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DIETARY EXPOSURE TO ESSENTIAL AND TOXIC ELEMENTS FROM FRESH, PASTEURISED AND POWDER MILK SAMPLES FROM PAKISTAN

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This study assesses the intake adequacy of elements determined in fresh, ultra high temperature treated and powder milk samples. The samples were analysed by semi-absolute k_0 instrumental neutron activation analysis, epithermal neutron activation analysis and flame atomic absorption spectrophotometry with proper method validation. Fourteen elements (Br, Ca, Cl, Cs, Cu, Fe, K, Mg, Na, P, Rb, Sn, Sr and Zn) were quantified in all samples. All essential elements were assessed for their daily intake by comparing with the dietary reference intakes. The daily intake of Pb, identified in only two samples, was compared with the provisional tolerable weekly intake defined by FAO/WHO. The Sr/Ca ratio was calculated for the assessment of radioactive dose. Additionally, the data was explored by cluster analysis and correlation coefficient to extract relationships between samples and elements respectively.

Keywords: Milk analysis, Mineral contents, Dietary intake, Cluster analysis, Correlation coefficient

1. Introduction

Milk is a very important source of nutrients. It contains minerals, vitamins (A, B1, B2, B6, B12, C, D, E and K), proteins, carbohydrates and fats. The number of substances in milk is more than 100 that are either in solution, suspension or emulsion form. Casein, the major protein of milk remains suspended as colloid, the fat and fat-soluble vitamins in the milk are in the form of emulsion. Lactose (milk sugar), some proteins (whey protein), mineral salts and other substances stay in the water of milk. The casein and fat give milk most of its physical characteristics, taste and flavour to dairy products. Proteins in milk contain all the essential amino acids. Milk also contains less than 1 % (w/w) minerals.

Milk is an excellent source of most minerals essential for the growth of young ones. An element is essential [1] if it is present in healthy tissue, its concentration must be relatively constant between different organisms and its deficiency induces abnormalities in different species and its supplementation with the element removes the abnormalities. There are four non-metallic elements (H, O, C and N) accounting for 99% of all elements in all biological systems. Seven elements Na, K, Ca, Mg, P, S and Cl provide another 0.9% of the total and essential trace elements share the remaining 0.1%.

Trace elements play crucial role in various biochemical functions of the body as some metals form the integral part of enzymes [2]. Sodium, potassium and chloride exist largely in the ionized state in the aqueous part of milk. The other, divalent cations of milk including calcium, magnesium and zinc have measurable concentrations of free ion and are part of the complex electrochemical equilibrium. Zinc is an essential component of over 200 enzymes, which may play both a catalytic and structural role. Depending upon the degree of zinc depletion, deficiency in the young may cause growth delay [3]. Iron act as oxygen carrier in the heme respiratory pigments, it is active in a variety of metalloenzymes involved in redox reactions with oxygen. Copper is a constituent of many metalloenzymes, which function to transport copper to the tissues and to release iron from stores and enzymes involved in the synthesis of connective tissues, melanin and catecholamines [3]. The symptoms of copper deficiency in the young include anemia, unresponsiveness to iron supplementation, defects in bone, cartilage and pigmentation, and diarrhea [4]. Manganese metalloenzymes have a wide range

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of metabolic functions. Fetal life and early infancy are the periods most vulnerable to manganese deficiency. Strontium helps in building matrix of bones and teeth.

Elemental analysis of milk is important for the assessment of the quality of milk as well as to determine any possible contamination through adulteration or by environment. Milk is adulterated by adding additional water, detergents, disinfectants, antibiotics, pesticides, insecticides and others. Although body requirement of mineral content is very low even then their excess or deficiency may disturb the normal biochemical functions. Many research studies indicated the presence of heavy metals like Pb and Cd in milk and its products [5]. Pakistan, with current estimates, is the 4th largest milk producing country in the world with 33 billion liters of milk produced annually. The main aim of this study is to analyse different types of milk available in market and dairy forms for their adequacy based on their mineral contents and to classify samples using statistical analysis.

2. Methods

In this study three techniques, k_0-Instrumental Neutron Activation Analysis (k_0-INAA), Epithermal Neutron Activation Analysis (ENAA) and Atomic Absorption Spectrophotometry (AAS) were employed. ENAA was required for iodine determination, AAS was inevitable for the determination of Cd, Cu, Ni and Pb. Rest of the elements was quantified by k_0-INAA. Each technique is described briefly, in next sections.

2.1. k_0 Instrumental Neutron Activation Analysis (INAA) and Epithermal Neutron Activation Analysis (ENAA)

Application of k_0-INAA requires the determination of thermal to epithermal neutron flux ratio (f) and epithermal neutron flux shape factor (α) and full energy peak efficiency (ε_p) calibration of the detector for the counting geometry. In our calculations f, α and (ε_p) were determined experimentally, while the factors $Q_0(\alpha)$ and $k_{0,Au}(a)$ were taken from published data [6].

In this study, phosphorus was quantified using $^{31}P(n,\alpha)^{28}Al$ reaction with the absolute standardization and fast neutron flux. The latter was measured by using $^{58}Fe(n,\gamma)^{59}Fe$ and $^{54}Fe(n,p)^{54}Mn$ reactions.

The application of ENAA requires the sample packed inside a Cd sheet and sent for irradiation. This increases the detection sensitivities of the isotopes having higher Q_0 value. In our study, we applied ENAA for the determination of iodine in milk samples. The quantification was performed by relative calibration, which involves irradiating and comparing peak areas of sample with a standard of known concentration.

2.2. Atomic Absorption Spectrophotometry (AAS)

In AAS, classical calibration was performed for quantification whereby the calibration points were fitted with a straight line having two coefficients, i.e. slope and intercept. Standard solutions were prepared by appropriate dilution of the element standards. A minimum of three values were recorded for each concentration and each calibration curve was constructed starting with a blank solution.

3. Experimental

3.1. Sampling and Sample pre-treatment

Thirteen samples of milk were included in this study. The samples were given codes from A to M consecutively. A list of the codes and sample descriptions is given in Table 1. The list comprises three types of samples: fresh liquid milk, UHT liquid milk and powder milk. UHT and powder samples were purchased from market. Fresh milk samples were taken from different dairy farms. Initially, the liquid samples were collected in polyethylene containers but soon after collection, they were freeze dried and stored in a refrigerator. All liquid samples were freeze dried in a Beta-A Christ freeze dryer for a period of about 72 hours. The dried samples were pulverized in a grinder and stored in clean air-tight polyethylene bottles as powders.

3.2. Instrumentation

3.2.1. k_0-NAA and ENAA

The f and α were determined using Al-0.1% Au wire (IRMM-530RC, Belgium, Geel) and ZrO_2 (99.99%, Aldrich, Wisconsin) powder. Full peak efficiency calibration of the detector was performed for different detector to source geometries using ^{241}Am, ^{133}Ba, ^{137}Cs, ^{60}Co and ^{152}Eu point calibration sources. Peak-to-total calibration was performed by ^{203}Hg, ^{65}Zn, ^{198}Au, ^{51}Cr and ^{137}Cs point sources.

Table 1. Sample descriptions with codes and comments.

Sample Code	Sample form	Comment
Milk-A	Fresh	Direct squirted from cow
Milk-B	Fresh	Supplied at home1
Milk-C	Fresh	Supplied at home2
Milk-D	UHT	Purchased from market
Milk-E	UHT	Purchased from market
Milk-F	UHT	Purchased from market
Milk-G	UHT	Purchased from market
Milk-H	UHT	Purchased from market
Milk-I	UHT	Purchased from market
Milk-J	UHT	Purchased from market; Fat free (skims)
Milk-K	UHT	Purchased from market, Fe-fortified
Milk-L	Powder	Purchased from market, Fe-fortified
Milk-M	Powder	Purchased from market

Each sample weighing upto 150 mg was packed alongwith the Au/Zr monitors in a polyethylene rabbit for irradiation, which was performed at Pakistan Atomic Research Reactor-1 (PARR-1) and PARR-2. Former is a 10 MW research reactor with nominal thermal neutron flux of about 2×10^{13} cm^{-2} s^{-1} and later is a 30 kW Miniature Neutron Source (MNSR) having nominal thermal neutron flux of 1×10^{12} cm^{-2} s^{-1}. In the present study, two irradiation channels, RS1 and RS3, of PARR-1 and two irradiation channels, B2 and B3, of PARR-2 were used. After irradiation the samples were shifted to pre-weighed clean polyethylene capsules for counting.

The gamma-ray spectra were acquired using a p-type coaxial HPGe detector (Eurisys Measures, France) coupled through a 570 ORTEC made spectroscopy amplifier to Trump PCI, 8k ADC/MCA card with Gamma Vision-32 ver. 6 Software (ORTEC, USA). The detector has 60% relative efficiency and FWHM of 1.95 keV at 1332 keV gamma peak of ^{60}Co.

3.2.2. AAS

All the quantification was carried out using Hitachi model Z-2000 Polarized Zeeman Atomic Absorption Spectrophotometer (AAS) coupled with software based data handling facility. A water cooled, premix, fish-tail type burner, having a slot of 10×0.05 cm, was used for the air-acetylene flame. Hollow cathode lamps from Hitachi were used as radiation source.

3.2.2.1. Reagents and Glassware

All the glassware was cleaned by overnight soaking in HNO_3 (E. Merck, Darmstadt) (1:1), followed by repeated rinsing with distilled water. Distilled and deionized water was used throughout this work. Concentrated HNO_3 (E. Merck) / $HClO_4$ (E. Merck) were used for digestion of milk samples. For the preparation of calibration curve, fresh working standards were made by appropriate dilution of stock solution of 1000 mg g^{-1} of Cu, Ni, Pb and Cd in distilled water immediately before use.

3.2.2.2. Dissolution Procedure

Complete mineralization of the sample is the fundamental requirement for accurate spectrometric analysis. Mixture of HNO_3 and $HClO_4$ was found to be the most suitable for the wet digestion of milk samples prior to AAS measurements. About 0.5 g of milk powder was taken in 100 mL digestion flasks fitted with 25 cm long air condenser, 5 mL conc. HNO_3 was added to the sample. The contents were heated at 80 ^{o}C for 80 minute with occasional shaking. After cooling 1.5 mL of concentrated perchloric acid (70%) was added and heated again at 250 ^{o}C with occasional shaking till white fumes evolve. The clear solution obtained was cooled and transferred to a 10 mL measuring flask and the volume was made up with deionized water, for subsequent measurements of desired metals. A blank was prepared under similar conditions. Certified reference material (IAEA-V-10) analysed for the study were also digested using the same procedure [7].

3.3. Data Analysis

In NAA, the data reduction steps after spectra acquisition were performed by our program *GammaLab* [8], which performs k_0-based quantifications with uncertainties after proper nuclide identification. Cluster analysis was done by MINITAB release 13.1 (MINITAB Inc. USA), Correlation coefficient analysis was performed in MATLAB release 2006b (Mathworks, Inc. USA) and rest of the calculations were done in Microsoft EXCEL release 2007 (Microsoft Corp. USA).

Table 2. Average flux characteristics at four irradiation channels of two reactors.

Reactor Name	Irradiation Channel	f	α	ϕ_{Th} (cm^{-2} s^{-1})
PARR-I	RS1	33.18±5.11	0.0427±0.0292	$2.43\times10^{13}\pm2.10\times10^{12}$
PARR-I	RS3	41.37±8.86	-0.0420±0.0042	$2.39\times10^{13}\pm1.30\times10^{12}$
PARR-II	B2	19.81±0.44	-0.0332±0.0051	$1.04\times10^{12}\pm1.66\times10^{10}$
PARR-II	B3	18.76±0.24	-0.0258±0.0121	$1.02\times10^{12}\pm6.62\times10^{10}$

4. Results and Discussion

The average values of f and α determined for PARR-1 and PARR-2, for the irradiation channels utilised in this study are presented in Table 2. Irradiations at PARR-1 were performed for elements found in lower concentration and form long-lived radionuclides, these include Co, Cr, Cs, Fe, Hf, Rb, Sc, Sr and Zn. PARR-2 was used for elements usually found in higher concentrations and form intermediate- or short-lived radionuclides, these include Br, Ca, Cl, K, I, Mg, Na and P. The analysis scheme for NAA can be found in our previous work [9, 10]. NAA and AAS methodologies were validated by analysing three reference materials: IAEA-336 (lichen), NIST-SRM-1572 (citrus leaves) and IAEA-V-10 (hay powder). The results of 10 elements (Br, Co, Cr, Cs, Fe, K, Na, Rb, Sc and Zn) determined in IAEA-336 by k_0-INAA, iodine in NIST-SRM-1572 (citrus leaves) by ENAA and 4 elements (Cd, Cu, Ni and Pb) by AAS are presented in Figure 1 with 95% confidence intervals. It is clear that the determined concentration of all elements except Rb were within the 95% confidence interval.

Specific gravity of bovine milk varies from 1.023 to 1.040 with an average of 1.030 [11], in our data it ranged from 1.010 (milk-I, UHT) to 1.074 (milk-A, fresh) with an average of 1.026. The amount of water in our fresh and UHT samples ranged from 81.0% (milk-A, fresh) to 90.3% (milk-J, skims) with an average of 87%. The average elemental concentrations of three types of milk samples (fresh, UHT and powder) are presented in Table 3. There are two powder samples in this study; their elemental concentrations were converted from (w/w) to (w/v) by adding 26g of powder milk into 200 mL of water, as suggested by the producer. The elemental analysis yielded quantitative results for 22 elements (Br, Ca, Cl, Co, Cr, Cs, Cu, Fe, Hf, I, K, Mg, Mn, Na, Ni, P, Pb, Rb, Sc, Sn, Sr, and Zn). Among all elements, 14 elements (Br, Ca, Cl, Cs, Cu, Fe, K, Mg, Na, P, Rb, Sn, Sr and Zn) were present in all samples. Ni was determined in eleven samples, Co, Mn and Sc in seven samples, iodine in five samples and Cr, Hf and Pb in two samples. Cd was not identified in any sample by AAS, having LoD as 0.01 μg mL^{-1}. Table 3 shows that maximum number of elements was determined in UHT type. The presence of Mn, Cr, Pb and slightly higher concentration of Fe, in UHT samples, indicates a source of steel and other alloy. This contamination might be from milk processing vessels used in the industry or containers used for transporting milk form dairy farms to the industry. The order of decreasing elemental concentration in fresh and powder samples is: Ca > K > Cl > Na > P > Mg > Sn > Zn > Sr > Br. The order in UHT is slightly different, it is: K > Ca > Na > Cl > P > Mg > Sn > Zn > Sr > Br.

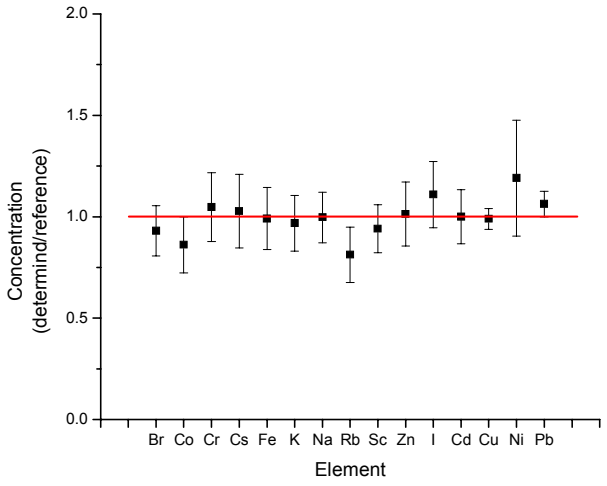

Figure 1. Concentration (determined/reference) in different reference materials by k_0-INAA (Br, Co, Cr, Cs, Fe, K, Na, Rb, Sc, Zn), ENAA (I) and AAS (Cd, Cu, Ni and Pb) for quality control.

Table 3. Average concentrations of elements determined in different types of milk samples.

Component	Fresh Milk	UHT Milk	Powder Milk
Ca (g/mL)	1.78±0.11	1.00±0.02	1.25±0.01
K (g/mL)	1.20±0.13	1.14±0.22	1.11±0.03
Na (g/mL)	0.48±0.07	0.97±0.31	0.67±0.09
Cl (g/mL)	0.74±0.11	0.81±0.17	0.70±0.17
P (g/mL)	0.38±0.14	0.20±0.07	0.23±0.03
Mg (g/mL)	0.14±0.03	0.11±0.03	0.10±0.02
Sn (μg/mL)	11.0±4.8	23.8±12.7	57.7±10.4
Zn (μg/mL)	6.04±0.92	3.24±0.61	5.31±1.53
Fe[a] (μg/mL)	0.95±0.05	1.59±0.74	1.29
Sr (μg/mL)	2.88±1.36	2.31±0.5	2.73±0.32
I[b] (μg/mL)	0.10	0.04±0.06	0.09
Br (μg/mL)	2.76±1.59	1.87±0.25	1.91±1.1
Rb (μg/mL)	1.10±0.40	0.79±0.16	0.75±0.31
Ni (μg/mL)	0.18±0.03	0.20±0.13	0.14±0.08
Cu (μg/mL)	0.13±0.02	0.12±0.06	0.24±0.04
Pb (ng/mL)	-	122±25	-
Mn (ng/mL)	-	124±37	-
Cr (ng/mL)	-	21±11	-
Hf (ng/mL)	-	5.6±1.8	-
Cs (ng/mL)	2.9±1.5	3.2±1.2	2.3±1.2
Co (ng/mL)	-	0.2±0.1	-

[a]: Fe-fortified samples are excluded.

[b]: One sample with high concentration of I was excluded.

Jenness [12] showed that the concentrations of phosphate and calcium are generally proportional to the concentration of casein in fresh milk. We used his estimate of slope and intercept of linear regression between the concentration of Ca and casein and calculated casein in our three fresh milk samples as: milk-A: 23.9±0.7 g/100 mL, milk-B: 27.5±1.2 g/100mL and milk-C: 32.0±1.6 g/100 mL.

4.1. Daily Intake

Since diet has prime importance in our life, health authorities and nutritionists have issued recommendations and guidelines on intake of nutrients and other food components. In 1941, the Food and Nutrition Board (FNB) of the US National Research Council (NRC) published the Recommended Dietary Allowances (RDAs). Later in 1989, RDAs [13] were revised to new Dietary Reference Intake (DRI) [1] for US and Canadian population. For certain elements whose DRIs are not available, FNB provided “adequate intake” (AI) values. To estimate the percent DRIs obtained from milk, we assumed a daily intake of 250 mL per person and calculated daily intake using average elemental concentrations in different types of milk after excluding two iron fortified samples (milk-K and milk-L) and one sample (milk-I) of unusual high concentration of iodine. The results are summarized in Table 4 after averaging concentrations over different types of milk, for those elements having RDAs available [1, 13]. The daily intakes were compared with the DRIs defined for male of age group 31-50 years. This comparison reveals that fresh milk is best in providing most of the essential elements (Ca, I, P,

Zn, Mg). UHT and powder samples are slightly better in Cu and Fe. It can be said that fresh milk is good source of Ca, I, P and Zn but poor source of Fe. World Health Organization (WHO) has given a provisional tolerable weekly intake (PTWI) for Pb as 25 μg/(kg body weight) [14]. Using this limit, two milk samples containing Pb (milk-E and milk-F, both UHT) were evaluated for a person drinking 250 ml of milk/day and weighing 70 kg. It produced weekly intake of 3.1 μg, a value well below the PTWI.

Table 4. % RDA provided by different types of milk for different elements calculated for male of 31-50 year.

Element	RDA/DRI/ AI	% RDA by milk		
		Fresh Milk	UHT Milk	Powder Milk
Fe***	8 mg	2.9	5.0	4.0
Cu	0.9 mg	3.7	3.4	6.7
K*	4.7 g	6.3	6.0	5.9
Na*	1.5 g	8.1	16.1	11.1
Cl*	2.3 g	8.1	8.8	7.6
Mg	420 mg	8.5	6.6	6.3
Zn	11 mg	13.7	7.4	12.1
P	700 mg	13.7	7.2	8.1
I**	150 μg	16.2	6.9	14.7
Ca*	1000 mg	44.4	25.1	31.3

* Adequate Intake
** Excluding results of milk-I having unusual high value.
*** Excluding results of Fe-fortified samples.

4.2. Sr/Ca ratio and K/Na ratio

There are many radioactive elements, which exist in our environment such as ^{60}Co, ^{137}Cs, ^{131}I, ^{40}K, ^{90}Sr, and progenies of ^{238}U and ^{232}Th. A radionuclide, having similar physical and chemical properties to another non-radioactive element belonging to a living body, may get absorbed in our body through food chain. For instance, an inadequate amount of Ca may lead to the absorption of ^{90}Sr, ^{140}Ba and ^{226}Ra. Similarly, K can be replaced by ^{137}Cs. ^{90}Sr (half-life 28 years) is present in our environment form the past nuclear weapon tests and accidents. It is a pure beta emitter and may be more harmful if it enters into living body. To minimize the absorption of ^{90}Sr, International Committee of Radiological Protection (ICRP) [15] recommends a Sr/Ca ratio in food as 1.7×10^{-3} or low. The Sr/Ca ratios in our samples revealed that only milk-A (fresh) and milk-B (fresh) had lower than the ICRP recommended value. The rest of samples had higher ratio, with an average of 2.3×10^{-3} for UHT and powder milks.

The ratio of K/Na in milk is also considered an important indicator of quality. Milk is considered good if it has a higher concentration of K than that of Na. A high K/Na ratio is required for the maintenance of the electrical gradient of milk [16]. The K/Na ratio calculated for our fresh milk samples show an average value of 2.5, which seems a reasonable good concentration ratio.

4.3. Cluster Analysis (CA)

Cluster analysis [17] was applied using Euclidean distance as a measure of similarity with average linkage. The data matrix contained 13 samples and 13 elements (Br, Ca, Cl, Cs, Cu, Fe, K, Mg, Na, P, Rb, Sr and Zn). CA was applied on raw dataset with results presented in Figure 2. It showed milk samples distributed among three clusters:

Cluster-1: Milk-A, B, C (All fresh milk samples)

Cluster-2: Milk-D, E, F, G, H, J, L and M

Cluster-3: Milk-I, K

These clusters show that fresh milk samples can be distinguished from rest of the samples. The classification here is due to major elements (Ca, K, Na, Cl, P and Mg).

When CA was applied using standardized data (z-standardization), the distance based dendrogram is presented in Figure 3, which shows some distinction between the fresh milk samples and UHT samples, however milk-C is not properly classified. The three main clusters created are:

Cluster-1: Milk-A, B (Both fresh milk samples)

Cluster-2: Milk-D, E, F, G, H, J and M

Cluster-3: Milk-I, K

Figures 2 and 3 show more or less similar classification.

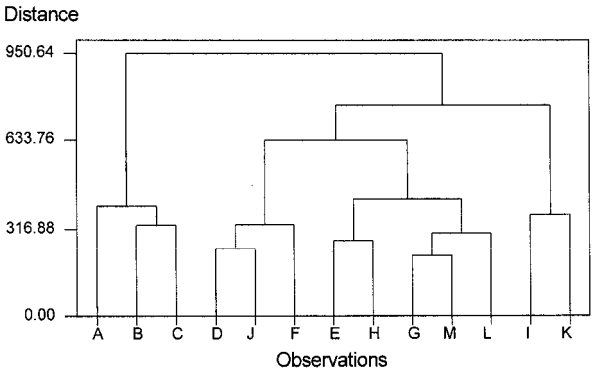

Figure 2. Dendrogram using raw data with Euclidean distance and average single.

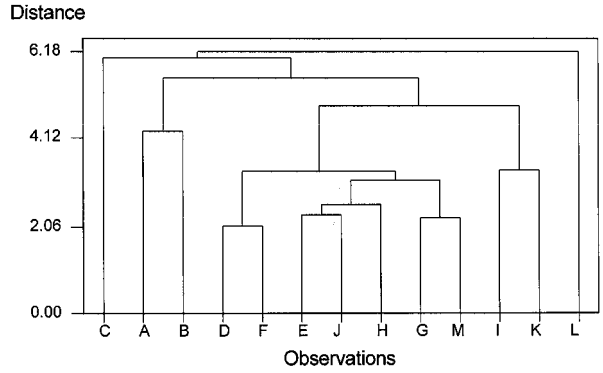

Figure 3. Dendrogram using standardized data with Euclidean distance and average single.

4.4. Correlation Coefficient (CC)

Inter-element relationship was extracted by using correlation coefficient. Correlation coefficients were calculated between elemental concentrations of UHT samples. The following pairs were found having significant ($p \geq 0.05$) correlations.

Ca-Cs (0.769), Ca-Sn (0.709), Ca-Sr (0.786), Ca-Zn (0.879), Cl-Na (0.840), Cs-Cu (0.727), Cs-Rb (0.830), Cs-Sn (0.719), Cu-Rb (0.822), Cu-Sn (0.732), Mg-Sr (0.813), Rb-Sn (0.904).

5. Conclusions

The study revealed that fresh milk was free from heavy metals. It provides Ca as much as 45% of DRI. Fresh milk has a Sr/Ca ratio below than the ICRP limit. The UHT and powder milk samples had Sr/Ca ratio 35% higher. Among toxic elements, only Pb was determined in two UHT samples with concentrations well below the PTWI limit defined by WHO. Similarly, K/Na ratio was almost 2.5 for fresh milk samples indicating good quality of milk. An analysis based on CA indicated the distinction possibility between fresh milk and others. The overall analysis reflects positively on the quality of fresh milk produced in Pakistan.

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