values for Cd, Cr and Pb were greater than unity in all samples and could be health risk to consumers (Parveen et al., 2020). The HRI values for Cd were higher in almost all groups of fruits' products, particularly in sauces, jam, and canned fruit. Likewise, HRI associated with Pb except sauces and marmalades and Cr except sauces, jams, tomato paste and marmalades was more than unity. Health protection standard of lifetime risk for THQ and HI is 1.0 (USEPA, 2006).

In the present study, THQ and HI values of HMs concentrations in all different groups of processed fruits' products (Fig. 5b&c) were within the safe limit (<1.0). Consequently, our findings demonstrated that ingestion of these products pertaining to these metals is safe and will not cause non-carcinogenic risk to consumers. The THQ and HI values of Cd, Cr and Pb were relatively higher than rest of the studied metals, therefore further assessed for TCR. As demonstrated by USEPA (2006), acceptable cancer risk limit for carcinogenic metals i.e. Cd, Cr and Pb is ranged 1×10^{-4} to 1×10^{-6}. Among, the studied samples TCR values for Cd, Cr and Pb were found within the acceptable limit (Fig. 6). However, TCR values of Cd was alarming in sauces, jams and canned fruits and may cause cancer to consumers over long-time exposure. Likewise, Cr and Pb concentration was alarming in pickles and canned fruits.

4. Conclusion

Present study was aimed to assess health risk related to HMs consumption present in processed fruit-based products sold in the local markets of North Pakistan. In total seven metals viz. Cd, Cr, Co, Cu, Fe, Pb and Zn were quantified in 345 samples categorized into eight groups (Sauces, Ketchup, Juices, Jams, canned fruits, tomato paste, marmalades and pickles). On the whole, Fe was dominating in term of concentration, followed by Cd, Cr and Pb. Whereas, Co and Cu were lowest in concentration. Pickles contain highest concentration of Fe, Pb and Co, while sauce, juice, canned fruits and tomato paste were rich in Cd, Cr, Zn and Cu, respectively. ANOVA test

Table 2 Correlation coefficient matrix of HMs in processed fruits' products.

HMs	Cd	Cr	Co	Cu	Fe	Pb	Zn
Cd	1.000						
Cr	−0.405	1.000					
Co	−0.200	0.626	1.000				
Cu	−0.315	0.104	0.529	1.000			
Fe	−0.074	0.843**	0.731	0.152	1.000		
Pb	−0.350	0.687	0.956**	0.428	0.746*	1.000	
Zn	−0.055	0.508	0.841*	0.163	0.781*	0.813*	1.000

** Correlation is significant at the 0.01 level (2-tailed).
* Correlation is significant at the 0.05 level (2-tailed).

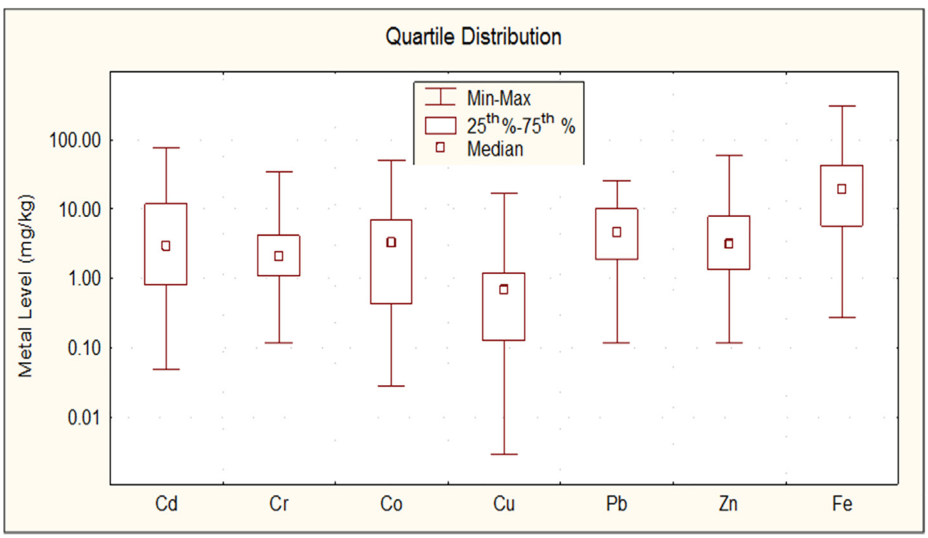

Fig. 2 Quartile distribution of heavy metals in studied samples.

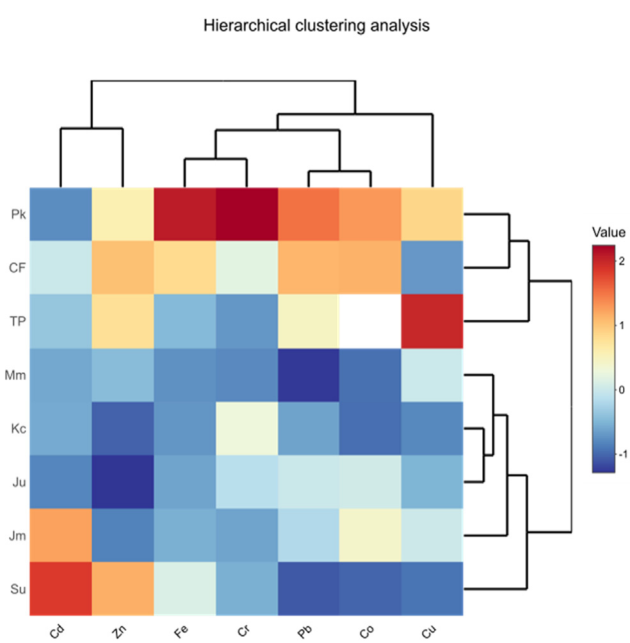

Fig. 3 Hierarchal clustering of heavy metals in processed fruits' products.

Table 3 Principal component analysis (PCA) of HMs in processed fruit products.

Variables	PC 1	PC 2
Eigen value	4.057	1.605
Total Variance (%)	57.96	23.92
Cumulative Variance (%)	57.91	80.88
Cd	−0.347	0.811
Cr	0.896	−0.159
Co	0.877	0.020
Cu	0.665	−0.349
Fe	0.929	0.317
Pb	0.937	−0.015
Zn	0.428	0.836

depicted that HMs contaminations in processed fruits' products varied considerably. Univariate and multivariate analysis revealed not only strong association among HMs i.e. Cr, Co, Pb and Fe, but also confirmed the contribution of natural and anthropogenic sources in processed foods' contamination. Measured levels of Cr in different brands of fruit juice, jams and ketchup were higher than the recommended level in processed food. The HRI values for Cd, Cr and Pb were greater

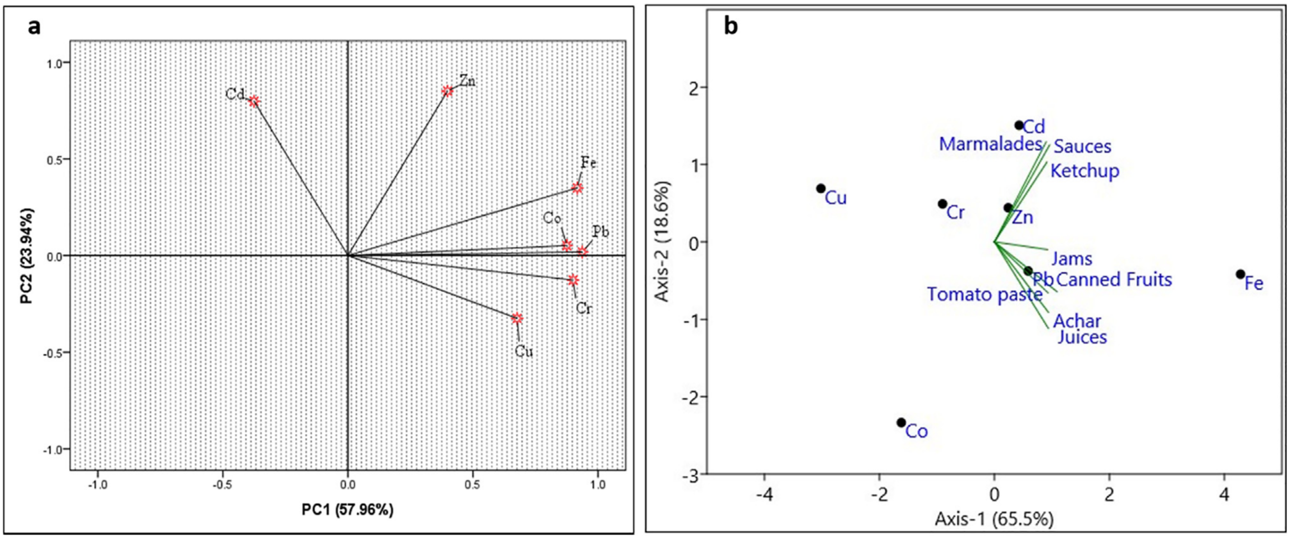

Fig. 4 Principle component analysis of heavy metals' concentration in samples analyzed.

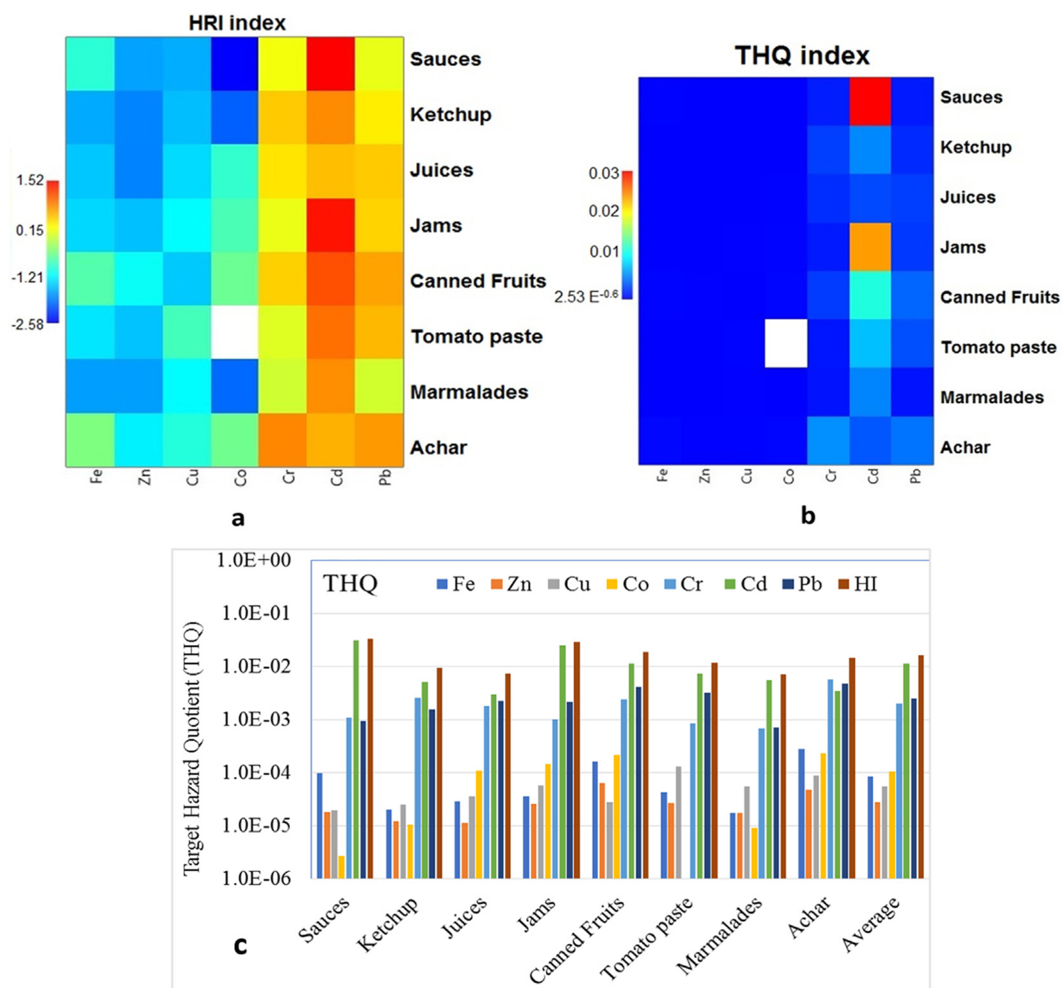

Fig. 5 HRI, THQ and HI associated with HMs contamination in processed fruits' products.

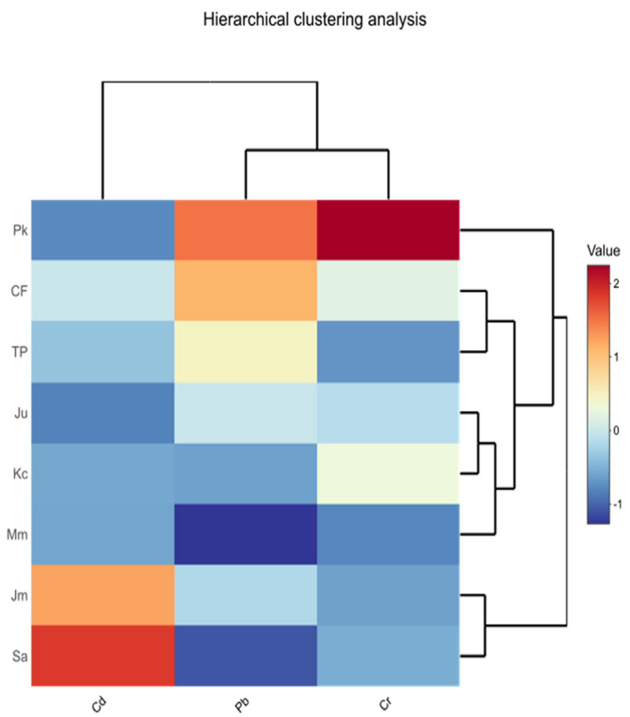

Fig. 6 Target cancer risk associated with heavy metals' concentration in processed fruits.

than unity (> 1.0), particularly HRI of Cd in sauces, jams and canned fruit was alarming. Likewise, THQ and HI levels for Cd, Cr and Pb were relatively high, but in TCR analysis were within the acceptable limit. However, TCR indicates that Cd concentration in sauces, jams and canned fruits may cause cancer to consumers over long-time ingestion.

Declaration of Competing Interest

All authors declared that they have no conflict of interest.

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Authors' contribution

HA did sampling, designed experiment work and prepared initial draft, MHS provided intellectual support and assist in data analysis, MM and ZH offered lab facilitation, WU involve in write up, MSE, JA and MSA, provided financial assistance and help in proof reading of manuscript. AMA supervised project, involved in data analysis and prepared final draft.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.arabjc.2020.10.020.

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