

Variation in macrominerals and trace elements in cows' retail milk and implications for consumers nutrition

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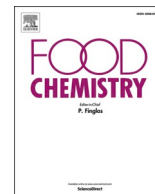
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Variation in macrominerals and trace elements in cows' retail milk and implications for consumers nutrition

Eric E. Newton^a, Ásta H. Pétursdóttir^b, Stephane Beauclercq^a, James Clarke^a, Natasa Desnica^b, Sokratis Stergiadis^{a,*}

^a University of Reading, School of Agriculture, Policy and Development, Department of Animal Sciences, PO Box 237, Earley Gate, Reading RG6 6EU, United Kingdom

^b Matís, Vínlandsleið 12, Reykjavík 113, Iceland

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ABSTRACT

Based on previous farm-level studies, this study hypothesised that production system (conventional, CON; organic, ORG; channel island, CHA) and season would cause variation in the concentrations of macrominerals and trace elements in retail milk. On average, milk retained its status as an excellent source of Ca, P, I, and Mo across different demographics, and a very good source of K, Mg, and Zn for children. Compared with CON and ORG, CHA milk contained higher concentrations of Ca, Mg, P, Cu, Mn, and Zn; and lower concentrations of K and I. Macrominerals did not show a clear seasonal pattern but trace elements were all at lower concentrations during the typical grazing season. Variation in mineral concentrations can have implications to Ca and P supply in children, and I and Zn supply across different consumer demographics; while the seasonal variation was more pronounced than that associated with production system.

1. Introduction

Milk is a major source of macrominerals and essential trace elements in human nutrition (Givens, 2020; Górska-Warsewicz, Rejman, Lasowski, & Czechtoko, 2019; Thorning et al., 2016). In the UK, milk and dairy products contribute 50–64% of I intake in children, 40% in adolescents, and 32–41% in adults (NDNS, 2020); which is essential to thyroid hormone generation and metabolism, as well as child physical and neurocognitive development in the womb as it relates to maternal I sufficiency (Dineva, Hall, Tan, Blaskova, & Bath, 2022; Gunnarsdóttir & Dahl, 2012; Levie et al., 2019). Milk and dairy products also contribute 44–59% of Ca intake in children, 34% in adolescents, and 34–44% in adults (NDNS, 2020); which is associated with roles in neurotransmitter secretion, skeletal health, and muscle contraction and as such assists in cardiovascular maintenance and thus positively associated with prevention of hypertensive disorders and vascular function in mothers and their children (Cormick & Belizán, 2019; Theobald, 2005). In addition, milk and dairy products are excellent sources of P, Zn, K, and Mg, covering 23–32%, 15–35%, 11–30%, 10–25% of the intakes of these minerals, respectively, across the different demographics of the population (McAlister et al., 2020). These minerals are of particular importance to human nutrition and health as they are involved in adenosine

triphosphate synthesis and bone structure (for P; (Calvo & Lamberg-Allardt, 2015; Serna & Bergwitz, 2020)); a multitude of biochemical functions, neurobehavioral development, and general positive growth outcomes (for Zn; (Brown, Wuehler, & Peerson, 2001; Li et al., 2022; Roohani, Hurrell, Kelishadi, & Schulin, 2013)); increased cardiovascular health, positive cell function, and renal health (for K, (He & MacGregor, 2008; Kumssa, Joy, & Broadley, 2021; Stone, Martyn, & Weaver, 2016)); and as a necessary component of enzymatic reactions within the body, transmembrane transport, and neuromuscular function (for Mg; (Al Alawi, Majoni, & Falhammar, 2018; Costello, Wallace, & Rosanoff, 2016; Schwalfenberg & Genuis, 2017)).

Retail milk mineral concentrations may vary due to differences in agricultural practices and in particular dairy management system, animal diet, and breed. For example, conventional milk is often found to be variable in trace element density when compared to organic milk (Bath, Button, & Rayman, 2012; Manzi & Durazzo, 2017; Payling, Juniper, Drake, Rymer, & Givens, 2015; Qin et al., 2021; Rey-Crespo, Miranda, & López-Alonso, 2013). In most studies, organic (ORG) milk had lower concentrations of I, Cu and Zn than conventional (CON) milk at retail and farm (Bath et al., 2012; Payling et al., 2015; Qin et al., 2021; Rey-Crespo et al., 2013; Stevenson, Drake, & Givens, 2018). Other studies at farm level, also added that ORG milk had higher concentrations of Ca,

* Corresponding author.

E-mail address: s.stergiadis@reading.ac.uk (S. Stergiadis).

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K, Mo, and P and lower concentrations of Al, Mn, Fe, and Se (Qin et al., 2021; Rey-Crespo et al., 2013). Such differences between dairy production systems have been previously associated to differences in animal diets, more specifically higher grazing intake, total forage intake, and type of matter intake in organic systems when compared with conventional systems (Qin et al., 2021; Rey-Crespo et al., 2013). Those differences in animal diets may also vary across seasons, and previous studies have identified a strong temporal variation on milk mineral concentrations (Li, Ye, & Singh, 2019; Nantapo & Muchenje, 2013; O'Brien, Mehra, Connolly, & Harrington, 1999; Payling et al., 2015; Qin et al., 2021; Stergiadis et al., 2021; Stevenson et al., 2018). Temporal variation in milk mineral concentrations is generally associated with the systemic use of feed, as the proportions of concentrate and grazing/cut-and-carry plant material is associated with the availability of such, with less availability during the colder months of the year. In most studies, milk collected in summer (typically associated with higher pasture intakes) contained less Cu, I, Mn, and Zn than milk collected in winter (Nantapo and Muchenje, 2013; O'Brien et al., 1999; Qin et al., 2021; Rey-Crespo et al., 2013; Stevenson et al., 2018). In some occasions, in studies performed at retail and farm levels, summer milk showed lower concentrations of Al, As, Ca, Na, Mg, Mo, and K, and higher concentrations of Ni (Li et al., 2019; Nantapo and Muchenje, 2013; O'Brien et al., 1999; Qin et al., 2021; Rey-Crespo et al., 2013). In addition, an effect of breed, especially the use of non-Holstein genetics in the herd, has also been reported to affect milk mineral concentrations (Cerbulis & Farrell, 1976; Manuelian, Penasa, Visentin, Zidi, Cassandro, & De Marchi, 2018; Nantapo and Muchenje, 2013; Qin et al., 2021; Stergiadis et al., 2021). For example, when compared with milk produced by Holstein/Friesian, Jersey cows have produced milk with lower concentrations of Na, and K, but higher concentrations of Al, Ca, Mg, and P; although these results were not statistically significant across all studies (Cerbulis and Farrell, 1976; Manuelian et al., 2018; Nantapo and Muchenje, 2013). In previous multivariate redundancy analyses, non-Holstein genetics were mainly positively correlated to Ca and P; and mainly negatively correlated to Al, Cu and Fe (Qin et al., 2021; Stergiadis et al., 2021).

Given that milk is a main contributor of minerals in human diets, and the fact that agricultural and seasonal parameters affects the concentration of minerals, this may have an impact on the mineral intakes of consumers of the produced milk. Previous work on variation of retail milk mineral concentrations relied on a small range of minerals, particularly I (Bath et al., 2012; Payling et al., 2015; Stevenson et al., 2018) sampling specific months (rather than across the whole year) and a restricted number of brands available to the consumer, and focused on milk products differentiated by the production system without accounting for products marketed based on animal breed. This study therefore aimed to (i) investigate the effect of milk production system, as available at retail (conventional, organic, channel island), month (January through to December) and their interaction, on retail milk concentrations of macrominerals and trace elements throughout the year, and (ii) assess the potential nutritional implications (total intakes, contribution towards reference nutrient intakes (RNI)) of macrominerals and trace elements on consumers.

2. Materials and methods

2.1. Experimental design

The present study analysed milk samples ($n = 473$) from retail outlets collected in South East England. Milk sample collection was performed monthly, over 12 months (January–December 2019) and included 42 brands of milk, of which 26 represented conventional (CON) milk, 12 represented organic (ORG) milk and 4 represented milk produced by channel island breeds (CHA; Jersey, or Jersey & Guernsey). All brands were whole pasteurised milk and samples collected to represent

the latest “use by” date available on the shelf at the day of sampling, to ensure maximum potential freshness. Milk samples, in their commercial packaging, were immediately transferred to the laboratories of the University of Reading, and aliquoted into 30 mL sterile polypropylene screw-top containers. One aliquot was preserved with bronopol and sent to National Milk Laboratories (Wolverhampton, UK) for determination of milk basic composition and somatic cell count (SCC), while two other aliquots were frozen at -20°C until analysis. One of the frozen aliquots was lyophilised and sent to Matis (Reykjavik, Iceland) for the determination of macrominerals, trace elements and potential toxic elements, while the other frozen aliquot was thawed in 4°C overnight and used for the determination of milk I concentrations at the University of Reading.

2.2. Milk analysis

Milk basic composition, including fat, protein, casein, whey protein, lactose, and urea, was estimated by National Milk Laboratories (Wolverhampton, UK) using a Milkoscan FT6000 (Foss Electric, Hillerød, Denmark), while milk somatic cell count (SCC) was assessed using a Fossomatic (Foss Electric, Hillerød, Denmark). Assessment of milk mineral concentrations was performed in technical duplicates, and included macrominerals (Ca, K, Mg, Na, P), essential trace elements (Cu, Fe, I, Mn, Mo, Zn), non-essential trace elements (Al, Sn) and potential toxic elements (As, Cd, Co, Cr, Hg, Ni, Pb). Analysis for all minerals, except I, was performed in Matis (accredited laboratory for mineral analyses; Reykjavik, Iceland) using lyophilised milk samples (200 mg), digested in an Ultra-wave Acid Digestion System (Milestone Inc., Italy) in 12 mL quartz tubes and 1 mL nitric acid (HNO_3 , ROTIPURAN Supra, 69%, Carl Roth, Karlsruhe, Germany) and 1 mL hydrogen peroxide (H_2O_2 , for trace analysis, $\geq 30\%$, Supelco, Sigma-Aldrich, France) and then proceeded to mineral analyses following dilution using inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7900, Agilent Technologies, Singapore) (NMKL 186, 2007; Qin et al., 2023). Certified reference material (CRM) DORM-5 (Fish protein certified reference material for trace metals, National Research Council Canada) and a matrix matched CRM skimmed milk powder ERM-BD150 (trace elements, European Reference Materials) were included with each analysis. Indium was used as a continuous internal standard. The analysis of I was performed at the University of Reading according to method described by Newton et al. (2021). In brief, 100 μL of milk was diluted to 10 mL with 2% tetramethylammonium hydroxide in ultrapure H_2O , syringed to remove milk solids, and then proceeded to mineral analyses following dilution using inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7900, Agilent Technologies, Singapore). CRM skimmed milk powder ERM-BD150 (trace elements, European Reference Materials) was included with each analysis. Instrumental parameters for the ICPMS, monitored masses, and the Ultrawave digestion program can be found in supplementary Table S1.

2.3. Statistical analysis

Statistical analysis was carried out in Minitab® 21.2. Data analysis was performed using linear mixed effects model using milk production system, month, and their interaction as fixed factors, and Milk ID (nested within milk production system) as a random factor. Milk ID was a unique identifier given to each individual brand collected. The normality of residuals was visually assessed and there was no deviation of normality for any of the presented variables; hence all variables were analysed untransformed. When the effect of the fixed factors was significant for a measured variable ($P < 0.05$), pairwise comparisons to assess significant differences between the predicted means were carried out using Tukey's Honestly Significant Difference test ($P < 0.05$).

3. Results

3.1. Milk basic composition

3.1.1. Effect of milk production system

There was a significant effect of milk production system on all basic composition parameters, except for SCC (Table 1). CHA milk contained more fat than CON (+0.58 g/100 g milk) and ORG (+0.29 g/100 g milk). CHA milk had higher concentrations of protein than CON (+0.54 g/100 g milk) and ORG (+0.57 g/100 g milk); and this was observed for both casein (+0.48 and +0.51 g/100 g milk, vs CON and ORG, respectively) and whey protein (+0.07 and +0.07 g/100 g milk, vs CON and ORG, respectively). Lactose concentrations were highest in CON milk than in ORG milk (+0.03 g/100 g milk), while the opposite has been observed for the fat:protein ratio. ORG milk showed lower concentrations of urea than CON (−40 mg/kg milk) and CHA (−42 mg/kg milk). There was also a tendency for higher SCC in CHA milk than CON and ORG milks, but the difference was not statistically significant.

3.1.2. Effect of month

There was a significant effect of month on all basic composition parameters (Table 2). However, the seasonal effect for fat, protein, casein, whey protein and lactose, although significant, has been relatively small in terms of numerical values. For example, the variation in their concentrations represented a difference of 0.55, 0.21, 0.20, 0.07 and 0.09 g/100 g milk between the minimum and maximum concentrations, respectively. The months April–August had higher SCC than milk in January but the differences when comparing to other months were not significant. Urea was higher between August–December and January, than in February–July.

Table 1

Effect of milk production system (conventional, CON; organic, ORG; channel island, CHA) on the basic composition and mineral profile of retail milk collected throughout the year.

	Milk production system			SE	P-values ^a
	CON	ORG	CHA		
Parameters assessed	n = 280	n = 135	n = 58		
Basic composition					
Fat (g/100 g milk)	3.59 ^B	3.88 ^A	4.17 ^A	0.137	**
Protein (g/100 g milk)	3.35 ^B	3.32 ^B	3.89 ^A	0.027	***
Casein (g/100 g milk)	2.63 ^B	2.60 ^B	3.11 ^A	0.021	***
Whey protein (g/100 g milk)	0.72 ^B	0.72 ^B	0.79 ^A	0.008	***
Lactose (g/100 g milk)	4.52 ^A	4.49 ^B	4.52 ^{AB}	0.014	*
SCC (x 10 ³ /mL milk)	54.3	65.6	96.6	15.6	†
Fat:protein (g/g)	1.07 ^B	1.17 ^A	1.08 ^{AB}	0.042	*
Urea (mg/kg milk)	221 ^A	181 ^B	223 ^A	8.2	***
Macrominerals (mg/kg milk)					
Ca	976 ^B	971 ^B	1087 ^A	21.4	***
K	1269 ^A	1256 ^A	1164 ^B	26.5	***
Mg	89 ^B	86 ^B	98 ^A	1.9	***
Na	303	300	304	7.0	ns
P	720 ^B	708 ^B	768 ^A	15.8	*
Essential trace elements (µg/kg milk unless otherwise stated)					
Cu	39.0 ^B	37.7 ^B	46.3 ^A	1.13	***
Fe (mg/kg milk)	0.22 ^B	0.24 ^A	0.24 ^{AB}	0.012	*
I	326 ^A	317 ^A	217 ^B	28.1	**
Mn	21.8 ^B	21.5 ^B	28.5 ^A	0.94	***
Mo	98 ^B	129 ^A	108 ^B	4.14	***
Zn (mg/kg milk)	3.59 ^B	3.64 ^B	4.26 ^A	0.083	***

n = number of samples, SE = standard error, SCC = somatic cell count.

^a Significances were declared at ***, P < 0.001; **, P < 0.01; *, P < 0.05; †, 0.05 < P < 0.10 (trend); ns, P > 0.10 (non-significant). Means for production system within a row with different upper-case letters are significantly different according to Tukey's Honestly Significant Difference test (P < 0.05).

3.1.3. Effect of production system × month interaction

The interaction milk production system × month significantly affected milk concentrations of fat, protein, casein, whey protein and urea (Supplementary Information; Fig. S1). CHA milk contained more fat than CON and ORG milk in February and March, but the differences between the three types of milk were not significant for any other months. The concentrations of protein, casein and whey protein in CHA milk were higher than CON and ORG milk throughout the study. CON milk had higher urea contents than ORG milk in April–July, but there were no other differences between the three types of milk at any other months.

3.2. Milk mineral profiles

3.2.1. Effect of milk production system

There was a significant effect of milk production system on the concentrations of all macrominerals and trace elements, except for Na (Table 1). When compared with CON and ORG milk, CHA milk contained more Ca (+111 and +116 mg/kg milk, respectively), Mg (+9 and +12 mg/kg milk, respectively), P (+48 and +60 mg/kg milk, respectively), Cu (+7.3 and +8.6 µg/kg milk, respectively), Mn (+6.7 and +7.0 µg/kg milk, respectively), and Zn (+0.67 and +0.32 µg/kg milk, respectively); while it contained less K (−127 and −125 mg/kg milk, respectively) and I (−109 and ORG −100 µg/kg milk, respectively). When compared with CON and CHA milk, ORG milk contained more Mo (+31 and +21 µg/kg milk, respectively); while it also contained 0.02 mg/kg milk more Fe than CON milk.

3.2.2. Effect of month

There was a significant effect of month on the concentrations of all macrominerals and essential trace elements in milk (Table 2). Milk Ca concentrations varied by 543 mg/kg milk (from 1358 in December to 815 in February); while November milk also had high Ca concentrations, the other months showing intermediate values without been significant differences between them. Milk K concentrations varied by 225 mg/kg milk (from 1345 in December to 1120 in February), but the difference was statistically significant only between these two months, plus between December and November (+204 mg/kg milk in December). Milk Mg concentrations varied by 29.7 mg/kg milk (from 108.7 in December to 79.0 in February), with the other months showing intermediate values without been significant differences between them. Milk Na concentrations varied by 83 mg/kg milk (from 258 in November to 341 in January), while other months with low and high values were January and Oct, respectively. Milk P concentrations varied by 223 mg/kg milk (from 626 in February to 849 in December), with the other months showing intermediate values without been significant differences between them.

Milk Cu concentrations varied by 23.5 µg/kg milk (from 33.7 in August to 57.2 in February), with May–October mainly showing lower concentrations (<40 µg/kg) than the rest of the months (40.0–57.2) although not all differences between individual months were statistically significant. Milk Fe concentrations varied by 0.17 mg/kg milk (from 0.19 in August–September to 0.36 mg/kg milk in February), while there were not statistically significant differences between the rest of the months. Milk I concentrations varied by 356 µg/kg milk (from 95 in May to 451 µg/kg milk in January). They were lowest between April–June (≤219 µg/kg milk); had increased values in February, July–September, December (244–284 µg/kg milk); and reached at their highest during January, March and November (>364 µg/kg milk). Milk Mn concentrations varied by 16.4 µg/kg milk (from 16.7 in October to 33.1 in February), with the monthly differences in Mn concentrations been primarily not significant for the remaining months. Milk Mo concentrations were lower between March–October (97–111 µg/kg milk) than January–February and November–December (114–142 µg/kg milk), although not all differences were statistically significant. Zn concentrations in milk varied by 1.9 mg/kg milk (from 3.3 in August to 5.24

Table 2

Effect of sampling month on the basic composition and mineral profile of retail milk collected throughout the year.

	Month												SE	P-values ^a
	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec		
Parameters assessed	n = 39	n = 42	n = 40	n = 39	n = 41	n = 40	n = 38	n = 40	n = 40	n = 40	n = 37	n = 37		
Basic composition														
Fat (g/100 g milk)	3.94 ^{AB}	4.14 ^A	3.94 ^{AB}	3.59 ^C	3.90 ^{ABC}	4.00 ^{AB}	3.82 ^{BC}	3.95 ^{AB}	3.81 ^{BC}	3.89 ^{ABC}	3.87 ^{ABC}	3.70 ^{BC}	0.092	***
Protein (g/100 g milk)	3.51 ^{DE}	3.46 ^{EF}	3.47 ^{EF}	3.51 ^{DE}	3.50 ^{DE}	3.49 ^{DE}	3.46 ^{EF}	3.40 ^F	3.56 ^{CD}	3.59 ^{BC}	3.67 ^A	3.65 ^{AB}	0.019	***
Casein (g/100 g milk)	2.76 ^{EF}	2.71 ^{FG}	2.73 ^{EFG}	2.75 ^{EFG}	2.78 ^{CDE}	2.77 ^{DE}	2.72 ^{EFG}	2.70 ^G	2.83 ^{BCD}	2.84 ^{BC}	2.91 ^A	2.86 ^{AB}	0.016	***
Whey protein (g/100 g milk)	0.75 ^{CD}	0.75 ^{BC}	0.74 ^{DE}	0.76 ^B	0.73 ^{EF}	0.72 ^F	0.73 ^{DE}	0.70 ^G	0.73 ^{EF}	0.75 ^{BC}	0.75 ^{BC}	0.79 ^A	0.004	***
Lactose (g/100 g milk)	4.54 ^{AB}	4.52 ^{BCDE}	4.54 ^{AB}	4.55 ^A	4.53 ^{ABC}	4.52 ^{ABCD}	4.50 ^{CDEF}	4.48 ^{FG}	4.48 ^{FG}	4.46 ^G	4.49 ^{DEFG}	4.49 ^{FG}	0.009	***
SCC (x 10 ³ /mL milk)	53.2 ^B	64.1 ^{AB}	71.4 ^{AB}	77.8 ^A	74.2 ^A	75.9 ^A	79.9 ^A	83.8 ^A	71.2 ^{AB}	73.2 ^{AB}	70.7 ^{AB}	70.2 ^{AB}	7.96	**
Fat:protein (g/g)	1.13 ^{ABCD}	1.19 ^A	1.14 ^{ABC}	1.04 ^{DE}	1.11 ^{ABCDE}	1.15 ^{ABC}	1.11 ^{ABCD}	1.16 ^{AB}	1.07 ^{BCDE}	1.10 ^{BCDE}	1.07 ^{CDE}	1.02 ^E	0.027	***
Urea (mg/kg milk)	232 ^B	162 ^C	188 ^C	178 ^C	162 ^C	176 ^C	162 ^C	223 ^B	292 ^A	246 ^B	249 ^B	230 ^B	7.0	***
Macrominerals (mg/kg milk)														
Ca	954 ^{BC}	815 ^C	911 ^{BC}	976 ^B	934 ^{BC}	1014 ^B	1005 ^B	927 ^{BC}	1004 ^B	1012 ^B	1227 ^A	1358 ^A	33.6	***
K	1201 ^{AB}	1120 ^B	1166 ^{AB}	1218 ^{AB}	1288 ^{AB}	1264 ^{AB}	1292 ^{AB}	1221 ^{AB}	1295 ^{AB}	1205 ^{AB}	1141 ^B	1345 ^A	41.0	***
Mg	89.4 ^{BC}	79.0 ^C	86.5 ^{BC}	90.9 ^{BC}	91.3 ^{BC}	93.4 ^B	94.3 ^B	86.7 ^{BC}	94.7 ^B	88.2 ^{BC}	89.4 ^{BC}	108.7 ^A	2.94	***
Na	341 ^A	285 ^{BCD}	313 ^{ABC}	280 ^{BCD}	283 ^{BCD}	305 ^{ABCD}	311 ^{ABC}	325 ^{AB}	345 ^A	276 ^{CD}	258 ^D	301 ^{ABCD}	12.2	***
P	711 ^{BC}	626 ^C	700 ^{BC}	728 ^{BC}	694 ^{BC}	753 ^{AB}	760 ^{AB}	723 ^{BC}	779 ^{AB}	735 ^B	724 ^{BC}	849 ^A	24.7	***
Essential trace elements (µg/kg milk unless otherwise stated)														
Cu	42.3 ^{BC}	57.2 ^A	40.0 ^{CD}	42.5 ^{BC}	39.4 ^{CD}	37.2 ^{CD}	34.4 ^D	33.7 ^D	36.1 ^{CD}	38.8 ^{CD}	41.9 ^{BC}	48.7 ^B	1.60	***
Fe (mg/kg milk)	0.20 ^C	0.36 ^A	0.21 ^C	0.21 ^C	0.30 ^{AB}	0.25 ^{BC}	0.21 ^C	0.19 ^C	0.19 ^C	0.23 ^{BC}	0.22 ^C	0.26 ^{BC}	0.018	***
I	451 ^A	284 ^C	401 ^{AB}	219 ^{CD}	95 ^E	166 ^{DE}	273 ^C	249 ^C	244 ^C	364 ^B	433 ^{AB}	262 ^C	20.6	***
Mn	22.0 ^{BCD}	33.1 ^A	25.6 ^{BC}	27.6 ^{AB}	26.5 ^{BC}	22.7 ^{BCD}	21.3 ^{BCD}	20.2 ^{CD}	22.9 ^{BCD}	16.7 ^D	22.3 ^{BCD}	25.9 ^{BC}	1.46	***
Mo	117 ^{BC}	142 ^A	99 ^C	107 ^C	111 ^C	106 ^C	98 ^C	97 ^C	104 ^C	110 ^C	114 ^{BC}	132 ^{AB}	4.7	***
Zn (mg/kg milk)	3.79 ^{BCD}	5.24 ^A	3.74 ^{BCD}	3.89 ^{BCD}	3.94 ^{BC}	3.51 ^{CD}	3.40 ^{CD}	3.33 ^D	3.35 ^D	3.84 ^{BCD}	3.71 ^{BCD}	4.23 ^B	0.130	***

n = number of samples, SE = standard error, SCC = somatic cell count

^a Significances were declared at ***, P < 0.001; **, P < 0.01; *, P < 0.05; †, 0.05 < P < 0.10 (trend); ns, P > 0.10 (non-significant). Means for month within a row with different upper-case letters are significantly different according to Tukey's Honestly Significant Difference test (P < 0.05)

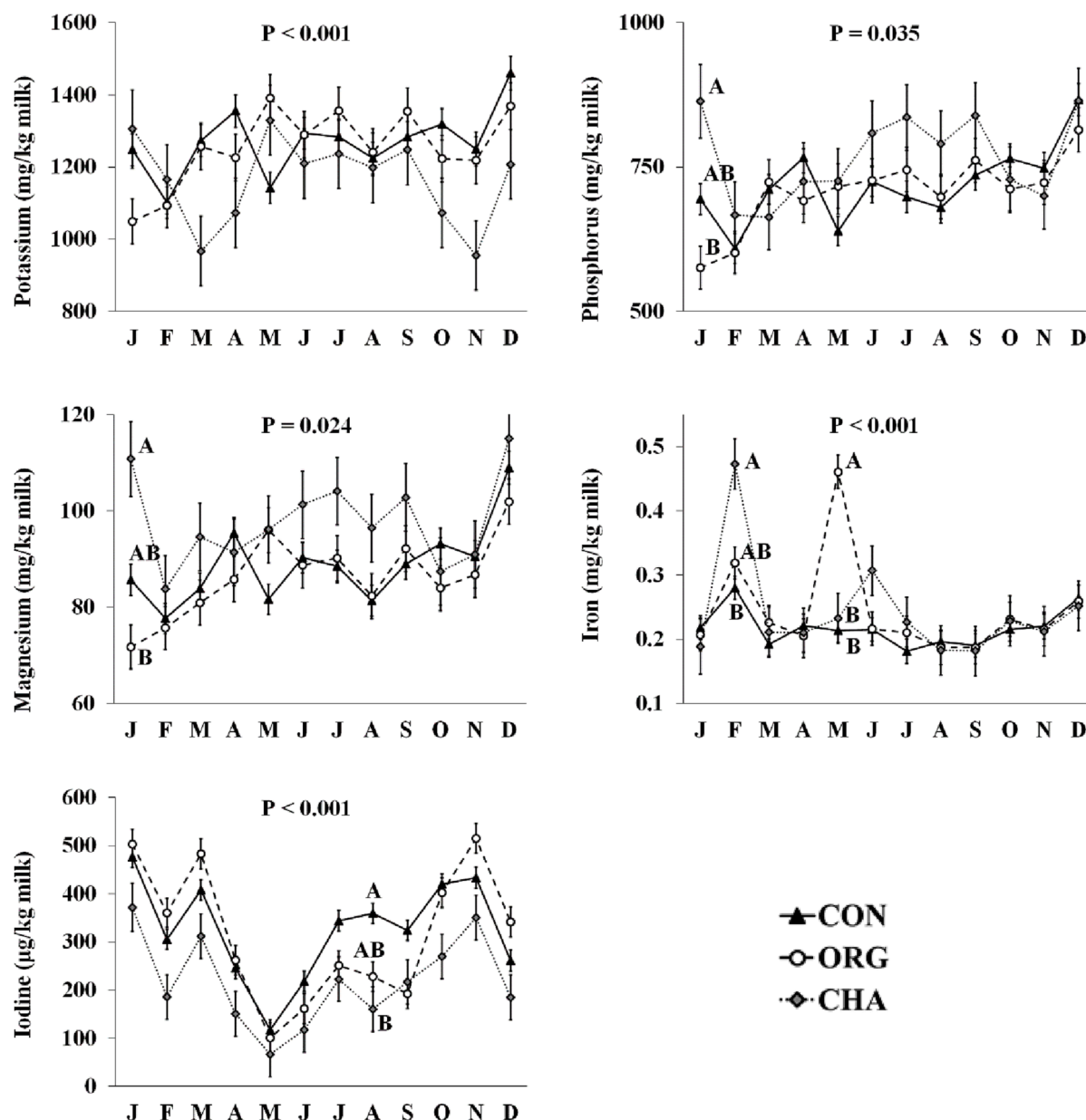


Fig. 1. Interaction means \pm SE (standard error bars) for the effects of milk production system (conventional, CON; organic, ORG; channel island, CHA) and month (in order of appearance from left to right in Axis Y: J, January; F, February; M, March; A, April; M, May; J, June; J, July; A, August; S, September; O, October; N, November; D, December) on mineral profiles of retail milk collected throughout the year. P represents the P-value for the interaction. Means for milk production system and within a month with different upper-case letters are significantly different according to Tukey's Honestly Significant Difference test ($P < 0.05$).

mg/kg milk in February), with the monthly differences in Zn concentrations been primarily not significant for the remaining months.

3.2.3. Effect of milk production system \times month interaction

The interaction milk production system \times month significantly affected milk concentrations of Ca, K, Mg, Na and P in milk (Fig. 1). CHA milk contained less Ca, K, Mg, Na, and P than CON and ORG milk in February, but higher concentrations of all these minerals in January (although the difference was not statistically significant for K and Na). Higher concentrations of Fe in CHA milk, than in CON and ORG milk, were also observed in February and May although the difference between CHA and ORG milk was not statistically significant in February. Milk I concentrations in CHA milk were numerically lower throughout the year, but the only month that this reached statistical significance, when the type \times month interaction was assessed, was August; with CHA

milk containing significantly less I than CON milk.

3.2.4. Non-essential trace elements and potential toxic elements

Most non-essential trace elements (Al, Sn) and potential toxic elements (As, Cd, Co, Cr, Hg, Ni and Pb) were present in trace concentrations and the majority of their individual measurements were below the limits of quantification (which were: 174.6 $\mu\text{g/kg}$ milk for Al, 3.5 $\mu\text{g/kg}$ milk for Sn, 5.8 $\mu\text{g/kg}$ milk for As, 0.6 $\mu\text{g/kg}$ milk for Cd, 5.8 $\mu\text{g/kg}$ milk for Cr, 3.5 $\mu\text{g/kg}$ milk for Co, 3.5 $\mu\text{g/kg}$ milk for Hg, 5.8 $\mu\text{g/kg}$ milk for Ni, and 3.5 $\mu\text{g/kg}$ milk for Pb). The proportion of measured values that were below these limits were as follows: Al (95.4%), Sn (96.0%), As (99.8%), Cd (91.2%), Cr (89.8%), Co (100%), Hg (100%), Ni (97.0%), Pb (94.8%). Therefore, the results of these elements were not statistically analysed; but scatter plots to present the measurements (and their relation to limits of quantification) for all macrominerals, trace elements

and potential toxic elements, are presented in Fig. S2.

4. Discussion

4.1. Milk basic composition

The higher concentrations of fat, protein and casein in CHA milk compared with CON milk are in line with a previous study in the North East England, with samples been collected between August 2006 and January 2008 (Stergiadis, Seal, Leifert, Eyre, Larsen, & Butler, 2013). Although the higher fat content in milk from Jersey and Guernsey cows has been previously reported at animal and retail levels (Carroll, DePeters, Taylor, Rosenberg, Perez-Monti, & Capps, 2006), it should be noted that retail milk fat content is also impacted by the processing method of the different supply chains (standardisation) and therefore not necessarily reflect the milk fat content produced by the animals; although CON and ORG milk are fat standardised in the dairy plant, the same may not necessarily be true for CHA milk. The higher protein concentrations in milk from Jersey and Guernsey cows, when compared with milk from Holstein cows has also been previously reported in studies at animal level (Carroll et al., 2006; Cerbulis and Farrell, 1976) and in retail milk (Stergiadis et al., 2013). The latter study has shown that the higher protein content is mostly an effect of the higher casein, rather than whey protein; a fact that is also confirmed in the present study. The seasonal variation in protein content was rather small (although statistically significant) with milk produced in October–December having higher protein contents than milk produced the rest of the year. It is likely that the increased supply in concentrates and the lower contribution of pasture and forages in winter diets (Qin et al., 2021; Stergiadis et al., 2012) may have increased the starch contents in cows' diets, which may support higher milk protein synthesis through the increased synthesis of propionic acid in the rumen and delivery to the mammary gland (McDonald et al., 2010).

4.2. Variation on the concentrations of minerals in milk from different production systems (conventional, organic, channel island)

When compared with CON and ORG milk, CHA milk contained more Ca, Mg, P, and Zn; while it contained less K and I. Casein micelles are responsible to host the most important milk cations and 70% of Ca and 35% of Mg are found within them, whilst 50% of inorganic phosphate (P) is also associated with the solid fraction in milk (Gaucheron, 2005; Holt & Jenness, 1984); thus milk protein concentration is typically positively correlated with Ca, Mg, and P in cows' milk (Dunshea et al., 2019). In the present study, CHA milk contained more protein and casein (thus in line with previous studies (Carroll et al., 2006; Cerbulis and Farrell, 1976; Stergiadis et al., 2013)), and this may have increased the concentrations of Ca, Mg, and P. The higher content of Ca, P, and Mg is in line with previous work (Cerbulis and Farrell, 1976; Manuelian et al., 2018), although Cerbulis and Farrell (1976) found no difference on Mg between Jersey and Holstein cows. The lower K in CHA milk is in line with other studies (Manuelian et al., 2018). A previous study showed positive correlation of K with grazing intake and total forage (Qin et al., 2021), but in the absence of clear indicators in literature that CHA farms offer less pasture to cows compared to organic or conventional herds, it is difficult to conclude that this can be the reason of the difference.

However, this hypothesis (that CHA herds may typically receive less pasture and more concentrates) would also align with the lower Zn and I concentrations in CHA milk; concentrates (fed in higher amounts in cases of lower pasture intake) typically contain more Zn and I than pasture and conserved forages (Orjales et al., 2018) while milk Zn and I concentrations have been found to be negatively correlated to pasture intakes (Qin et al., 2021). Interestingly, the present study contradicts previous work, in South East England, showing that retail ORG milk contained less I than retail CON milk in the UK, with milk samples

collected in 2009 (Bath et al., 2012), 2014 (Payling et al., 2015) and 2015 (Stevenson et al., 2018); but agrees with a farm survey, in South East England, with milk samples collected in 2019 (same year as the present retail survey) (Qin et al., 2021). It is likely that following the original findings in the earlier retail surveys (Bath et al., 2012; Stevenson et al., 2018), organic dairy farmers may have enriched cows' diet in I in the form of potassium iodide, anhydrous calcium iodate (permitted by organic regulations (Soil Association, 2021)) or via organic seaweed supplementation which is known to increase milk I concentrations (Newton et al., 2021; Qin et al., 2023), and by the time of sample collection in the published farm survey (Qin et al., 2021) and the present retail survey, the concentrations of I in ORG and CON retail milks did not differ.

When compared with CON and CHA milk, ORG milk contained more Mo; while it also contained more Fe than CON milk. A previous study in the South East England (Qin et al., 2021), which measured milk minerals concentrations from 43 conventional and 27 organic farms, also found that Mo was higher in ORG milk although the result for Fe was the opposite (higher in CON milk). The same study has showed that milk Mo concentrations were positively correlated with total forage intakes and the contribution of non-Holstein breeds in the herd, and this may potentially explain their higher concentrations in ORG milk both in this previous, but also in the present study, as ORG herds in South East England typically consume more forage and are made of more cows, either purebred or crossbreds (Qin et al., 2021). However, milk Fe concentrations were negatively correlated to the same husbandry practices, as well as pasture intake, while previous work shows that the feed-to-milk transfer efficiencies are very low (<1%; Qin et al., 2023) while other studies highlighted the important role of breed on milk Fe concentrations (Zwierchowski & Ametaj, 2018). It is therefore unclear why ORG milk would have more Fe than CON milk in the present study but it should be noted that the difference was rather minimal (+20 µg per kg milk).

4.3. Seasonal variation in the concentrations of minerals in milk

The minimum concentrations of most macrominerals (Ca, K, Mg, P) were found in February, except for Na which was found in November; while the maximum amounts were found in December, except for Na which was found in September. Milk protein concentration is typically positively correlated with Ca, Mg, and P in cows' milk (Dunshea et al., 2019; Gaucheron, 2005; Holt and Jenness, 1984). In the present study, milk protein concentrations were among the highest in December and among the lowest in February; and therefore, the differences in milk mineral concentrations may not necessarily be due to differences in mineral supplementation practices in dairy farms, but more a result of the lower protein concentrations in these months. A previous multivariate redundancy analysis in data from dairy farms in the South East England showed that husbandry practices which were positively correlated with milk protein concentrations, had also a positive correlation with Ca and P concentrations (Qin et al., 2021); while, in addition, such relationship was also observed for milk Mg concentrations in case of organic dairy farms (Stergiadis et al., 2021). K and Na are found in the aqueous phase liquid part of milk as free ions, while other amounts of K and Na associate with citrate, inorganic phosphate and chloride (Gaucheron, 2005). A previous study showed a positive correlation of Na and K with intakes of pasture and total forage respectively (both typically higher in dairy farms between April–October (Qin et al., 2021; Stergiadis et al., 2012), but the variation in their milk concentrations in the present study did not show any association with potential seasonal variation in forage intakes.

The minimum concentrations of all trace elements (Cu, Fe, I, Mn, Mo, Zn) were found during grazing/outdoor periods (typically April–October in the UK dairy farms, as recorded by studies in South East and North East previously (Qin et al., 2021; Stergiadis et al., 2012; Stergiadis et al., 2021)) while the maximum were observed during the indoor housing

Table 3Estimated macromineral intake from different milk production systems and contributions towards the reference nutrient intakes (RNI)^a for the different age groups in the UK population.

Age group	Milk intake ^b (g/d)	% of Ca RNI covered from				% of K RNI covered from				% of Mg RNI covered from				% of P RNI covered from			
		RNI for Ca (mg/d)	CON	ORG	CHA ^c	RNI for K (mg/d)	CON	ORG	CHA ^c	RNI for Mg (mg/d)	CON	ORG	CHA ^c	RNI for P (mg/d)	CON	ORG	CHA ^c
Children 1.5–3 years	247	350	69	69	77	800	39	39	36	85	26	25	28	270	66	65	70
Boys 4–10 years	181	500	35	35	39	1550	15	15	14	160	10	10	11	400	33	32	35
Girls 4–10 years	160	500	31	31	35	1550	13	13	12	160	9	9	10	400	29	28	31
Children 4–10 years	170	500	33	33	37	1550	14	14	13	160	9	9	10	400	31	30	33
Boys 11–18 years	129	1000	13	13	14	3300	5	5	5	290	4	4	4	775	12	12	13
Girls 11–18 years	113	800	14	14	15	3300	4	4	4	290	3	3	4	625	13	13	14
Adolescents 11–18 years	121	900	13	13	15	3300	5	5	4	290	4	4	4	700	12	12	13
Men 19–64 years	136	700	19	19	21	3500	5	5	5	300	4	4	4	550	18	17	19
Women 19–64 years	106	700	15	15	16	3500	4	4	4	270	3	3	4	550	14	14	15
Adults 19–64 years	121	700	17	17	19	3500	4	4	4	285	4	4	4	550	16	16	17
Men 65+ years	186	700	26	26	29	3500	7	7	6	300	6	5	6	550	24	24	26
Women 65+ years	150	700	21	21	23	3500	5	5	5	270	5	5	5	550	20	19	21
Adults 65+ years	166	700	23	23	26	3500	6	6	6	285	5	5	6	550	22	21	23
Men 65–74 years	154	700	21	21	24	3500	6	6	5	300	5	4	5	550	20	20	21
Women 65–74 years	122	700	17	17	19	3500	4	4	4	270	4	4	4	550	16	16	17
Adults 65–74 years	137	700	19	19	21	3500	5	5	5	285	4	4	5	550	18	18	19
Men 75+ years	214	700	30	30	33	3500	8	8	7	300	6	6	7	550	28	28	30
Women 75+ years	177	700	25	25	27	3500	6	6	6	270	6	6	6	550	23	23	25
Adults 75+ years	193	700	27	27	30	3500	7	7	6	285	6	6	7	550	25	25	27
Pregnant women ^d	106	805	13	13	14	3750	4	4	3	300	3	3	3	550	14	14	15
Lactating women ^d	106	805	13	13	14	3750	4	4	3	300	3	3	3	550	14	14	15

^a Reference nutrient intakes, as presented in the National Diet and Nutrition Survey UK Results from Years 9–11 of the Rolling Programme (NDNS, 2020).^b Estimated based on energy intakes and % of energy intake from milk for the different demographics in the National Diet and Nutrition Survey UK Results from Years 9–11 of the Rolling Programme (NDNS, 2020) and the average caloric content of different milks according to the McCance and Widdowson's 'composition of foods integrated dataset' on the nutrient content of the UK food supply (Public Health England, 2021).^c Significantly different ($P < 0.05$) compared to the other two types of milk.^d Assuming a similar milk intake with women 19–64 years of age, and using the population average requirement for Ca, adequate intakes for K, Mg and P for pregnant and lactating women from the European Food Safety Authority Panel on Dietetic Products & Allergies (European Food Safety Authority, 2019).

Table 4
Estimated trace element intake from different milk production systems and contributions towards the reference nutrient intakes (RNI)^a or adequate intakes (AI)^a for the different age groups in the UK population.

Age group	Milk intake ^b (g/d)	% of Cu RNI covered from				% of I RNI covered from				% of Mn AI covered from				% of Mo AI covered from				% of Zn RNI covered from			
		RNI for Cu (µg/d)	CON	ORG	CHA ^c	RNI for I (µg/d)	CON	ORG	CHA ^c	AI for Mn (µg/d)	CON	ORG	CHA ^c	AI for Mo (µg/d)	CON	ORG	CHA ^c	RNI for Zn (µg/d)	CON	ORG	CHA ^c
Children 1.5–3 years	247	400	2.4	2.3	2.9	70	115	112	77	500	1.1	1.1	1.4	15	161	213	178	5000	18	18	21
Boys 4–10 years	181	650	1.1	1.1	1.3	105	56	55	37	1250	0.3	0.3	0.4	25	71	93	78	6750	10	10	11
Girls 4–10 years	160	650	1.0	0.9	1.1	105	50	48	33	1250	0.3	0.3	0.4	25	63	82	69	6750	9	9	10
Children 4–10 years	170	650	1.0	1.0	1.2	105	53	51	35	1250	0.3	0.3	0.4	25	67	88	74	6750	9	9	11
Boys 11–18 years	129	900	0.6	0.5	0.7	135	31	30	21	2500	0.1	0.1	0.1	55	23	30	25	9250	5	5	6
Girls 11–18 years	113	900	0.5	0.5	0.6	135	27	27	18	2500	0.1	0.1	0.1	55	20	27	22	8000	5	5	6
Adolescents 11–18 years	121	900	0.5	0.5	0.6	135	29	29	20	2500	0.1	0.1	0.1	55	22	28	24	8625	5	5	6
Men 19–64 years	136	1200	0.4	0.4	0.5	140	32	31	21	3000	0.1	0.1	0.1	65	20	27	23	9500	5	5	6
Women 19–64 years	106	1200	0.3	0.3	0.4	140	25	24	16	3000	0.1	0.1	0.1	65	16	21	18	7000	5	6	7
Adults 19–64 years	121	1200	0.4	0.4	0.5	140	28	27	19	3000	0.1	0.1	0.1	65	18	24	20	8250	5	5	6
Men 65+ years	186	1200	0.6	0.6	0.7	140	43	42	29	3000	0.1	0.1	0.2	65	28	37	31	9500	7	7	8
Women 65+ years	150	1200	0.5	0.5	0.6	140	31	28	22	3000	0.1	0.1	0.1	65	23	30	25	7000	8	8	9
Adults 65+ years	166	1200	0.5	0.5	0.6	140	35	34	23	3000	0.1	0.1	0.2	65	25	33	28	8250	7	7	9
Men 65–74 years	154	1200	0.5	0.5	0.6	140	39	38	26	3000	0.1	0.1	0.1	65	23	30	26	9500	6	6	7
Women 65–74 years	122	1200	0.4	0.4	0.5	140	28	28	19	3000	0.1	0.1	0.1	65	18	24	20	7000	6	6	7
Adults 65–74 years	137	1200	0.4	0.4	0.5	140	32	31	21	3000	0.1	0.1	0.1	65	21	27	23	8250	6	6	7
Men 75+ years	214	1200	0.7	0.7	0.8	140	50	48	33	3000	0.2	0.2	0.2	65	32	42	36	9500	8	8	10
Women 75+ years	177	1200	0.6	0.6	0.7	140	41	40	27	3000	0.1	0.1	0.2	65	27	35	29	7000	9	10	11
Adults 75+ years	193	1200	0.6	0.6	0.7	140	45	44	30	3000	0.1	0.1	0.2	65	29	38	32	8250	8	9	10
Pregnant women ^d	106	1500	0.3	0.3	0.3	200	17	17	12	3000	0.1	0.1	0.1	65	16	21	18	8600	4	5	5
Lactating women ^d	106	1500	0.3	0.3	0.3	200	17	17	12	3000	0.1	0.1	0.1	65	16	21	18	9400	4	4	5

^a Sources of RNIs and AIs: Cu ([British Nutrition Foundation, 2021](#)), Mn ([European Food Safety Authority, 2019](#)), Zn and I ([NDNS, 2020](#)).

^b Estimated based on energy intakes and % of energy intake from milk for the different demographics in the National Diet and Nutrition Survey UK Results from Years 9–11 of the Rolling Programme ([NDNS, 2020](#)) and the average caloric content of different milks according to the McCance and Widdowson's 'composition of foods integrated dataset' on the nutrient content of the UK food supply ([Public Health England, 2021](#)).

^c Significantly different compared to the other two types of milk.

^d Assuming a similar milk intake with women 19–64 years of age, and using the AIs for Cu, I and Mn intake for pregnant and lactating women, and corrected population RNI for Zn intakes (+1.6 mg/d for pregnant women and +2.9 mg/d for lactating women) from the European Food Safety Authority Panel on Dietetic Products & Allergies ([European Food Safety Authority, 2019](#)).

Table 5Differences in the contribution of retail milk, across all types, towards the reference nutrient intakes (RNI)^a or adequate intakes (AI)^a for the different age groups in the UK population.

		Macrominerals (% RNI)										Trace elements (% RNI)											
		Ca		K		Mg		Na		P		Cu		Fe		I		Mn		Mo		Zn	
Age group	Milk intake ^b (g/d)	min Feb	max Dec	min Feb	max Dec	min Feb	max Dec	min Nov	max Sep	min Feb	max Dec	min Aug	max Feb	min Aug	max Feb	min May	max Jan	min Oct	max Feb	min Aug	max Feb	min Aug	max Feb
Children 1.5–3 years	247	58	96	35	42	23	32	13	17	57	78	2.1	3.5	0.7	1.3	34	159	0.3	0.7	160	234	17	26
Boys 4–10 years	181	29	49	13	16	9	12	5	7	28	38	0.9	1.6	0.5	0.9	16	78	0.2	0.4	70	103	9	14
Girls 4–10 years	160	26	43	12	14	8	11	4	6	25	34	0.8	1.4	0.4	0.8	14	69	0.2	0.4	62	91	8	12
Children 4–10 years	170	28	46	12	15	8	12	5	6	27	36	0.9	1.5	0.4	0.8	15	73	0.2	0.4	66	97	8	13
Boys 11–18 years	129	11	18	4	5	4	5	2	3	10	14	0.5	0.8	0.2	0.4	9	43	0.1	0.2	23	33	5	7
Girls 11–18 years	113	12	19	4	5	3	4	2	2	11	15	0.4	0.7	0.2	0.3	8	38	0.1	0.2	20	29	5	7
Adolescents 11–18 years	121	11	18	4	5	3	5	2	3	11	15	0.5	0.8	0.2	0.3	9	41	0.1	0.2	21	31	5	7
Men 19–64 years	136	16	26	4	5	4	5	2	3	15	21	0.4	0.7	0.3	0.6	9	44	0.1	0.2	20	30	5	8
Women 19–64 years	106	12	21	3	4	3	4	2	2	12	16	0.3	0.5	0.2	0.3	7	34	0.1	0.2	16	23	5	8
Adults 19–64 years	121	14	23	4	5	3	5	2	3	14	19	0.3	0.6	0.2	0.4	8	39	0.1	0.2	18	26	5	8
Men 65+ years	186	22	36	6	7	5	7	3	4	21	29	0.5	0.9	0.4	0.8	13	60	0.1	0.3	28	41	7	10
Women 65+ years	150	17	29	5	6	4	6	2	3	17	23	0.4	0.7	0.3	0.6	10	48	0.1	0.3	22	33	7	11
Adults 65+ years	166	19	32	5	6	5	6	3	4	19	26	0.5	0.8	0.4	0.7	11	54	0.1	0.3	25	36	7	11
Men 65–74 years	154	18	30	5	6	4	6	2	3	17	24	0.4	0.7	0.3	0.6	10	50	0.1	0.2	23	34	5	9
Women 65–74 years	122	14	24	4	5	4	5	2	3	14	19	0.3	0.6	0.3	0.5	8	39	0.1	0.2	18	27	6	9
Adults 65–74 years	137	16	27	4	5	4	5	2	3	16	21	0.4	0.6	0.3	0.6	9	44	0.1	0.2	20	30	6	9
Men 75+ years	214	25	41	7	8	6	8	3	5	24	33	0.6	1.0	0.5	0.9	15	69	0.2	0.3	32	47	8	12
Women 75+ years	177	21	34	6	7	5	7	3	4	20	27	0.5	0.8	0.4	0.7	12	57	0.2	0.3	26	39	8	13
Adults 75+ years	193	22	37	6	7	5	7	3	4	22	30	0.5	0.9	0.4	0.8	13	62	0.2	0.3	29	42	8	12
Pregnant women ^c	106	11	18	3	4	3	4	2	2	12	16	0.2	0.4	0.2	0.3	5	24	0.1	0.1	16	23	4	7
Lactating women ^c	106	11	18	3	4	3	4	2	2	12	16	0.2	0.4	0.2	0.3	5	24	0.1	0.1	16	23	4	6

^a Sources: RNIs Na and Cu (British Nutrition Foundation, 2021), Ca, K, Mg, P, I, Fe and Zn (NDNS, 2020), AIs Mn and Mo (European Food Safety Authority, 2019).^b Estimated based on energy intakes and % of energy intake from milk for the different demographics in the National Diet and Nutrition Survey UK Results from Years 9–11 of the Rolling Programme (NDNS, 2020) and the average caloric content of different milks according to the McCance and Widdowson's 'composition of foods integrated dataset' on the nutrient content of the UK food supply (Public Health England, 2021).^c Assuming a similar milk intake with women 19–64 years of age, and using the average requirement for Ca, AIs for K, Mg, P, Cu, I and Mn intake for pregnant and lactating women, and corrected population RNI for Zn intakes (+1.6 mg/d for pregnant women and +2.9 mg/d for lactating women) from the European Food Safety Authority Panel on Dietetic Products & Allergies (European Food Safety Authority, 2019).

periods (typically November–March (Qin et al., 2021; Stergiadis et al., 2012; Stergiadis et al., 2021)). This finding agrees with previous work at farm level, which also reported that these macrominerals were negatively correlated with pasture and/or total forage intake and positively correlated with maize silage, concentrate feeds and mineral supplementation (Qin et al., 2021; Stergiadis et al., 2021). These all represent typical practices during the indoor/housing period of dairy cows (Qin et al., 2021; Stergiadis et al., 2012; Stergiadis et al., 2021) and may reflect the higher concentrations of these minerals in the milk within the supply chain through to retail.

4.4. Implications of the variation in retail milk mineral profile for the mineral intakes of consumers

To assess the impact of milk mineral variation and whether this is a result of different production systems (conventional, organic, channel island), the intakes of liquid milk for different age groups were calculated from their total energy intakes and the percentages of the energy intake contributed by liquid milk, as recorded by the National Diet and Nutrition Survey (NDNS, 2020) and the average milk energy content in liquid milk according to the McCance and Widdowson's The Composition of Foods integrated dataset (Public Health England, 2021). The daily intakes of macrominerals and trace elements (mg/d) that were significantly different between milk production systems or different months were then calculated by multiplying the milk intakes (g/day) with the measured concentrations of minerals in the present study (mg/g milk). These mineral intakes were then compared with the nutrition recommendations (reference nutrient intakes, RNI; or adequate intakes, AI) for these minerals (European Food Safety Authority, 2019; NDNS, 2020; Public Health England, 2021) to estimate the proportionate contribution of milk from different types and seasons towards the RNI and AI for the different nutrients (Tables 3–5). As this exercise relies on the average intakes of each demographic (NDNS, 2020), it represents on the effect that milk's compositional differences have on the consumers of average liquid milk amounts. The proportionate contribution of milk towards the intakes of certain minerals would be lower for those consuming less milk than the average for their demographic group and higher for those consuming higher amounts.

4.4.1. The role of milk from different production systems (conventional, organic, channel island) in delivering macrominerals and trace elements in consumers' diet

The National Diet and Nutrition Survey has indicated that milk and dairy products are an excellent source of Ca in consumers' diets, delivering 44–59% of Ca intakes in children, 34% of Ca intakes in adolescents and 34–44% of Ca intakes in adults (NDNS, 2020). The NDNS have also reported that milk and dairy products cover 23–32% of P intakes across the different consumer demographics (Henderson et al., 2003; McAlister et al., 2020). Similarly, in the present study milk has been found to be a good source of Ca and P, contributing from 13% to 77% and from 12% to 70% of the respective RNIs in different consumer demographics. In children 1.5–3.0 years of age, consuming CHA milk, instead of ORG or CON milk would increase the contribution of milk towards the RNIs for Ca (from 69% to 77%) and P (from 65% to 70%) which is nutritionally desirable. Ca is hallmarked as critical to bone development and structure and health particularly in infancy and childhood according to Bouziani et al. (2018) and Shertukde, Cahoon, Prado, Cara, and Chung (2022). P also serves as a crucial mineral in a wide range of regulatory, metabolic, and synthesis processes along with bone growth which is required for optimal adolescent and infantile development (Daniels, Hutton, Knott, Wright, & Forman, 1935; Koljonen et al., 2021). However, the relative difference in the contribution of Ca and P to respective RNIs in the other demographics would be rather limited (less \pm 5%) mainly as a result that demographics over 3 years of age drink less milk and have higher requirements for Ca and P. Milk was a reliable source of K and Mg in children 1.5–10 years of age, contributing from 12% to 39% and from

9% to 28% of their respective RNIs; thus aligning with previous reports that highlighted the importance of milk and dairy products in K and Mg intakes in the consumers' diets, representing approximately 16–30% of their intakes in children, 11–13% in adolescents and 9–14% in adults (NDNS, 2020). However, the differences in milk's contribution towards RNIs for K and Mg when these demographics consume ORG, CON or CHA milk were lower than \pm 3% and it is unlikely that these would cause any nutritional or health impacts. In case of female adolescents (reported at 47% below the lower RNI for Mg (NDNS, 2020)), any increase in Mg intake would be desirable.

The National Diet and Nutrition Survey has indicated that milk and dairy products are an excellent source of I in the consumers' diets, delivering 51–64% of I intakes in children, 40% of I intakes in adolescents and 32–41% of I intakes in adults (NDNS, 2020). I is an essential component to thyroid hormone production and is crucial for growth and development (Gunnarsdottir et al., 2012). The results from this study reinforce this finding and under the current recorded milk intakes (NDNS, 2020) and the I concentrations found in the milk in the present study, milk covers up to 115% of the RNI for children 1.5–3.0 years of age, 56% of the RNI for children 4–10 years of age, 31% of the RNI in adolescents and 50% of the RNI in adults. This is particularly important as I deficiency affects 435.5 million people globally and is the most common single cause for preventable brain damage and leads to reduced work and school performance, associated with lower intellectual ability (WHO, 2007). In the UK, particular demographics shown to exhibit substantial rates of I deficiency have been women, and particularly during adolescence, with rates of mild/moderate deficiency ranging from 10% to 51% of the participants across different studies (Miller, Spiro, & Stanner, 2016; Vanderpump et al., 2011). The present study also highlighted that the contribution of milk towards the RNI of pregnant or lactating women (assuming same milk intakes as females 19–64 years of age) is even lower compared with other demographics, because of their lower milk intakes and their higher I requirements. I sufficiency in pregnant or lactating women is critical, as even mild-to-moderate I deficiency has negative neurocognitive implications for the child stemming from maternal I supply while pregnant (Bath & Rayman, 2015). Given the high rates of I sub-optimal iodine status in UK women of childbearing age, and that milk and dairy products are an affordable, readily available, safe, and a reliable dietary staple which is the main source I in most human diets, health professionals should take this into consideration and review the nutritional advice to recommend, where feasible, the consumption of milk and dairy products as a means to overcome I deficiencies (e.g. female adolescents) or meet increased requirements (e.g. pregnant and lactating women).

In relation to the differences in I concentrations between the available milk production systems, switching from CON or ORG milk to CHA milk would reduce the contribution of milk towards RNIs for I from 115% to 77% in children 1.5–3.0 years of age, from 53% to 35% in children 4–10 years of age, from 29% to 20% in adolescents, from 28% to 19% in adults 19–64 years of age, from 45% to 30% in adults 75 + years of age, and from 17% to 12% in pregnant and lactating women (assuming same milk intakes as females 19–64 years of age). Based on the discussions above, this is potentially an undesirable nutritional outcome. However, milk I concentrations are highly correlated to I intakes (Flachowsky, Franke, Meyer, Leiterer, & Schöne, 2014) and increasing the supplementation of cows' diet with I at farm level (within the permitted inclusion rate of 5 mg/kg DM; (Flachowsky et al., 2014)) would potentially improve milk I status in CHA supply chain. This is the first study at retail level in the UK that organic milk has not shown consistently lower concentrations of I than conventional milk, in contrast with other studies which collected samples between 2009 and 2015 (Bath et al., 2012; Payling et al., 2015; Stevenson et al., 2018), aligning with a more recent farm survey that collected samples in 2019 (Qin et al., 2021). Therefore, based on the results of the present work and the recent farm survey (Qin et al., 2021), consumption of ORG and CON milk would not affect I intakes of consumers and there would be no

risk to affect I status of pregnant/lactating women as it has been previously recommended (Payling et al., 2015; Stevenson et al., 2018).

In relation to the differences in Mo concentrations between the available milk production systems, switching from CON or CHA milk to ORG milk would increase the contribution of milk towards RNIs for Mo from 170% to 213% in children 1.5–3.0 years of age, from 71% to 88% in children 4–10 years of age, from 19% to 24% in adults 19–64 years of age, from 31% to 38% in adults 75 + years of age, and from 17% to 21% in pregnant and lactating women (assuming same milk intakes as females 19–64 years of age). This can be considered desirable from a nutritional point of view, as Mo is an essential mineral for proper enzymatic function (EFSA Panel on Dietetic Products Nutrition, and Allergies, 2013). Despite the fact that organic milk contributes substantially more than the Mo requirements to children 1.5–3.0 years of age, this does not pose any health risks as these contributions represent just the 32% of upper tolerable limit (100 µg/d; (European Food Safety Authority, 2019)) for children 1.5–3.0 years of age. It should however be noted that Mo intakes in UK diets are not expected to present risk of deficiencies and Mo is generally abundant in the diet also via plant material (legumes, leafy vegetables, cauliflower, nuts, cereals) (EVM, 2003).

Milk was a good source of Zn in children 1.5–10 years of age, contributing from 9% to 21% RNIs for Zn; less important source of adolescents (5–6% contribution to RNI for Zn), but then again contributions increased from adults from 5% to 11% RNI for Zn. However, the differences in milk's contribution towards Zn's RNI by consuming ORG, CON or CHA milk were lower than $\pm 3\%$ and any nutritional or health implications cannot be assumed. Across all milk production systems and demographics, the contribution of milk towards RNI for Cu and Mn were 0.3–2.9% and 0.1–1.4%, which reveals that milk is not a main source of these minerals in UK diets and their variation is not necessarily relevant to human health implications.

4.4.2. Nutritional implications from the seasonal variation in milk minerals

Milk Ca, Mg, and P concentrations substantially varied between months, potentially as a result in the variation of milk protein concentrations, a compound that all these nutrients are highly correlated with (Dunshea et al., 2019; Gaucheron, 2005; Holt and Jenness, 1984). Consuming milk from the month with the lowest Ca concentration (February) than the highest Ca concentration (December) would reduce the contribution of milk towards Ca RNI from 96% to 58% for children 1.5–3.0 years of age, from 46% to 28% for children 4–10 years of age, from 18% to 11% for adolescents, from 32% to 19% in adults 65 + and from 18% to 11% in pregnant and lactating women (assuming same milk intakes as females 19–64 years of age). For the same months, the contribution of milk towards P RNI would be reduced from 78% to 57% for children 1.5–3.0 years of age, from 36% to 27% for children 4–10 years of age, but the impact for all other demographics would be less than $\pm 4\%$. For the same months for Mg, the impact will only be relevant to children 1.5–3.0 years of age where milk's contribution towards RNI would be reduced from 32% to 23% because the impact to the RNI for the other demographics would be lower than $\pm 4\%$. However, given that adolescents (especially females) have very low intakes of Mg (47% below the lower RNI in previous studies (NDNS, 2020)), even a small increase of Mg in this demographic can be considered nutritionally desirable. This highlights that there might be nutritional implications for the population due to milk macromineral variation, more relevant to Ca (across all demographics) but also for P (particularly for children) and Mg (for children and adolescents). Therefore, maintaining stable milk protein concentration across the year may provide a dual benefit towards improving returns for farmers (in case that payments account for milk solids concentrations) but also maintain a more constant supply of Ca, Mg and P via liquid milk in the population. Actions towards this may include the provision of adequate dietary energy and protein levels in cows' diets, good body condition, and reduced body weight loss via protein feeds and concentrates (especially in the first stage of lactation),

avoidance of overfeeding with oils, an increase of grazing upon clover swards, and the provision of appropriate nutrition during autumn grazing (AHDB, 2022; Murphy & O'Mara, 1993). Despite the seasonal variation in milk K and Na concentrations there would probably be no nutritional and health implications for the consumers as the relative impact to the contribution of milk towards the RNI is less than $\pm 4\%$ across all demographics, except for children 1.5–3.0 years of age which would have a reduction from 42% to 35% their RNI covered from milk by consuming December milk than February milk.

The seasonal variation in milk I concentrations would substantially affect consumers' I intakes. Consuming milk from May (a typical grazing/outdoors month) compared to January (a typical indoor housing month) would reduce milk's contribution towards I RNI from 159% to 34% in children 1.5–3.0 years of age, from 73% to 15% in children 4–10 years of age, from 41% to 9% in adolescents, from 39% to 8% in adults 65 + years of age and from 24% to 5% in pregnant and lactating women (assuming same milk intakes as females 19–64 years of age). It appears that the seasonal variation in milk I concentrations is substantially higher than the variation caused by the different milk production systems; and this may increase the risk of I deficiencies across all consumer demographics, including those at higher risk due to previously documented deficiencies (female adolescents and adults; (Miller et al., 2016; Vanderpump et al., 2011) and/or higher I requirements (pregnant and lactating women; (European Food Safety Authority, 2019)). Given that milk and dairy products are the best single sources of I in UK diets (NDNS, 2020), and the alternative sources are either rarely used (iodised salt) or not consumed in sufficient amounts to cover I requirements (seaweed, fish), it can be recommended that actions are taken in order to improve milk I content in certain months in order to ensure an optimum I supply to the population. Research at farm level, has shown a strong negative correlation between milk I and grazing intake which results in reduced I concentration in milk when cows are grazing (Qin et al., 2021). Milk I concentrations are highly responsive to increased I supply, either provided as I supplements (Flachowsky et al., 2014) or via I-rich feeds (e.g. seaweed; (Qin et al., 2023)), and therefore I supplementation (in any form) should be considered during the months that cows are grazing in order to maintain an optimum I concentration in milk and supply to consumers. In all cases of dietary enrichment of cows' diet with I, special care should be taken to ensure that diet I content does not exceed the upper permitted inclusion rates (5 mg/kg DM; (Flachowsky et al., 2014)).

The seasonal variation in milk Mo concentrations would substantially affect consumers' Mo intakes. Consuming milk from May (a typical grazing/outdoors month) compared to January (a typical indoor housing month) would reduce milk's contribution towards Mo RNI from 234% to 160% in children 1.5–3.0 years of age, from 103% to 70% in children 4–10 years of age, from 31% to 21% in adolescents, from 36% to 25% in adults 65 + years of age and from 23% to 16% in pregnant and lactating women (assuming same milk intakes as females 19–64 years of age). Therefore, the consumption of milk produced during the indoor periods provides more Mo in the diet of consumers. In addition, despite the fact milk contributes substantially more than the Mo requirements to children 1.5–3.0 years of age, this does not pose any health risks as these contributions represent just the 32% of upper tolerable limit (100 µg/d; (European Food Safety Authority, 2019)) for children 1.5–3.0 years of age. Despite the variation, in the absence of current evidence on populations' Mo status (mainly due to absence of suitable biomarkers for Mo status) and the fact that Mo is present in nearly all foods, there is no reason to recommend any interventions in the dairy supply chain in order to reduce the seasonal variation of Mo in milk as this does not pose any risks to population Mo intakes.

Despite the seasonal variation in milk Zn concentrations there would probably be no nutritional and health implications for the consumers as the relative difference to the contribution of milk towards the RNI is less than $\pm 5\%$ across all demographics, except for children 1.5–3.0 years of age which would have a reduction from 26% to 17% their RNI covered from Zn by consuming February milk than August milk. Considering the

average seasonal variation across all milk production systems and demographics, the contribution of milk towards RNI for Cu, Fe, and Mn were 0.2–3.5%, 0.2–1.3% and 0.1–0.7%, which reinforces the finding that milk is not a main source of these minerals in UK diets and their seasonal variation in milk will not affect consumers' nutrition and health.

5. Conclusions

The present study provided evidence that milk is an excellent source of Ca, P, I, and Mo across different consumer demographics, and additionally a very good source of K, Mg, and Zn for children. The work highlighted that both the type of milk (conventional, organic, channel island) and season affect the concentrations of certain macrominerals and trace elements, having in some cases implications to the nutrient supply to consumers' diets. The differences observed were most likely associated with the relationship of certain nutrients to milk protein (e.g. for Ca, P, Mg) and animal diet including mineral supplementation, and intakes of forage, pasture and concentrate feeds (e.g. for Cu, Cu, Fe, I, Mn, Mo and Zn) as these vary between different production systems and seasons. When compared with conventional and organic milk, channel island milk (from Jersey and Guernsey cows) contained more Ca, Mg, P, Cu, Mn, and Zn; but less K and I. Despite the higher concentrations of these macrominerals and Zn in channel island milk, the implications of these differences for consumers' nutrition, under the recorded milk consumption rates in the UK, would be minimal because the numerical values were not different enough to create a meaningful change to the intakes of these macronutrients for consumers. The highest impacts would be for (i) Ca supply to children 1.5–3.0 years of age, where consumption of channel island milk would increase the contribution of milk to Ca and P requirements from 69% and 65% to 77% and 70%, respectively, compared with the consumption of conventional or organic milk, and (ii) for I supply that would be reduced, including demographics which are at higher risk such as adolescents (from 29% to 20%) and pregnant/lactating women (from 17% to 12%; assuming same milk intakes as females 19–64 years of age). However, the impact of the seasonal variation has been more pronounced than that of the type of milk and this may have implications to the intakes of Ca, P, I and Zn; which may be substantially reduced during certain months of the year depending on the requirements and milk intakes of certain demographics. Notably the months with the higher milk concentrations for Ca, P, I and Zn had 1.7, 1.4, 4.7, and 1.6 times more of these minerals, respectively, than the months with the lower concentrations. Notably for I, seasonal variation may cause a reduction of the contribution of milk to RNI from 78–159% to 16–34% in children, from 41% to 9% in adolescents, from 39–62% to 8–13% in adults, and from 24% to 5% in pregnant/lactating women. It may therefore be recommended that mineral supplementation in dairy cows aligns with changes the cows' diets (e.g. supplementation of I and Zn during the grazing season) to minimise the variation in macrominerals and trace elements concentrations in milk and ensure an optimum supply of minerals to the population throughout the year.

CRedit authorship contribution statement

Eric E. Newton: Methodology, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Ásta H. Pétursdóttir:** Methodology, Resources, Writing – review & editing, Project administration. **Stephane Beauclercq:** Methodology, Investigation, Data curation, Writing – review & editing. **James Clarke:** Validation, Investigation, Writing – review & editing. **Natasa Desnica:** Methodology, Validation, Investigation, Writing – review & editing. **Sokratis Stergiadis:** Conceptualization, Methodology, Data curation, Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

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

Data availability

Data will be made available on request from the corresponding author.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.foodchem.2023.135809>.

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