

Mineral Profile for Milk Traceability

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Abstract

The purpose of this study is to figure out geographical indication in whole milk powder. Thus, 32 whole milk powder samples labeled as originating in New Zealand, Australia, the Netherlands and the Czech Republic were analyzed. The amounts of Ag, Al, As, Ba, Cd, Co, Cr, Cs, Cu, Fe, Li, Mn, Ni, Pb, Rb, Se, Sr, V and Zn were determined by an inductively coupled plasma-mass spectrometer (ICP-MS). The data obtained was analyzed by principal component analysis (PCA). The results revealed that New Zealand milk samples could be differentiated from Australia and the Netherlands milk samples by the amounts of Al, As, Ba, Cs, Fe, and Pb. However, there was a limit of using mineral profiles to distinguish the milk samples of New Zealand and Czech Republic.

Keywords: Whole milk powder, Traceability, Dry-ashing digestion, Microwave-assisted acid digestion, Inductively coupled plasma-mass spectrometry, Principal component analysis

Introduction

Milk is a unique fluid produced by female mammals. It is usually considered as a sole source of food for newborn offspring because it contains most of nutrients required for growing. Even though milk is generated from the same species, its composition is varied. In terms of minerals, the previous study reported that the condition of a mammary gland and extrinsic conditions were two main factors of the milk variation. The former factor comprises the lactation stage, calf birth weight and health status, while the latter factor comprises season, diet, geological origin, contamination, and production system (Kondyli *et al.* 2007; Vergara *et al.* 2003; Hamann and Krömker 1997; Pennington 1990; Herwig *et al.* 2011; Patra *et al.* 2008; do Nascimento *et al.* 2010). The EU Regulation No. 178/2002 stated that any food, feed, food-producing animal or substance intended to be needs to be performed traceability in all stages. This requirement can ensure about food safety, fair trading system, and the reliability in such a product. Moreover, traceability also applies in

Free Trade Agreement (FTA). The customs officer in the importing country has to carry traceability in the goods imported whether they are produced or obtained in the importing country or not before he abolishes tariff in such products.

Because one of the factors influencing the milk composition is geological origin, variation in minerals can be used as criteria in traceability. Santos *et al.* (2012) reported that the concentration of Al, Br, Ca, Cr, Cu, K, Mn, Ni, Rb and Sr demonstrates the potential to differentiate the regional origin of milk from two dairy farms located in Brazil. In their study the researcher team applied the high performance ion chromatography (HPIC), inductively coupled plasma atomic emission spectroscopy (ICP-AES), nuclear magnetic resonance (NMR) and isotope ratio mass spectrometry (IRMS), and Principle Component Analysis (PCA). Moreover, Bilandžić *et al.* (2011) reported that the concentration of Cd and Hg provides the capacity of determining the region of milk production in Northern and Southern region of Croatia. They conducted their research by using graphite furnace-atomic absorption spectroscopy. The authors suggested that the variation of Cd depended on type of food i.e. feeding crops, trace element premixes, fish meal and minerals, while the variation of Hg is based on the place animal rears. However, the previous studies focus on the differentiation on the region within a country.

Objective

This study proposed to investigate the possibility to distinguish the origin among countries by using mineral profiling in order to apply this approach for distinguishing in milk based on its country of origin.

Research Methodology

Materials

Sample

Thirty two samples of whole milk powder labeled as originating in New Zealand (n=10), Australia (n=10), the Netherland (n=8), and the Czech Republic (n=4) were analyzed. They were obtained from the ports of entry of the The Customs Department of Thailand and from bakery-supplied shops located in Bangkok, Nonthaburi, and Nakhon Pathom during December 2010 to May 2011.

Reagent

65% nitric acid was from Merck (Darmstadt, Germany); Multi-element standard of Al, As, Ba, Br, Cd, Cs, Cr, Co, Cu, Fe, Pb, Li, Mn, Mo, Ni, Rb, Se, Ag, Sr, V and Zn (10 µg/mL) in 5% nitric acid, RhO (1,000 µg/mL) in 10% HCl and ScO (1,000 µg/mL) in 2% HNO₃ were supplied by Accutrace, Inc. (PA, USA), and deionized and free organic water (ASTM type 1) was prepared by Ultreapure water system (Barnstead D9015-33, Barnstead International, USA) with a resistivity of 18.2 MΩ cm⁻¹.

Instrument and quality control of measurement

Inductively coupled plasma-mass spectrometer (ICP-MS) (SCIEX ELAN DRC-e, Perkin Elmer, USA) was used for mineral analysis. The condition of the instrument was shown in Table 2. The instrumental limit of detection (*LOD*) was determined based on IUPAC recommendation, calculated from $LOD = (3s)/S$, where *s* was the standard deviation of blank measurement and *S* was the slope of three standard concentrations determining in five replicates. The repeatability of a method was reported in the Horwitz ratio, which is also called Horrat value. It is the ratio of relative standard deviation (*RSD*) of three concentrations with five replicates, to the corresponding predicted relative standard deviation (*PRSD*). *PRSD* is calculated by

using Horwitz equation where $PRSD_R = 2^{(1-0.5\log C)}$. Here, C is concentration of analyte in the standard as a decimal fraction (g/g). The acceptable Horwitz ratio is 0.5 – 0.7 (Horwitz 2006).

Methods

Sample treatment

Dry-ashing digestion Whole milk powder sample (0.5 g) was accurately weighed in a crucible and charred on a hot plate at 150° C. The charred sample was moved into a furnace for ashing at 550° C for four hours. The white ash obtained was dissolved with HNO₃ and adjusted the volume to 25 mL by deionized water. In case, the ash was not completely ashed, 1 mL conc. HNO₃ was added and repeated ashing steps.

Microwave-assisted acid digestion Sample (0.5 g) was accurately weighed in the digestion vessel and 2 mL of concentrated HNO₃ and 10 mL deionized water were added. Closed vessels were placed inside a microwave oven (Microwave Activated Reaction System Mar X, CEM Corporation, USA) and decomposition was carried out according to Table 1. After digested, the sample was adjusted the volume to 25 mL by deionized water.

All samples were prepared in triplicate. The final concentration of 10 mgL⁻¹ Sc and Rh were added as internal standards. All diluted sample solutions were filtered and kept at 4° C until performing mineral analysis.

Statistical analyses

The data obtained was performed the Descriptive and Principal Component Analyses (PCA) by using the software XLSTAT version 2012.4.02 (ADDINSOFTTM 1995-2012). PCA was applied to identify patterns in data with a large number of variables.

Results/ Discussion

Sample pretreatment

Two sample treatment techniques including dry-ashing digestion and microwave-assisted acid digestion were compared for averages of measured values, precision, and recovery percentage (Table 3). In terms of precision, microwave-assisted acid digestion gave precision better than dry-ashing digestion because the samples digested by microwave digestion provided lower *RSD*. Moreover, the replicates results of Al, Fe, Li, and Ni in the samples treated by dry-ashing digestion was unacceptable because their values were higher than *PRSD*. For recovery percentage, the samples treated by dry-ashing digestion gave unacceptable recovery percentage in seven minerals *i.e.* Al, Cs, Fe, Ni, Se, Sr, and Zn. In contrast, the sample treated by microwave-assisted digestion provided acceptable recovery percentage in all minerals. These ranges of recovery percent were set according to AOAC Requirements for Single Laboratory Validation of Chemical Methods. Method verification concluded that microwave-assisted digestion was suitable for whole-milk-powder digestion. This conclusion was in agreed with Herwig *et al.* (2011) and Soylak *et al.* (2004).

Quantitation

Table 4 revealed that all minerals except Li showed their amounts higher than *LOD*. Hence, all minerals except Li were performed PCA. Theoretically, PCA uses two mathematical concepts – Statistics and Matrix Algebra. The concepts of standard deviation, variance, and co-variance in Statistics are applied to observe distribution measurements and spread of data. Furthermore, the concepts of eigenvector and eigenvalue in Matrix Algebra are used in analysing and rearranging

PCA results. Descriptive statistics in Table 4 also demonstrated that some minerals *e.g.* Ag or Cd could not distinguish the difference in the country of origin for milk samples because their mineral amounts observed in milk from different countries were closed to each other. Ag, for example, showed its amount ranged from 0.035 – 0.041 $\mu\text{g/g}$, 0.034 – 0.038 $\mu\text{g/g}$, and 0.035 – 0.041 $\mu\text{g/g}$ for samples from New Zealand, Australia, the Netherlands, and the Czech Republic, respectively. However, some minerals showed potential to differentiate the country of origin. For instance, Ba showed its amounts ranged from 0.368 – 0.735 $\mu\text{g/g}$, 0.279 – 0.508 $\mu\text{g/g}$, 0.305 – 0.408 $\mu\text{g/g}$, and 0.406 – 0.562 $\mu\text{g/g}$ for samples from New Zealand, Australia, the Netherlands, and the Czech Republic, respectively. It meant that by using Ba, New Zealand samples could be differentiated from Australia and the Netherlands samples, but they could be distinguished from the Czech Republic samples with limitation.

Principal component analysis

In this study, PCA was applied with determinations of 18 minerals in each milk sample. The Figure 1 showed the trends of mineral profiles in a two-dimension graph. The results in Figure 1 showed that New Zealand milk could be differentiated from Australia and the Netherlands milks by PC2, which was 22.92% of the total variance. On this PC New Zealand milk showed high scores, while Australia and the Netherlands showed low scores. The highest loading value on PC2 included Al, As, Ba, Fe and Pb, and all of them were positive. However, PC3, which showed 16.42 % of the total variance, showed the possibility to discriminate New Zealand milk from the Czech Republic milk because the trend of mineral profiles in New Zealand milk showed higher scores when comparing with the trend in the Czech Republic milk. The highest values on PC3 consisted of Ag, Co, Cr, Ni, Rb, and V, which were all positive.

The results showing that New Zealand samples could be differentiated from Australia and the Netherlands samples were in agreement with the study of dos Santos *et al.* and Bilandžić *et al.* The authors concluded that by using the difference of mineral amounts the area of origin of milk could be determined (dos Santos *et al.* 2012; Bilandžić *et al.* 2011). However, the results shown the limited discrimination in New Zealand samples from the Czech Republic samples supported the conclusion in Sola-Larranaga nad Navarri-Blasco's study. The authors suggests that not only feeding plays a role in the variation of mineral content in milk, but other factors such as nutrition additives and supplementary contamination, or stress factors have an impact as well (Sola-Larranaga and Navarro-Blasco 2009) Furthermore, numerous study works revealed the variability in milk occurs in many factors. Therefore, in order to avoid variability, the sample size from each country should be increased.

However, regarding the diagram, there were two outliers from Australia and the Netherlands sample groups. This phenomenon could describe by using descriptive statistical data. It showed that two outliers show higher amount in Fe and Zn comparing to the sample from the same country.

Conclusions

The results revealed that the set of mineral comprising Al, As, Ba, Cs, Fe, and Pb could distinguish New Zealand whole milk powder samples from Australia and the Netherlands milk samples. These minerals were high in New Zealand milk sample when comparing with Australia and the Netherlands samples. However, there was limitation in the differentiation of New Zealand samples and the Czech Republic samples based on this data set.

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Table**Table 1** Working conditions for microwave-assisted acid digestion

Operating parameter	Setting
Program	Ramp to temperature
Max powers	1200 watts
Pressure	200 psi
Temperature	200°C
Hold time	30 min

Table 2 Working conditions of ICP-MS

Operating parameter	Setting
RF power (W)	1000
Nebulizer (carrier gas) flow rate (L/min)	0.95
Lens voltage (V)	6.00
Analog stage voltage (V)	-2250
Pulse stage voltage (V)	11550
Discriminator threshold (V)	55
Quadrupole rod offset (V)	0
Detector	Dual
Speed of peristaltic pump	24
Sweeps / reading	40
Replicates	3
Dwell time	50 ms
Scan mode	Peak hopping

Table 3 Mean values of mineral amount (dry weight), relative standard deviation (*RSD*, $n=3$), predicted relative standard deviation (*PRSD*) and recovery percentage of each mineral in the samples prepared by different techniques

Minerals	Dry-ashing digestion				Microwave-assisted acid digestion			
	Mean	<i>RSD</i>	<i>PRSD</i>	Recovery	Mean	<i>RSD</i>	<i>PRSD</i>	Recovery
Ag ($\mu\text{g/g}$)	0.055	2.61	49.09	104.56 ^d	0.071	4.55	8.51	97.64 ^d
Al ($\mu\text{g/g}$)	3.470	66.73	26.27	157.17 ^c	0.692	6.01	7.81	102.44 ^d
As ($\mu\text{g/g}$)	0.038	1.56	51.89	127.18 ^d	0.054	6.08	8.59	99.15 ^d
Ba ($\mu\text{g/g}$)	0.259	17.97	38.82	109.39 ^d	0.432	5.89	7.96	97.24 ^d
Cd ($\mu\text{g/g}$)	0.036	5.56	52.36	117.95 ^d	0.045	4.18	8.65	99.80 ^d
Co ($\mu\text{g/g}$)	0.047	13.57	50.18	122.07 ^d	0.096	6.56	8.41	99.57 ^d
Cr ($\mu\text{g/g}$)	0.136	28.36	42.77	121.66 ^d	0.107	5.90	8.38	96.87 ^d
Cs ($\mu\text{g/g}$)	0.165	0.05	41.55	134.70 ^d	0.206	0.11	8.18	101.86 ^d
Cu ($\mu\text{g/g}$)	12.770	20.31	21.59	105.57 ^b	10.465	6.92	6.99	95.80 ^b
Fe ($\mu\text{g/g}$)	3.030	28.54	26.81	139.44 ^c	7.280	6.48	7.10	100.63 ^c
Li ($\mu\text{g/g}$)	22.370	49.13	19.84	88.97 ^b	0.120	7.51	8.34	96.80 ^d
Mn ($\mu\text{g/g}$)	0.131	13.61	43.02	80.91 ^d	0.073	7.88	8.49	96.71 ^d
Ni ($\mu\text{g/g}$)	0.260	51.29	38.80	134.70 ^d	0.104	8.34	8.39	98.05 ^d
Pb ($\mu\text{g/g}$)	0.116	40.83	43.81	123.24 ^d	0.194	6.84	8.20	101.72 ^d
Rb ($\mu\text{g/g}$)	0.087	13.08	45.75	95.41 ^d	0.418	2.28	7.97	99.21 ^d
Se ($\mu\text{g/g}$)	0.025	10.55	55.30	137.84 ^d	0.064	5.25	8.54	102.86 ^d
Sr ($\mu\text{g/g}$)	0.315	12.32	37.69	136.07 ^d	0.363	3.32	8.01	99.56 ^d
V ($\mu\text{g/g}$)	0.033	7.07	52.98	107.63 ^d	0.047	3.74	8.63	103.13 ^d
Zn (mg/g)	20.430	4.62	7.11	84.25 ^a	14.243	4.25	4.79	99.73 ^b

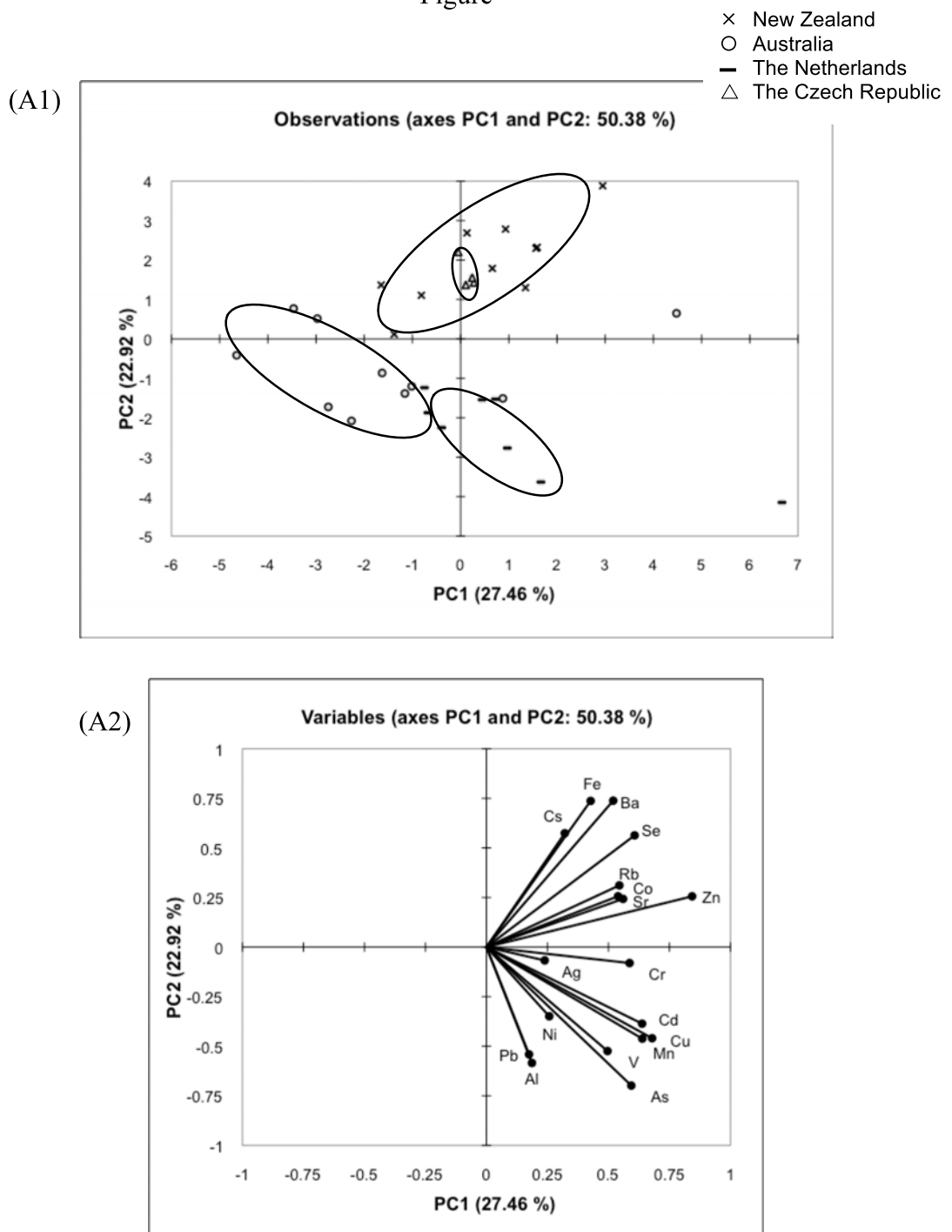
Acceptable recovery based on AOAC requirement for Single Laboratory Validation of Chemical Methods, a, b, c, and d are for 80-110%, 80-115%, 75-120%, and 70-125% respectively.

Table 4 Descriptive statistics of minerals found in the whole milk powder samples from New Zealand (n=10), Australia (n=10), the Netherlands (n=8), and the Czech Republic (n=4) comparing with instrumental LOD

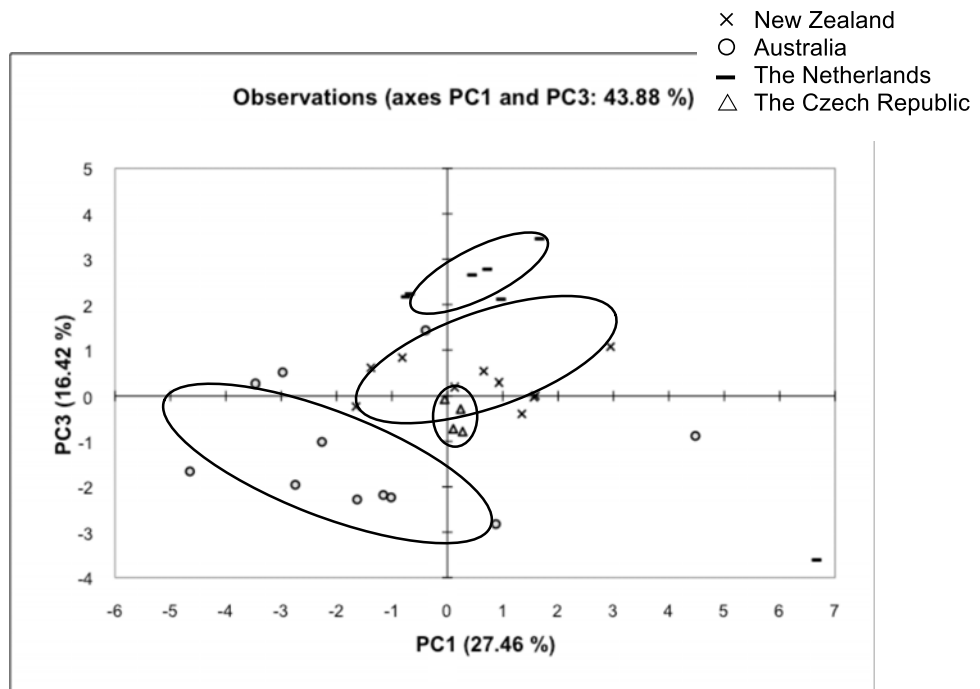
Mineral	LOD	Country	Mean \pm SD	Min	Max
Ag ($\mu\text{g/g}$)	0.48 pg/g	NZ	0.037 \pm 0.002	0.035	0.041
		AU	0.035 \pm 0.001	0.034	0.035
		NL	0.038 \pm 0.002	0.035	0.041
		CZ	0.036 \pm 0.002	0.034	0.039
Al ($\mu\text{g/g}$)	2.27 pg/g	NZ	8.694 \pm 2.942	1.079	11.789
		AU	11.028 \pm 3.280	7.221	16.868
		NL	29.514 \pm 28.033	4.127	83.646
		CZ	3.613 \pm 4.485	0.809	10.298
As ($\mu\text{g/g}$)	3.60 pg/g	NZ	0.052 \pm 0.002	0.049	0.008
		AU	0.054 \pm 0.007	0.044	0.017
		NL	0.068 \pm 0.005	0.057	0.017
		CZ	0.051 \pm 0.001	0.049	0.002
Ba ($\mu\text{g/g}$)	1.85 pg/g	NZ	0.514 \pm 0.132	0.368	0.735
		AU	0.343 \pm 0.064	0.279	0.508
		NL	0.339 \pm 0.037	0.305	0.408
		CZ	0.455 \pm 0.073	0.406	0.562
Cd ($\mu\text{g/g}$)	2.07 pg/g	NZ	0.034 \pm 0.001	0.033	0.735
		AU	0.035 \pm 0.001	0.033	0.508
		NL	0.035 \pm 0.001	0.034	0.408
		CZ	0.035 \pm 0.001	0.034	0.562
Co ($\mu\text{g/g}$)	0.30 pg/g	NZ	0.119 \pm 0.006	0.111	0.128
		AU	0.106 \pm 0.006	0.100	0.122
		NL	0.119 \pm 0.010	0.108	0.137
		CZ	0.115 \pm 0.004	0.111	0.120
Cr ($\mu\text{g/g}$)	0.27 pg/g	NZ	0.947 \pm 0.165	0.733	1.173
		AU	0.919 \pm 0.338	0.389	1.483
		NL	0.799 \pm 0.415	0.545	1.811
		CZ	0.830 \pm 0.167	0.729	1.079
Cs ($\mu\text{g/g}$)	0.23 pg/g	NZ	0.212 \pm 0.067	0.159	0.333
		AU	0.160 \pm 0.006	0.153	0.169
		NL	0.164 \pm 0.005	0.160	0.176
		CZ	0.173 \pm 0.023	0.161	0.208
Cu ($\mu\text{g/g}$)	0.35 pg/g	NZ	0.220 \pm 0.069	0.161	0.384
		AU	0.298 \pm 0.147	0.115	0.463
		NL	0.376 \pm 0.264	0.237	1.023
		CZ	0.287 \pm 0.062	0.229	0.360
Fe ($\mu\text{g/g}$)	7.69 pg/g	NZ	78.068 \pm 24.778	49.490	115.649
		AU	25.421 \pm 15.657	3.269	49.153
		NL	21.648 \pm 11.512	15.209	49.904
		CZ	61.514 \pm 18.966	51.416	89.936
Li ($\mu\text{g/g}$)	83.85 pg/g	NZ	0.045 \pm 0.044	0.019	0.162
		AU	0.028 \pm 0.004	0.023	0.032
		NL	0.084 \pm 0.022	0.035	0.105

Mineral	LOD	Country	Mean \pm SD	Min	Max
		CZ	0.025 \pm 0.004	0.023	0.031
Mn ($\mu\text{g/g}$)	0.80 pg/g	NZ	0.240 \pm 0.027	0.204	0.283
		AU	0.281 \pm 0.102	0.163	0.416
		NL	0.357 \pm 0.260	0.226	0.988
		CZ	0.204 \pm 0.028	0.181	0.245
Ni ($\mu\text{g/g}$)	3.24 pg/g	NZ	0.293 \pm 0.097	0.128	0.461
		AU	0.260 \pm 0.133	0.141	0.579
		NL	0.548 \pm 0.212	0.219	0.872
		CZ	0.346 \pm 0.105	0.200	0.450
Pb ($\mu\text{g/g}$)	1.60 pg/g	NZ	0.061 \pm 0.034	0.045	0.156
		AU	0.059 \pm 0.009	0.050	0.071
		NL	0.083 \pm 0.011	0.065	0.096
		CZ	0.046 \pm 0.003	0.044	0.051
Rb ($\mu\text{g/g}$)	0.25 pg/g	NZ	10.275 \pm 2.590	7.039	14.733
		AU	4.083 \pm 3.923	0.306	11.106
		NL	10.747 \pm 1.691	7.319	12.311
		CZ	7.833 \pm 1.565	5.486	8.668
Se ($\mu\text{g/g}$)	38.70 pg/g	NZ	0.204 \pm 0.030	0.170	0.262
		AU	0.096 \pm 0.357	0.024	0.243
		NL	0.149 \pm 0.017	0.122	0.183
		CZ	0.249 \pm 0.045	0.213	0.308
Sr ($\mu\text{g/g}$)	1.17 pg/g	NZ	1.064 \pm 0.202	0.775	1.338
		AU	1.138 \pm 1.494	0.296	5.329
		NL	0.869 \pm 0.192	0.745	1.302
		CZ	1.255 \pm 0.222	0.981	1.471
V ($\mu\text{g/g}$)	0.32 pg/g	NZ	0.050 \pm 0.003	0.046	0.054
		AU	0.047 \pm 0.005	0.041	0.058
		NL	0.069 \pm 0.007	0.059	0.078
		CZ	0.049 \pm 0.001	0.048	0.049
Zn (mg/g)	158.65 pg/g	NZ	15.240 \pm 3.316	10.705	19.852
		AU	9.306 \pm 9.628	0.895	30.985
		NL	9.818 \pm 9.902	5.172	34.223
		CZ	16.259 \pm 2.199	13.402	18.766

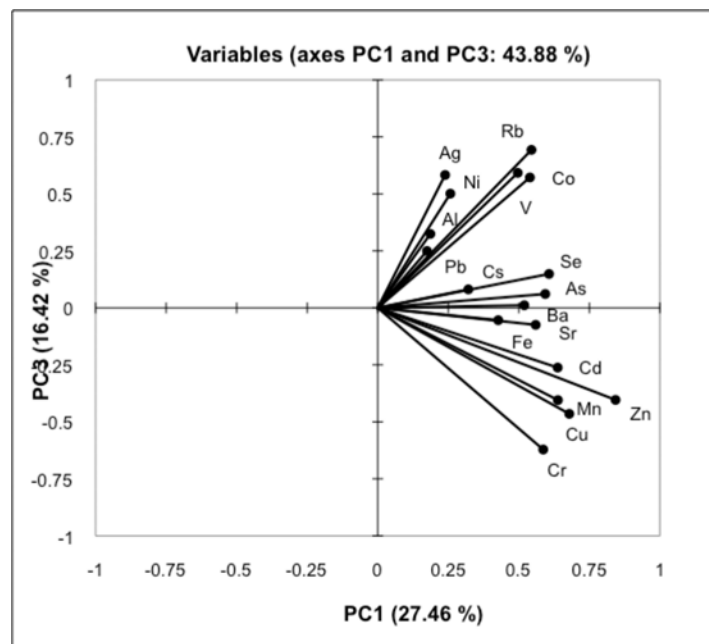
Figure



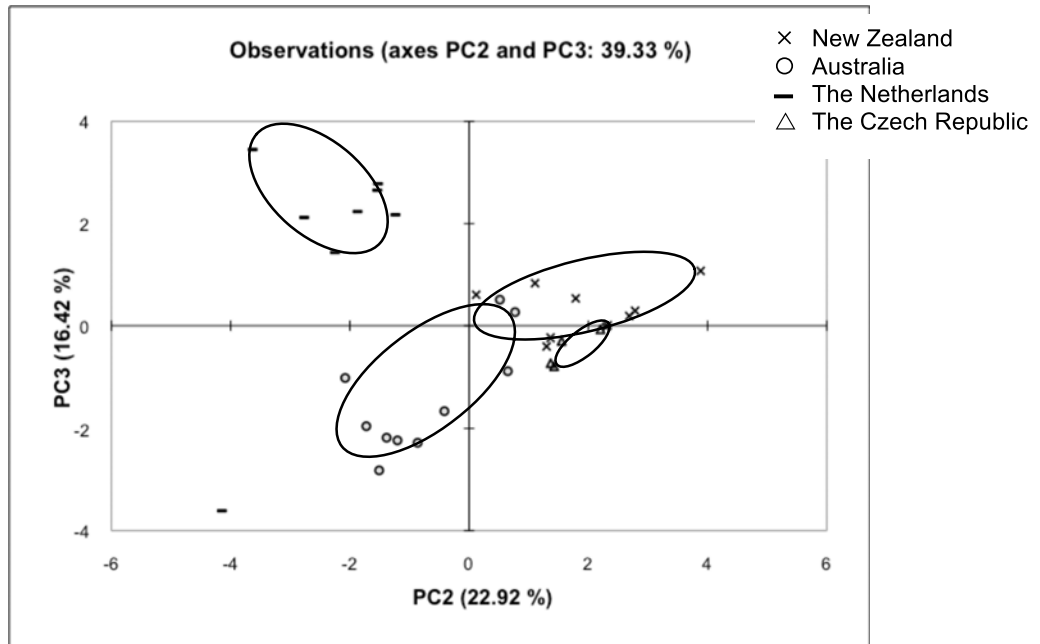
(B1)



(B2)



(C1)



(C2)

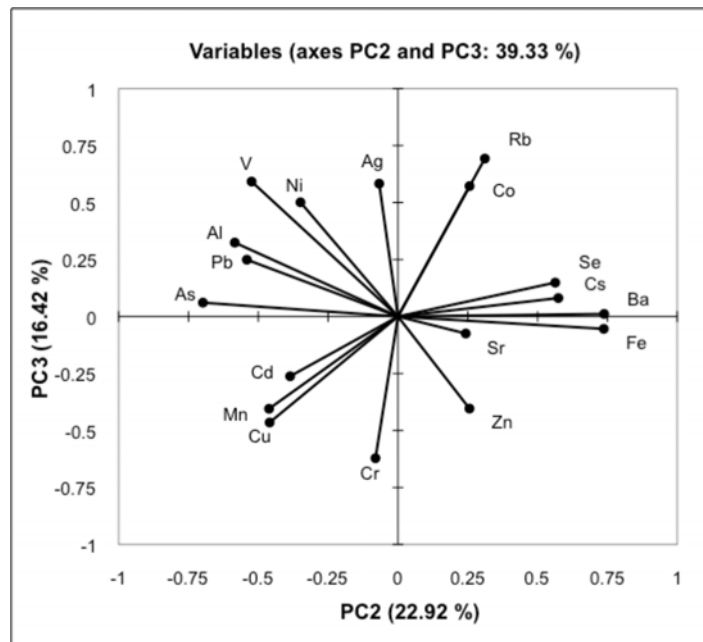


Fig. 1 PCA graph showing correlation between 32 whole milk powder samples and 18 variables (Ag, Al, As, Ba, Cd, Co, Cr, Cs, Cu, Fe, Mn, Ni, Pb, Rb, Se, Sr, V and Zn)