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Macromineral and trace element concentrations and their seasonal variation in milk from organic and conventional dairy herds

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Abstract

To study the effects of dairy production system on milk macromineral and trace element concentrations, milk samples were collected monthly in 2019 from 43 conventional and 27 organic farms. Organic milk contained more Ca (1049.5 vs. 995.8 mg/kg), K (1383.6 vs. 1362.4 mg/kg), P (806.5 vs. 792.5 mg/kg) and Mo (73.3 vs. 60.6 μg/kg) but less Cu (52.4 vs. 60.6 μg/kg), Fe (0.66 vs 2.03 mg/kg), Mn (28.8 vs. 45.0 μg/kg), Zn (4.51 vs. 5.00 mg/kg) and Al (0.32 vs. 1.14 μg/kg) than conventional milk. Significant seasonal variation was observed in all determined minerals' concentrations. Milk I concentration was not consistently affected by production system, whereas organic milk contained less I in June and July than conventional milk. Dietary factors contributing to different milk mineral concentrations between production systems included intakes of maize silage, dry-straights and oils (higher in conventional diets), and pasture, clover and wholecrop (higher in organic diets).

Keywords: dairy cow, milk, organic, minerals, macrominerals, trace elements, heavy metals, iodine, production system

1. Introduction

Organic farming promotes the use of naturally produced substances and processes and limits the application of chemically synthesised ingredients. In organic dairy farming, cows consume more pasture and lower amounts of concentrate feeds and additives when compared with those managed in conventional production systems (Weller & Bowling, 2007). As a consequence of differences in cow nutrition, the chemical composition and nutritional value of organically produced milk, including contents of saturated fat, n-3 fatty acids, trans fatty acids, tocopherols, antioxidants and minerals, may differ from those of conventionally produced milk (Schwendel et al., 2015; Stergiadis et al., 2012).

The difference in mineral composition between organic and conventional milk has been reported based on data from various European countries (Poulsen, Rybicka, Poulsen, Larsen, Andersen, & Larsen, 2015; Rey-Crespo, Miranda, & Lopez-Alonso, 2013). These differences in minerals can be attributed to (i) the mineral status of organically managed cows reflecting local environmental conditions when compared with conventionally managed cows, usually as a consequence of their greater reliance on grazing rather than conserved forage (Orjales, Herrero-Latorre, Miranda, Rey-Crespo, Rodriguez-Bermudez, & Lopez-Alonso, 2018) and (ii) the limited (if any) mineral supplementation to organically managed cattle. These organic farming practices are considered as factors that reduce the concentrations of some essential trace elements in milk, with I and Se being the most commonly reported ones (Srednicka-Tober et al., 2016; van de Kamp, Saridakis, & Verkaik-Kloosterman, 2019).

Organic systems may conversely affect the concentrations of toxic heavy metal elements in milk, as reported previously that organic milk contains less As and Cr, but more Cd than conventional milk (Lopez-Alonso, Rey-Crespo, Herrero-Latorre, & Miranda, 2017; Rodriguez-Bermudez, Lopez-Alonso, Miranda, Fouz, Orjales, & Herrero-Latorre, 2018). The differences in heavy metal concentrations are contributed by the greater ingestion of soil by organically managed cows during grazing when compared with their conventional counterparts (Lopez-Alonso et al., 2017).

In addition, organic milk has been reported to have greater seasonal variation in mineral composition than conventional milk (van de Kamp et al., 2019). The seasonal variation in organic milk mineral composition is attributed to the inconsistency in pasture intake of organically managed cows during a year. Moreover, pasture intake is known to be correlated with the concentrations of a number of minerals in milk, for instance, it increases Ni and reduces Cu and I concentrations (O'Brien et al., 1999). In contrast, the diets of conventionally managed cows may be more homogeneous throughout the year, which results in more consistent mineral composition of milk between seasons.

Since cow milk is an important source of a number of essential nutrients for human, variation in mineral composition between production systems and different seasons potentially impacts mineral supply and balance of consumers. Although data regarding other minerals are scarce, previous works on milk iodine concentration have reported that consumption of organic milk could reduce iodine intake of the UK population, with potential implications particular in sub-groups with higher requirements, e.g., pregnant women (Bath, Button, & Rayman, 2012). Given that I deficiency is observed in 23% of women 11-18 years old and 10% of women 19+ years old (Miller, Spiro, & Stanner, 2016), it was previously claimed that consumption of organic milk may further reduce the chance of these age groups to meet their requirements. However, there has been limited systematic analyses of husbandry practices at a farm level that may influence milk mineral concentrations, as well as implications these might have for human nutrition when taking into account a wider range of minerals from organic and conventional milk.

The objectives of the current study were to (i) compare the macromineral and trace element profiles of organic and conventional milk produced in Southern England and their variation throughout the year, (ii) identify husbandry practices (management, breeding, diet) that impact milk macromineral and trace element concentrations and (iii) estimate the impact that differences between organic and conventional milk macromineral and trace element profiles may have on consumers' mineral intakes. It was hypothesised that the concentrations of specific macrominerals and trace elements differ between organic and conventional milk and the difference in milk mineral composition between production systems mainly results from cow diet and herd breeding parameters.

2. Materials and Methods

2.1 Experiment/survey design

The current study presents results from 809 milk samples collected from 70 dairy herds in Southern England. Twenty-seven of the farms were certified organic according to Soil Association or Organic Farmers and Growers standards, including 11 farms applying all-year round calving, nine farms applying autumn calving, three farms applying spring and autumn calving and four farms applying spring calving. The remaining 43 farms were non-organic (conventional), among which autumn calving, spring and autumn calving and spring calving were applied in one farm, respectively, and all-year round calving was applied in the rest. Although regulations for organic certification may differ between countries across the world, organic dairy diets are generally associated with higher intakes of pasture, forage:concentrate ratios and contribution of legumes in the pastures and conserved forages; with considerable within-system variation being also apparent (Stergiadis et al., 2015; Stergiadis et al., 2012). In the present work, both organic and conventional farms allowed access to outdoors/pasture during the grazing season (March-October), and the average grazing intake throughout the year was 25% of dry matter intake (DMI) in the organic and 8% in the conventional herds, with maximum average across-farms pasture intake being observed in April for organic herds (58%

DMI) and July (16% DMI) for the conventional herds. Over the study, concentrate feeds represented 26% DMI and 39% DMI in the organic and conventional herds, respectively. In conventional production system, pastures and silages were predominantly perennial ryegrass, while in organic systems silage typically has variant grass:clover ratios. Both production systems kept Holstein and non-Holstein breeds, and the latter averaged at 36% and 21% in the organic and conventional herds, respectively. Detailed analysis of the herd composition and feed intake in conventional and organic farms is presented in Table 1.

Milk was collected monthly (3rd week in each month) from farms' bulk tanks following stirring between January and December 2019. Corresponding herd breeding records (proportion of each breed/crossbreed in the herd) and feeding practices (type/amounts of conserved forage, other feeds, supplements offered) were collected during each sampling session, using questionnaires completed collaboratively by each producer and an interviewer. Average herd live weights were estimated based on average breed live weights (supplementary Table S1) and the proportionate number of cows from each breed/crossbreed in the total herd, as described by Stergiadis et al. (2018). Estimated DMI and pasture intake were calculated based on average breed live weight and recorded milk yield, according to Butler et al. (2008).

2.2 Milk analysis

Following collection, milk samples were kept cool and transferred to the laboratory where each sample was aliquoted into three 7 mL containers, one remained refrigerated and two were frozen at -20°C. The chilled aliquot was sent to National Milk Laboratories (Wolverhampton, UK) for analysis of fat, protein and lactose by Fourier Transform InfraRed spectroscopy (MilkoScanTM 7RM; FOSS, Denmark) and somatic cell count (SCC) by flow cytometry (FossomaticTM 7; FOSS, Denmark).

The frozen aliquots were lyophilised at the University of Reading and sent on ice to Matis (Reykjavik, Iceland) for analysis of Al, As, Ca, Cd, Co, Cr, Cu, Fe, Hg, I, K, Mg, Mn, Mo, Na, Ni, P, Pb, Se, Sn amd Zn. The samples were digested using an Ultra-wave Acid Digestion System (Milestone Inc., Italy) and then proceed to macromineral and trace element analyses using inductively coupled plasma mass spectrometry (Agilent 7900, Agilent Technologies, Singapore). The analysis of I was performed according to the method described by Payling et al. (2015). Analyses of Al, As, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Mo, Na, Ni, P, Pb, Se, Sn and Zn were performed according to an NMKL method (NMKL 186, 2007).

2.3 Statistical analysis

Analyses of variance were performed with a mixed linear model (residual maximum likelihood analysis; REML) in GenStat (VSN International, 18^{th} Edition, Hempstead, UK). The fixed effects included production system (Conventional, CON; Organic, ORG), month (January, February, March, April, May, June, July, August, September, October, November, December) and the interaction between them. Farm ID was included as a random effect. In all analyses, normality plots were used to assess the residual diagnostics of the final model. Mineral concentrations and SCC were log(x+1) and log(x) transformed, respectively, prior to REML analysis. Descriptive statistics, to generate means and standard errors, were performed in untransformed data. When the fixed effect was significant ($p < 0.05$), pairwise comparisons of means were performed using Fisher's Least Significant Difference test.

Multivariate redundancy analysis (RDA) was performed to assess the relative impact of breed and diet composition on milk yield, basic composition and macrominerals and trace elements, using CANOCO 5 Windows Release 5.12, with the automatic forward selection of variables and significances calculated using Monte Carlo permutation tests. In the RDA biplots, arrow length and direction represent the relative effects of driver variables (herd breed and diet composition parameters) on the response variables (milk yield, basic composition and mineral profile). The driver related to breed represents the proportion of non-Holstein genetics in the herd (the summary of British Friesian, NZ Friesian, Jersey, Scandinavian Red, Shorthorn, Ayrshire, Brown Swiss, Guernsey and other breed/crossbreed). Drivers related to nutrition were dietary proportions of estimated grazing, grass/grass-clover silage, maize silage, lucerne silage, other mixed silage, wholecrop (ensiled whole wheat plants, harvested approximately 1 month before grain maturity), hay/straw, moist by-products, dry straights, cereals, blends, oil, minerals and vitamins. The response variables included milk yield, basic composition (fat, protein, lactose and SCC) and concentrations of minerals (Al, Ca, Cu, Fe, I, K, Mg, Mn, Mo, Na, P, Sn, Zn).

3. Results

3.1 Herd composition and feed intake

The conventional and organic farms had similar herd compositions, with Holstein being the major breed comprising more than 60% of total cows in both systems (Table 1). There was significant but numerically minor difference between production systems in the proportions of Scandinavian Red ($p = 0.019$) and Shorthorn ($p = 0.002$), being marginally higher in organic farms than conventional farms, although in both cases their contribution to the total herd was <1%.

Estimated daily DMI was higher in conventional farms than in organic farms ($p < 0.001$; Table 1). Significant difference between production systems was also observed in almost all diet composition parameters (expressed as % DMI). Conventionally managed cows had a lower total forage intake ($p < 0.001$) and a higher total concentrate intake ($p < 0.001$) when compared with organically managed cows. Intake of maize silage ($p < 0.001$) was higher in conventional farms than in organic farms. In contrast, intakes of grass/grass-clover silage ($p < 0.001$), other mixed silage ($p < 0.001$) and wholecrop silage ($p < 0.001$) were lower in conventional farms than in organic farms. When compared with conventional farms, organic farms fed cows lower amounts of moist by-products ($p < 0.001$), dry straights ($p < 0.001$), oils ($p < 0.001$) and minerals ($p = 0.007$). In addition, total concentrate intake ($p = 0.004$), grazing intake ($p < 0.001$) and intakes of grass/grass-clover silage ($p < 0.001$) and maize silage ($p < 0.001$) showed significant monthly variation (supplementary Figure S1). Significant interactions between production system and month were observed in grazing ($p < 0.001$), grass/grass-clover silage ($p < 0.001$) and maize silage ($p < 0.001$) intakes (supplementary Figure S1).

3.2 Milk yield and composition

Average milk yield was considerably higher in conventional production system (+7.3 kg/cow/d; $p < 0.001$) with slightly higher milk lactose content (+0.4 g/kg milk; $p = 0.009$) when compared with organic production system (Table 2). Average milk fat, protein and SCC contents were not different between production systems. Significant interactions between production system and month were observed in milk yield ($p = 0.010$) and contents of fat ($p = 0.005$), protein ($p < 0.001$) and lactose ($p < 0.001$; supplementary Figure S2). The within-month comparisons showed that when compared with organic farms, conventional farms had greater milk yields ($p < 0.05$) in all months; higher milk lactose contents ($p < 0.05$) in January, June, July, August and September; a lower milk fat content ($p < 0.05$) in September; and a lower milk protein content ($p < 0.05$) in August.

Across production systems, milk yield and composition fluctuated markedly over time (time effect $p < 0.05$; Table 3). Milk yields in April and May were greater ($p < 0.05$) than in other months. Milk fat and protein contents decreased from January and reached their lowest levels during summer, followed by a gradual increase thereafter. Milk lactose content remained relatively stable from January to June and declined marginally thereafter. Milk SCC was noticeably lower ($p < 0.05$) in February and September when compared with other months.

3.3 Milk macromineral and trace element concentrations

Of the determined macrominerals, organic milk contained higher average concentrations of Ca (+53.7 mg/kg milk, $p < 0.001$), K (+21.2 mg/kg milk, $p = 0.048$) and P (+14.0 mg/kg milk, $p = 0.042$) than conventional milk (Table 2). Significant interactions between production system and month were observed in the concentrations of K ($p < 0.001$) and P ($p < 0.001$; Figure 1). In January, the concentrations of K and P in conventional milk were higher ($p < 0.05$) than those in organic milk, while the opposite ($p < 0.05$) was observed in August, October and November for K and in April and August for P. Regardless of production system, concentrations of Ca, K, Na, Mg and P shared a similar pattern over time (Table 3). Their concentrations increased from January to March and remained stable thereafter except for a sharp decrease in June.

Of the determined essential trace elements, conventional milk contained higher average concentrations of Cu (+8.2 µg/kg milk, $p < 0.001$), Fe (+1.3 mg/kg milk, $p < 0.001$), Mn (+16.2 µg/kg milk, $p < 0.001$) and Zn (0.5 mg/kg milk, $p < 0.001$) when compared with organic milk, while the opposite was observed for milk Mo concentration (-12.7 µg/kg milk, $p < 0.001$; Table 2). Average milk I concentration was not significantly affected by production system. In the analyses of Co and Se, 78% and 84% of the individual measurements were below the limits of quantification (Co, 0.59 µg/kg milk; Se, 35.38 µg/kg milk), respectively, and these results were not included in the statistical analysis. Significant interactions between production system and month were observed in milk concentrations of Cu ($p < 0.001$), Fe ($p < 0.001$), I ($p < 0.001$), Mn ($p < 0.001$), Mo ($p < 0.001$) and Zn ($p < 0.001$; Figure 2). Comparisons within months suggested that conventional milk had a higher ($p < 0.05$) Cu concentration in January, March, May, July and August, but a lower ($p < 0.05$) Cu concentration in February and September, when compared with organic milk. Milk I concentration was higher ($p < 0.05$) in conventional milk than organic milk in June and July. The concentrations of Fe, Mn and Zn were greater (p

< 0.05) in conventional milk than organic milk in January, March, July and August, while conventional milk had a lower ($p < 0.05$) Fe concentration in February and lower ($p < 0.05$) Zn concentrations in February and September when compared with organic milk. In addition, organic farms produced milk containing more ($p < 0.05$) Mo in February, April, June, September, October, November and December, when compared with conventional production system, while the opposite effect ($p < 0.05$) was observed in July and August. Across production systems, milk Cu, Mo and Zn concentrations were higher between January and March when compared with the other months. Their concentrations were relatively stable between April and December except that higher ($p < 0.05$) concentrations were observed in conventional milk than organic milk in July and August (Table 3 and Figure 2). The concentrations of Fe and Mn in milk were lowest between April and June and then peaking in July. Milk I concentration was markedly lower in summer than in winter, presented as a gradual decrease from March to May, before increasing in July, a trend that persisted for the remainder of the year.

Most non-essential trace and toxic heavy metal elements (including As, Cd, Cr, Hg, Ni and Pb) were present in low concentrations and the majority of their individual measurements was below the limits of quantification (As, 5.90 μg/kg milk; Cd, 0.24 μg/kg milk; Cr, 7.08 μg/kg milk; Hg, 2.36 μg/kg milk; Ni, 4.72 μg/kg milk; Pb, 8.26 μg/kg milk): As (99%), Cd (57%), Cr (81%), Hg (100%), Ni (88%), Pb (99%). The results of these elements were thus not included in statistical analysis. Of the accurately quantified elements, conventional milk had a higher ($p < 0.001$) average Al concentration (+0.8 mg/kg milk) than organic milk (Table 2). In addition, the concentrations of Al ($p < 0.006$) and Sn ($p < 0.001$) showed significant interactions between production system and month (Figure 1). The within-month comparisons showed that conventional milk contained more ($p < 0.05$) Al than organic milk in January, March and July. The interaction in Sn concentration was characterized by a higher ($p < 0.05$) Sn concentration in January and March and a lower ($p < 0.05$) Sn concentration in February and July in conventional milk when compared with organic milk. Across production systems, significant time effects were observed in the concentrations of Al ($p < 0.001$) and Sn ($p < 0.001$; Table 3). Milk Al concentration was significantly higher ($p < 0.05$) in January, March and July when compared with other months. The concentration of Sn fluctuated throughout the year and had its minimum between June and September.

3.4 Effect of breeding and diet parameters

The RDA examined relationships between driver variables related to cow breed and diet compositions and response variables including milk yield, composition and mineral concentrations (Figure 3). Drivers explained 7.9% of the variation, of which 4.2% was explained by axis 1 and a further 2.1% was explained by axis 2. Estimated grazing intake and maize silage intake accounted for 3.3% and 1.3% of the total explained variation, while all other feed ingredients and breed individually accounted for less than 1% of the variation. Milk yield and concentrations of lactose, Al, Cu, Fe Mn, Sn and Zn, and to a lesser extent Sn, were positively correlated with intakes of maize silage, dry straights, oils, minerals and hay/straw; and negatively correlated with intakes of pasture, total forage and alternative (non-Holstein) breeds. Milk yield and concentrations of lactose, Al and Fe were also positively correlated with intakes of moist by-products. Milk SCC was positively correlated with intakes of moist by-products, oils, maize silage and pasture and negatively correlated with intakes of grass/grass-clover silage, cereals, wholecrop and to a lesser extent with hay/straw. On the contrary, milk concentrations of protein, Ca, P, K and Na were negatively correlated with intakes of maize silage, dry straights, minerals, oils and moist-by products (Na was not correlated with moist by-products); and positively correlated with grazing and total forage intakes and the proportion of alternative (non-Holstein) breeds. Milk Ca content was also positively correlated with wholecrop, cereals and grass/grass-clover silage intakes. Concentration of I was negatively correlated with grazing and total forage intakes, as well as alternative (non-Holstein breeds) and positively correlated with intakes of grass/grass-clover silage, cereals, wholecrop, dry straights and minerals, and to a small extent with hay/straw. Milk concentrations of fat and Mo were positively correlated with intakes of cereals, grass/grass-clover silage, wholecrop, total forage and the proportion of alternative (non-Holstein) breed, while Mg was positively correlated only with intakes of cereals, grass/grass-clover silage and wholecrop. Milk contents of fat and Mo were negatively correlated with intakes of moist by-products, oils, maize silage minerals and dry straights.

4. Discussion

4.1 Milk yield and basic composition

4.1.1 Effects of production system on milk yield and basic composition

Organic farms produced 25% less milk per cow than conventional farms. Decreased milk yield in organic production system has been reported a number of times in previous studies (Stergiadis et al., 2015; Stergiadis et al., 2012), in which 15-30% less milk production was observed in organic farms than conventional farms. The reduced milk production by the organic farms has been mainly associated with the lower intake of concentrates and higher intake of pasture, thus resulting in a lower energy density diet (Schwendel et al., 2015). The reduction of diet energy also gives rise to the necessity to use alternative breeds to Holstein (Stergiadis et al., 2015; Stergiadis et al., 2012). In the present study, total concentrate intake was considerably lower in organic farms than in conventional farms, while pasture and forage intakes were higher. The results of the RDA confirmed the strong negative correlation of milk yield with total forage and grazing intakes as well as the proportion of alternative (non-Holstein) breeds, and the positive correlation with intakes of maize silage and some concentrate ingredients (in particular oils, dry straights, moist by-products and minerals), with some of these findings previously demonstrated (Stergiadis et al., 2012). In addition, previous work reported that milk yield in organic farms may be also limited by farmers in order to reduce production intensity and the incidence of mastitis (Valle, Lien, Flaten, Koesling, & Ebbesvik, 2007).

Milk fat content was not consistently affected by production system, although there was a higher fat content in organic milk when compared with conventional milk in September. Previously, contradictory results from the comparison of milk fat content between different production systems have been reported (Butler et al., 2008; Sundberg, Berglund, Rydhmer, & Strandberg, 2009). Schwendel et al. (2015) suggested that a higher fat content in organic than conventional milk can be expected because the greater intakes of starch-based concentrates in conventional farms may reduce milk fat synthesis. In addition, findings of the RDA and other studies suggest that the higher contribution of alternative breeds in the herd may also contribute to the higher fat content of organic milk (Palladino, Buckley, Prendiville, Murphy, Callan, & Kenny, 2010; Stergiadis et al., 2015). In the present study, the difference in concentrate intake between production systems was greater during the grazing season (April – September) compared with other months, which may explain the higher milk fat content of organic milk at the end of the grazing season, while the potential impact of breed would be minimal due to the relatively similar breed make-up of the conventional and organic herds.

The influence of production system on milk protein content was also minor, as only characterized by a higher protein content of organic milk in August. Milk protein content was suggested to be positively associated with intake of starch-based concentrate (Rius et al., 2010) and higher protein contents of conventional milk than organic milk have been previously reported by others (Sundberg et al., 2009). However, similar results were not observed in the present study even though a considerably higher level of concentrate was involved in the diets of conventional managed cows compared with those of their organic counterparts. The similar breed make-up of conventional and organic herds may also explain the similarities in milk protein content, as breed is also a strong driver for milk protein content, according to previous studies (Palladino et al., 2010; Stergiadis et al., 2015) and the RDA results in the current study. Milk lactose content was marginally lower in organic production system when compared with conventional system. The reduction of milk lactose content in organic milk occurred mainly during the grazing season (June – September). Previously, a lower lactose content in organic milk than in conventional milk has been reported by others (Tunick, Van Hekken, Paul, Ingham, & Karreman, 2016). As milk lactose content has been reported to be increased by infusion of starch to cows (Rius et al., 2010), the reduction in organic milk lactose content during the grazing season may be explained by the enlarged difference in concentrate intake between production systems (higher in conventional) during this period. The RDA revealed positive correlations between intakes of maize silage, oils, dry straights and minerals and milk lactose concentration. Therefore, the lower intakes of these feeds in organic herds may have also contributed to the slightly lower lactose content of milk.

4.1.2 Seasonal variation of milk composition

Across production systems, milk fat and protein contents showed a similar pattern over time; their concentrations were higher in winter and lower in summer. Previously, the reduction of milk fat and protein contents from winter to summer have been reported by others (Bernabucci et al., 2015; Heck, van Valenberg, Dijkstra, & van Hooijdonk, 2009). The seasonal variation in milk fat and protein contents is likely associated with grazing, as a previous study showed that milk fat decreased linearly with increased fresh grass intake (Couvreur, Hurtaud, Lopez, Delaby, & Peyraud, 2006). The lipid in fresh pasture contains a relatively high level of long-chain unsaturated fatty acids, exemplified by linolenic acid, as potential inhibitors of milk fat synthesis (Bauman & Griinari, 2001; Couvreur et al., 2006). The reduction in milk protein content in summer may be attributed to the decreased propionate production in the rumen as a consequence of the higher forage:concentrate ratio in the diet during the grazing season (Bannink et al., 2006). Milk lactose content in both production systems also dropped when cows were provided with increasing amounts of fresh pasture, whereas the minimal level appeared at late grazing season. The seasonal variation of lactose was greater in the organic than in the conventional production system, which was likely associated with the different dietary starch levels between production systems (Rius et al., 2010), as concentrate intake was kept relatively stable during the year in coventional farms, whereas it decreased to a lower level during the grazing season in organic farms, reaching the minimum in August when the lowest milk lactose content was also observed.

4.2 Milk macromineral and trace element concentrations

4.2.1 Effects of production system on milk mineral and trace element concentrations

The concentrations of Ca and P were higher in organic milk than conventional milk. Our results disagree with the findings of Poulsen et al. (2015) who compared the mineral compositions of bulk milk from two conventional dairies and one organic dairy in Denmark and reported lower Ca and P concentrations in organic milk. Moreover, a meta-analysis investigating the mineral profile of bovine raw milk based on 72 different studies from 37 countries showed that Ca and P concentrations were lower in organic milk when compared with conventional milk, although differences between countries were acknowledged as well (Zwierzchowski & Ametaj, 2018). Poulsen et al. (2015) suggested a positive correlation between milk P concentration and protein content. However, this explanation is not applicable in the present study as protein contents were generally similar in both conventional and organic milk throughout the year.

Milk K concentration was observed higher in organic milk than conventional milk. However, our result is contrary to the finding of the meta-analysis by Zwierzchowski et al. (2018), in which it was reported that conventional milk tended to contain more K than organic milk due to the excessive use of K-rich fertilizer in conventional swards. This may not necessarily be the case in the present study which represented the organic and conventional systems in southern England.

The concentrations of Ca and P in milk are relatively constant and are mainly determined by genetics (van Hulzen, Sprong, van der Meer, & van Arendonk, 2009). The contradiction between our results and others' might be explained by differences in breed make-up of the herds, which is supported by the strong positive correlations of Ca, P and K concentrations with alternative (non-Holstein) breeds in the RDA results. Regarding the effects of diet, the RDA results suggested that Ca, P and K concentrations were positively correlated with grazing and total forage intakes, while Ca concentration was also positively correlated with wholecrop, cereals and grass/grass-clover silage intakes. In addition, their concentrations were negatively correlated with intakes of maize silage, dry-straights, minerals, moist by-products and oils. Moreover, a previous finding suggested that maize silage has low Ca and P concentrations and supplementation of these minerals is required when maize silage comprises 25% or more of DMI (Kolver, Roche, Miller, & Densley, 2001). Therefore, the considerably higher proportion of maize silage in conventional diets when compared with organic diets (24.8% DMI vs. 0.9% DMI) could be an important driver of the higher Ca and P concentrations in organic milk. In addition, the higher Ca, P and K concentrations in organic milk may have been partly driven by the numerically higher contribution of grazing, wholecrop, grass/grass-clover silage and cereals to organic diets, as well as the extremely low intakes of dry-straights (<1% DMI), moist by-products (<1% DMI) and oils (non-fed). The fact that Ca and P contents in milk may represent the combined effect of cow diet and breed has been previously reported by Zwierzchowski et al. (2018).

Milk concentrations of five essential trace elements, including Cu, Fe, Mn, Mo and Zn, were different between production systems. The average concentrations of Cu, Fe, Mn and Zn during the year were lower in organic milk. These results correspond to the findings of several studies in which the profiles of essential trace elements of conventionally and organically produced milk were compared (Rey-Crespo et al., 2013; Rodriguez-Bermudez et al., 2018; Zwierzchowski et al., 2018). The lower concentrations of these essential trace elements in organic milk may have resulted from the lower concentrate intake in organic herds, because concentrates typically contain higher levels of Co, Cu, I, Se and Zn than forages due to the routine supplementation of these minerals (Orjales et al., 2018). Previous analyses of mineral contents of organic and conventional diets have shown that conventional diets contained higher concentrations of Cu and Zn (Lopez-Alonso et al., 2017; Orjales et al., 2018). The RDA results of the current study provide further evidence on the potential reasons for this observation, showing that concentrations of Cu, Fe, Mn and Zn were mainly driven by intakes of maize silage, dry-straights, moist by products, minerals and oils (all higher in conventional herds) and were negatively correlated with grazing and total forage intakes and the proportion of non-Holstein breed in the herd (all higher in organic herds). Previous findings also highlighted the influence of cow breed on the concentrations of Fe, Mn and Zn in milk (Zwierzchowski et al., 2018).

Conversely, the average Mo concentration was higher in organic milk than conventional milk. The higher Mo concentration in organic milk may, again, be associated with higher dietary Mo intake, as a previous study has reported higher Mo concentrations in the diet and blood of organically managed cows when compared with conventionally managed cows (Lopez-Alonso et al., 2017). The RDA results in the present study confirmed the previous findings that both diet and breed may influence milk Mo concentration (Zwierzchowski et al., 2018). The higher Mo content in organic milk may be explained by the higher contribution of alternative (non-Holstein) breeds, higher intakes of wholecrop, grass/grass-clover silage and cereals, and lower intakes of maize silage, dry straights, moist by-products and oils, according to the RDA results.

The concentrations of forementioned essential trace elements in milk shared common patterns related to the interaction between production system and month, i.e., similar differences between production systems were observed in specific months. For instance, conventional milk contained higher levels of Cu, Fe, Mn, Mo and Zn in July and August (when differences in grazing intake and forage: concentrate ratio between the systems were maximised), higher levels of Cu, Fe, Mn and Zn in January and March, lower levels of Cu, Mn, Mo and Zn in February and lower levels of Cu, Mo and Zn in September. Given that genetics in the herds were stable throughout the year, these patterns potentially reflect changes in dietary supply of these minerals via various dietary changes.

Previous studies have also summarised other potential parameters that influence the concentrations of these minerals, such as water quality and composition, Cu originating from infrastructure, distance from industrial facilities, negative interactions/correlations between minerals (e.g. Cu vs Zn), skimming of milk (Zwierzchowski et al., 2018). However, data in relation to these additional parameters were not available in the present study.

The average milk I concentrations were similar between production systems. A decreased I concentration has been commonly observed in organic milk when compared with conventional milk (Bath et al., 2012; Srednicka-Tober et al., 2016; Zwierzchowski et al., 2018). The factors that may decrease I concentration in organic milk have been attributed to less use of iodophor sanitizer, lower intake of mineral-supplemented concentrates and higher intake of goitrogen-enriched forage, which hampers iodine absorption and transfer into the mammary gland and milk (Flachowsky, Franke, Meyer, Leiterer, & Schone, 2014; Rey-Crespo et al., 2013). Despite the absence of a difference in the average concentration, organic milk had a lower I concentration than conventional milk in June and July when the difference in grazing and concentrate intakes was almost maximised between production systems. Therefore, the lower I concentration in organic milk in June and July may have resulted from (i) greater grazing intake, which is known to reduce milk I content (Flachowsky et al., 2014) and also confirmed by their strong negative correlation in the RDA of this study and (ii) potential higher ingestion of clover-containing pastures used in organic farming, which contain greater quantities of goitrogenic compounds than perennial ryegrass pastures used in conventional farming (Flachowsky et al., 2014).

Of the determined non-essential trace and toxic heavy metal elements that were accurately quantified in this study, only the average concentration of Al showed a significant effect of production system. The higher content of Al in conventional milk is in line with a previous study which showed that Al concentration in conventional milk was 6.5-fold greater than that in organic milk (Zwierzchowski et al., 2018). Differences in soil acidity and water quality/process may influence Al content in milk (Zwierzchowski et al., 2018) but such records were not available in the present study. In addition, the concentrations of Al and Sn showed similar patterns related to the interaction between production system and month as the forementioned essential trace elements did, as a higher Al concentration was observed in January, March and July, and a higher Sn concentration was observed in January and March in conventional milk. The RDA results indicated that potential drivers for higher milk Al and Sn concentrations can be intakes of maize silage, dry-straights, minerals, moist by-products and oils (used more extensively in conventional farms), while their concentrations were negatively correlated with grazing and total forage intakes as well as the proportion of alternative breeds. The concentration of heavy metal elements in milk largely depends on the environment and the use of fertilizer (Lopez-Alonso, 2012). The difference in the concentrations of As, Cd, Cr and Pb between conventional and organic milk has been reported by others (Rodriguez-Bermudez et al., 2018; Zwierzchowski et al., 2018), although most measured values for these elements were below the quantification limit of analysis in the present study. In general, the present study suggested that concentrations of heavy metal elements were very low in both types of milk and their concentrations were well below the tolerable intake values (Vracko, Tuomisto, Grad, & Kunseler, 2007).

4.2.2 Seasonal variation of milk macromineral and trace element concentrations

The concentrations of all determined minerals varied significantly over time. Although the concentrations of macrominerals showed similar patterns over sequential months, no obvious pattern over seasons was observed. Most determined essential trace elements (except Fe) had a relatively higher concentration in milk between January and March than the rest of the year. Their profiles over months may correspond to the previous finding that milk essential trace element concentrations are generally higher during winter than summer due to higher concentrate intake and lower grazing intake (Rey-Crespo et al., 2013). The RDA results confirmed the previous findings, as grazing intake was identified as the main driver for the concentrations of many milk minerals (except Ca, K, P, Mg and Na, on which the impact is rather low). Milk I concentration showed an obvious decrease with the increase of grazing intake in summer, which is also supported by the negative correlation between them in the RDA. The reduction of milk I concentration during the grazing season has been prevalently reported and has been attributed to decreased I intake and increased goitrogen ingestion (Flachowsky et al., 2014; Pennington, 1990). The seasonal variation of I concentration was more marked in organic milk than conventional milk, in line with the findings of previous works which compared I concentrations in retail milk between summer and winter (Stevenson, Drake, & Givens, 2018; van de Kamp et al., 2019). The greater seasonal variation of I concentration in organic milk may have resulted from the greater change in grazing intake during the year in organic farms when compared with conventional farms.

4.2.3 Potential impact of dairy production system on the mineral intakes of UK consumers

Milk is a major source of various minerals (notably Ca, I, Mg), particularly for age groups that may have increased requirements, such as pregnant women and nursing mothers (Scholz-Ahrens, Ahrens, & Barth, 2020). Given the differences in mineral composition between conventional and organic milk, the choice of consuming one or the other may impact the mineral intake of consumers. To assess the impact of dairy production system on the mineral intakes, the intakes of liquid milk for different age groups in the UK population 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 (Roberts et al., 2018) and the average milk energy content in liquid milk (Pinchen, Powell, Weiner, & Finglas, 2019). The daily intakes of macrominerals and essential trace elements that had significantly different average concentrations between production systems (Ca, K, P, Cu, Fe, Mn, Mo and Zn) were then calculated based on milk intakes and the quantified concentrations in the present study and were compared with the nutrition recommendations (Roberts et al., 2018).

Depending on the intake, milk provides all age groups with an essential part of the reference nutrient intake (RNI) for macrominerals Ca (34%-75%), K (20%-43%) and P (35%-75%). The greatest contribution of milk to Ca, K and P intakes is seen for children 1.5-3 years old whose diet relies more on milk than that of other age groups. Substituting conventional with organic milk in children 1.5-3.0 years old will slightly increase the contribution towards RNIs of Ca (from 70.6% to 74.5%; +13.4 mg/day), K (from 42.3% to 42.9%; +5.3 mg/day) and P (from 72.9% to 74.2%; +3.4 mg/day). Similar increased contribution of milk are observed in the other age and gender groups, but given that children 1.5-3.0 years old have the greatest reliance on milk intake as well as the lowest RNIs of minerals among the age groups, the corresponding impact on the other age and gender groups would be at an even lower scale. Although milk is the main supplier of Ca, K and P in human diets and concentrations of these minerals in milk are influenced by dairy production system, it is unlikely that substituting conventional with organic milk would have any effect on human health because of the relatively low impact towards changing the contribution to RNIs.

As regards to the essential trace elements which were affected by dairy production system, milk (regardless of production system) provides with a proportion of RNI for Cu (1.6-3.8%), Fe (1.2-7.3%), Mn (0.3-0.9%), Mo (0.04-0.11%) and Zn (10.9-24.8%). Taking as an example of the age group children 1.5-3.0 years old that receives the highest potential impact due to high milk intake and low RNI, substituting conventional with organic milk would reduce the intakes of Cu (from 3.8% to 3.3% RNI; -2.1 μg/day), Fe (from 7.3% to 2.4% RNI; -0.34 mg/day), Mn (from 0.9% to 0.6% RNI; -4.0 μg /day) and Zn (from 24.8% to 22.4% RNI; -0.12 mg/day) and increase the intake of Mo (from 0.09% to 0.11% RNI; +3.16 μg/day). Similar to the macrominerals, differences in intakes of essential trace elements from milk appear to be too small to impact human health.

Iodine concentration of organic milk has often been reported to be lower than that of conventional milk and previous studies highlighted that certain consumer groups of organic milk with high I requirements (e.g. nursing mothers and pregnant women) may need to consume higher quantities of milk or alternative food sources of iodine to meet their nutritional requirements (Bath et al., 2012; Stevenson et al., 2018). In this study, although there was no significant overall difference in I content between the two types of milk, organic milk contained less I than conventional milk during two months of the grazing season (278 μg/kg vs. 316 μg/kg in June; 248 μg/kg vs. 269 μg/kg in July). For children 1.5-3.0 years old, switching from conventional milk to organic milk would reduce I intake from 108.8% to 95.6% RNI (-9.3 μg/day). In case of pregnant women and nursing mothers, for whom RNI of I is 250 μg/d (Andersson, de Benoist, Delange, Zupan, & Secretariat, 2007), assuming average milk intake similar to women 19-65 years old, consuming organic milk would reduce I intake from 30.5% to 26.8% RNI (-13.2 μg/day). The potential impact on nutrition is therefore lower than reported in other studies (Bath et al., 2012; Stevenson et al., 2018). This is possibly because the conventional systems in the present study also allowed their cows to graze in summer (supplementary Figure S1). This may have reduced the difference in pasture intakes between the two production systems, subsequently reducing the potential difference in I intakes based on organic or conventional milk because pasture intake was seen as a major driver for milk I content in the RDA. Notably, I contents in conventional and organic milk were decreased, e.g. from 426 μg/kg and 485 μg/kg in March to 269 μg/kg and 248 μg/kg in July, respectively. The variation over months may have a greater impact on consumers' I intake than dairy production system does.

5. Conclusions

The organic farms in the present study had lower average daily milk yield per cow and milk lactose content when compared with the conventional farms, while milk fat and protein contents during late summer were higher in organic milk than in conventional milk. Across production systems, milk protein, fat, lactose contents were lower during the grazing season. Organic milk contained higher levels of macrominerals Ca, K, P when compared with conventional milk. In contrast, with respect to essential trace elements, organic milk contained on average less Cu, Fe, Mn and Zn but more Mo than conventional milk. Milk I concentration was lower in organic milk in June and July when compared with conventional milk. The concentrations of all determined elements varied significantly over months with higher concentrations of Cu, I, Mn, Mo and Zn observed between January and March compared with the other months. Differences in milk mineral concentrations between organic and conventional systems may be explained by the different dietary ingredients making up conventional and organic dairy cow diets, including intakes of maize silage, moist-by products, dry-straights, minerals and oils (higher in conventional cow diets) and intakes of total forage, pasture, grass/grass-clover silage and wholecrop (higher in organic farms). In addition, the multivariate redundancy analysis suggested a secondary effect of cow breed, in particular the higher contribution of alternative breeds and crossbreeds in organic herds in expense of Holstein. The magnitude of the differences in milk mineral composition appeared to have a rather small impact on the expected contribution of milk to the reference nutrient intakes, according to recorded milk intakes of the UK population. Therefore, the differences in milk mineral composition are likely not large enough to have any meaningful impact on human health.

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Table 1. Herd composition and feed intake in conventional and organic farms.

Parameters assessed	Mean[1]		SE	*p*-value
	Conventional n = 490	Organic n = 319		
Herd composition (% herd)				
Holstein	78.9	64.2	6.64	0.125
British Friesian	4.5	2.3	2.33	0.510
Ayrshire	2.5	7.8	3.46	0.279
New Zealand Friesian	0.4	1.3	0.75	0.421
Jersey	0.3	2.1	0.83	0.129
Scandinavian Red	0.1	0.9	0.22	0.019
Shorthorn	0	0.7	0.15	0.002
Montbeliarde	0.1	0.1	0.05	0.905
Brown Swiss	0.3	1.8	0.84	0.212
Guernsey	0.1	0	0.03	0.285
Other breed/crossbreed	12.3	18.8	5.81	0.435
Feed intake and composition (% dry matter intake unless otherwise stated)				
Dry matter intake (kg/cow/d)	20.0	18.6	0.27	<0.001
Total forage	61.0	74.3	1.22	<0.001
Total concentrate	39.0	25.7	1.22	<0.001
Grazing intake	8.3	25.4	1.86	<0.001
Grass/grass-clover silage[2]	24.4	35.1	1.63	<0.001
Maize silage	24.8	0.9	1.44	<0.001
Lucerne silage	0.4	0.1	0.20	0.307
Other mixed silage	0.3	2.6	0.45	<0.001
Wholecrop silage	0.4	8.0	0.57	<0.001
Hay and straw	2.5	2.2	0.39	0.631
Moist by-product	4.8	0.4	0.77	<0.001
Dry-straight	10.0	0.9	1.62	<0.001
Cereal	2.4	4.0	0.64	0.073
Blend	20.9	20.4	1.92	0.874
Oil	0.8	0	0.11	<0.001
Mineral (g/cow/d)	131.1	68.8	15.64	0.007
Vitamin (g/cow/d)	10.3	0	5.81	0.215

[1]Predicted means from the fitted mixed linear model.

[2]In the conventional production system, this was predominantly perennial ryegrass silage, while in organic systems silage typically has variant grass: clover ratios.

Table 2. Milk yield, basic composition and macromineral and trace element concentrations in conventional and organic farms

Parameters assessed	Mean[1]		SE	*p*-value[2]
	Conventional n = 484	Organic n = 316		
Yield and basic composition				
Milk yield (kg/cow/d)	29.5	22.2	0.67	<0.001
Fat (g/kg milk)	39.2	39.8	0.30	0.193
Protein (g/kg milk)	33.4	33.3	0.21	0.560
Lactose (g/kg milk)	45.2	44.8	0.09	0.009
SCC (×1000/ml milk)	149.9	136.0	2.91	0.102
Macrominerals (mg/kg milk)				
Ca	995.8	1049.5	5.06	<0.001
K	1362.4	1383.6	7.03	0.048
Mg	95.7	95.0	0.53	0.457
Na	343.5	344.7	2.86	0.906
P	792.5	806.5	4.45	0.042
Essential trace elements (µg/kg milk unless otherwise stated)				
Cu	60.6	52.4	1.84	<0.001
Fe (mg/kg milk)	2.03	0.66	0.222	<0.001
I	331.2	364.1	15.99	0.547
Mn	45.0	28.8	2.57	<0.001
Mo	60.6	73.3	1.72	<0.001
Zn (mg/kg milk)	5.00	4.51	0.114	<0.001
Non-essential trace elements				
Al (mg/kg milk)	1.14	0.32	0.120	<0.001
Sn (µg/kg milk)	2.66	2.68	0.300	0.166

[1]The means of milk yield, fat, protein and lactose contents are the predicted means obtained from the fitted mixed linear model. The means of somatic cell count (SCC), macrominerals and trace elements are the arithmetic means of the measured values.

[2]The *p*-value of SCC was obtained from the fitted mixed linear model based on log(x)-transformed values. The *p*-values of macrominerals and trace elements were obtained from the fitted mixed linear model based on the log(x+1)-transformed values.

Table 3. Changes of average milk yield, basic composition and macromineral and trace element concentrations of all farms over time.

Parameter	Mean[1]												SE	*p*-value[2]
	Jan n = 67	Feb n = 69	Mar n = 68	Apr n = 68	May n = 68	Jun n = 68	Jul n = 68	Aug n = 67	Sep n = 67	Oct n = 67	Nov n = 67	Dec n = 65		
Yield and basic composition														
Milk yield (kg/cow/d)	26.0[cd]	25.7[cd]	25.8[cd]	26.9[ab]	27.1[a]	26.1[bc]	25.8[cd]	25.3[cd]	25.2[cd]	25.1[d]	25.3[cd]	25.9[cd]	0.57	<0.001
Fat (g/kg milk)	40.6[bc]	39.5[d]	39.7[d]	38.6[e]	37.7[f]	38.3[ef]	36.8[g]	39.5[d]	40.3[cd]	41.1[ab]	41.7[a]	40.8[bc]	0.34	<0.001
Protein (g/kg milk)	33.3[c]	33.2[cd]	32.8[e]	33.1[cde]	32.9[de]	32.8[ef]	32.4[f]	33.2[cd]	33.5[c]	34.1[b]	34.7[a]	34.1[b]	0.20	<0.001
Lactose (g/kg milk)	45.3[b]	45.2[bc]	45.5[a]	45.3[b]	45.2[bc]	45.0[cd]	44.8[e]	44.5[f]	44.6[f]	44.8[e]	45.0[de]	45.0[de]	0.09	<0.001
SCC (×1000/ml milk)	147.6[bcd]	127.7[ef]	155.6[abc]	150.6[abcd]	141.3[cde]	164.9[a]	148.1[abcd]	159.7[ab]	114.4[f]	142.9[cde]	142.4[cde]	137.7[de]	6.61	<0.001
Macrominerals (mg/kg milk)														
Ca	960.3[e]	983.1[cde]	1061.2[b]	976.3[de]	986.7[cd]	925.9[f]	1058.8[b]	1006.3[c]	1069.3[b]	1094.2[a]	1039.8[b]	1042.0[b]	10.56	<0.001
K	1217.4[i]	1328.9[g]	1401.4[cde]	1345.8[fg]	1375.1[ef]	1256.3[h]	1463.0[ab]	1378.4[def]	1435.2[bc]	1468.0[a]	1363.2[ef]	1417.4[cd]	13.41	<0.001
Mg	88.0[f]	91.2[de]	100.9[b]	90.2[ef]	93.5[cd]	85.3[g]	99.1[b]	95.1[c]	101.4[b]	104.7[a]	102.1[ab]	93.9[cd]	1.02	<0.001
Na	275.2[h]	318.0[g]	380.6[b]	354.9[cd]	330.2[f]	309.7[g]	410.2[a]	336.2[ef]	353.2[d]	364.3[c]	348.1[de]	348.1[de]	5.26	<0.001
P	668.8[h]	778.6[f]	842.6[abc]	812.8[d]	820.7[bcd]	732.5[g]	841.6[ab]	783.7[ef]	813.4[de]	819.8[cd]	803.0[de]	860.5[a]	8.33	<0.001
Essential trace elements (μg/kg milk unless otherwise stated)														
Cu	67.8[cd]	71.3[b]	112.1[a]	50.3[ef]	42.0[hi]	40.3[i]	56.6[efgh]	65.0[c]	41.8[ghi]	44.0[fghi]	50.9[de]	46.6[efg]	3.23	<0.001
Fe (mg/kg milk)	4.77[a]	0.86[c]	2.14[bc]	0.42[ef]	0.21[f]	0.26[ef]	6.34[ab]	1.27[cd]	0.30[ef]	0.25[ef]	0.75[de]	0.33[ef]	0.371	<0.001
I	426.2[a]	382.8[ab]	449.7[a]	338.8[de]	270.6[fg]	300.8[ef]	260.9[g]	297.8[ef]	334.0[cd]	396.2[ab]	349.8[bc]	323.1[cd]	35.46	<0.001
Mn	77.9[b]	44.2[bc]	80.2[a]	24.4[e]	18.9[f]	19.8[ef]	73.2[cd]	42.5[d]	20.3[ef]	19.0[f]	22.9[ef]	19.5[ef]	4.85	<0.001
Mo	63.3[cd]	82.3[b]	105.7[a]	57.3[cd]	54.3[d]	53.3[d]	60.7[d]	79.7[b]	52.7[d]	55.9[d]	64.0[c]	58.4[cd]	3.18	<0.001
Zn (mg/kg milk)	4.98d	5.86[b]	8.21[a]	4.12[def]	3.72[g]	4.36[d]	4.88[de]	5.84[c]	3.91[defg]	3.79[fg]	4.16[def]	3.86[efg]	0.192	<0.001
Non-essential trace elements														
Al (mg/kg milk)	2.92[a]	0.52[cd]	2.42[b]	0.26[ef]	0.06[f]	0.09[f]	2.24[bc]	0.50[de]	0.18[ef]	0.13[ef]	0.31[def]	0.13[ef]	0.234	<0.001
Sn (μg/kg milk)	3.37[bcd]	2.24[bcd]	4.18[a]	2.08[cde]	2.96[bc]	1.54[ef]	3.87[ef]	2.28[cde]	1.81[f]	1.72[ef]	1.67[de]	4.33[abc]	0.532	<0.001

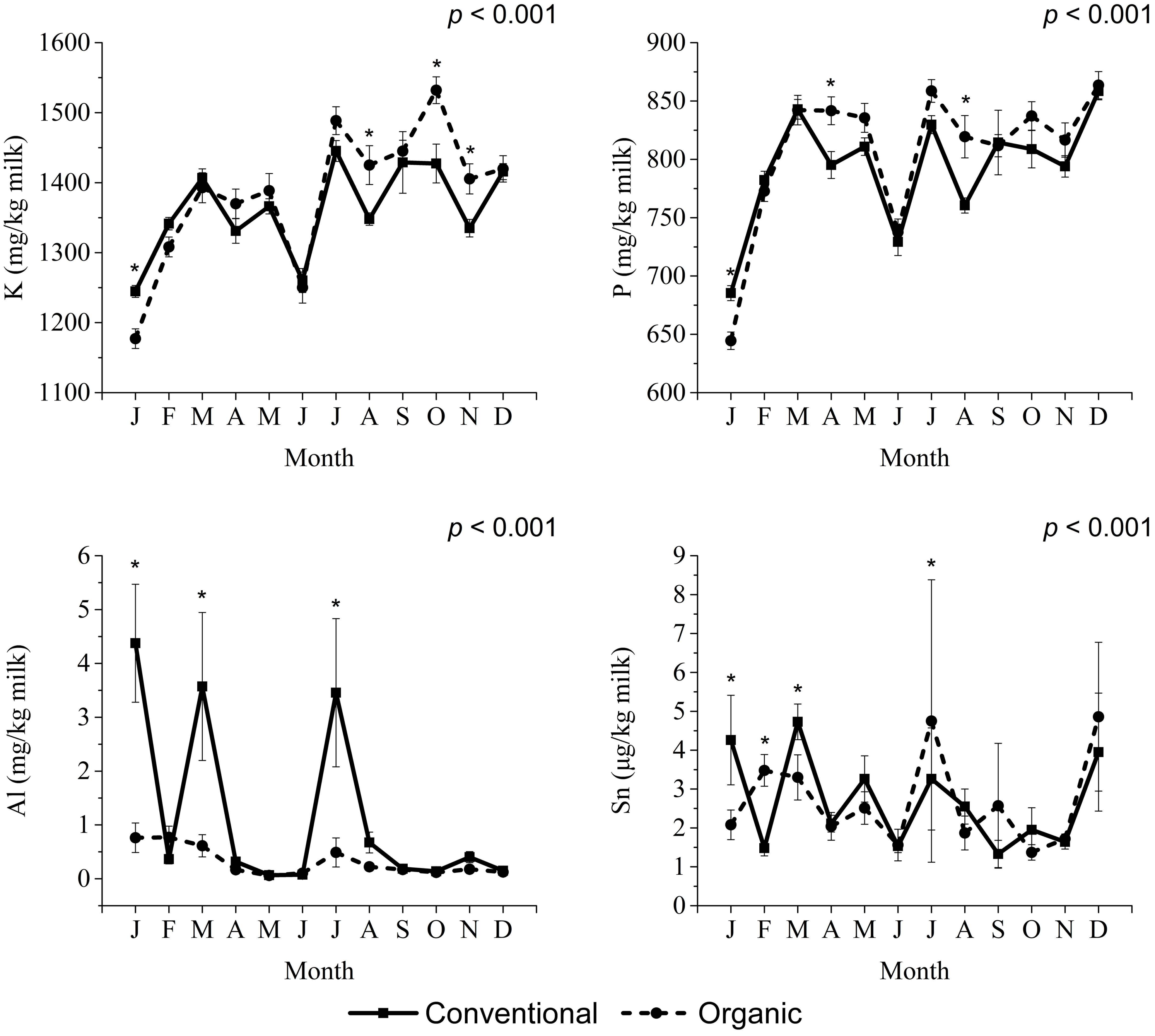
[1]The means of milk yield, fat, protein and lactose contents are the predicted means obtained from the fitted mixed linear model. The means of somatic cell count (SCC), macrominerals and trace elements are the arithmetic means of the measured values.

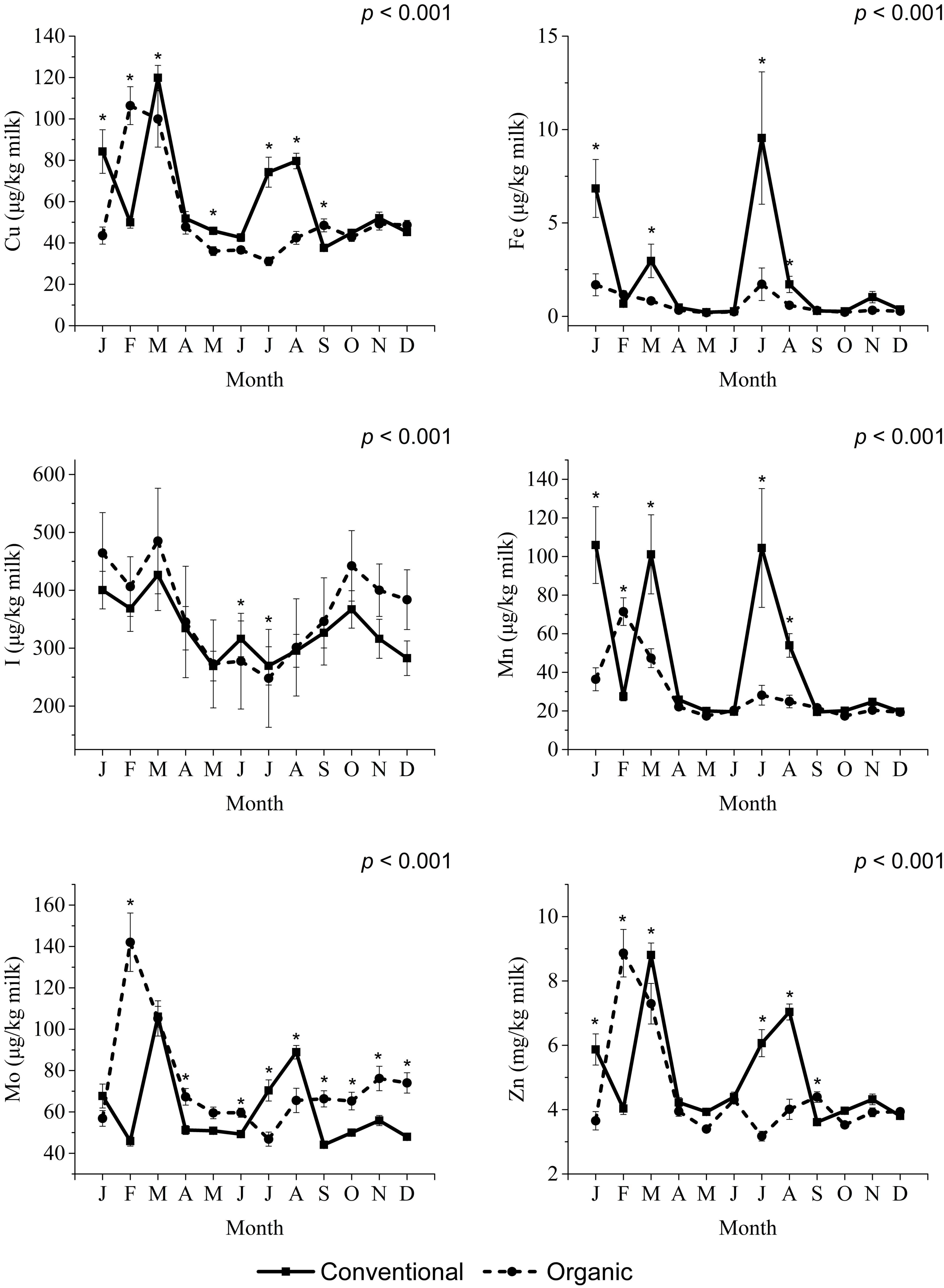
[2]The p-value of SCC was obtained from the fitted mixed linear model based on log(x)-transformed values. The p-values of macrominerals and trace elements were obtained from the fitted mixed linear model based on the log(x+1)-transformed values. Different letters indicate significant ($p < 0.05$) difference between months.

Figure 1. Concentrations of macrominerals (K, P) and non-essential trace elements (Al, Sn) in milk where a significant ($p < 0.05$) interaction between production system and month was observed. Means were calculated from the measured values. The error bars represent the standard error of the means. The *p*-values were obtained from the fitted mixed linear model based on the log(x+1)-transformed values. *: significant difference between conventional and organic farms within the month ($p < 0.05$).

Figure 2. Concentrations of essential trace elements in milk where a significant ($p < 0.05$) interaction between production system and month was observed. Means were calculated from the measured values. The error bars represent the standard error of the means. The *p*-values were obtained from the fitted mixed linear model based on the log(x+1)-transformed values. *: significant difference between conventional and organic farms within the month ($p < 0.05$).

Figure 3. Biplot derived from the redundancy analysis using our database, showing the relationship between herd breeding (Non-Holstein, NHOL) and diet composition (total forage, TFOR; estimated grazing, GRA; grass and grass/clover silage, GGCS; maize silage, MS; lucerne silage, LS; wholecrop (ensiled whole wheat plants, harvested approximately 1 month before grain maturity), WC; hay/straw, HS; moist by-products, MBP; dry straights, DRY; cereals, CER; oil, OIL; minerals, MIN; and vitamins, VIT) parameters relative to the (i) milk yield (*yield*), milk contents of fat (*fat*), protein (*prot*), lactose (*lact*), somatic cell count (*scc*) and milk concentrations of *Al*, *Mn*, *Fe*, *Cu*, *Zn*, *Mo*, *Sn*, *Na*, *Mg*, *P*, *K*, *Ca* and *I*. Axis 1 explained 4.2% of the variation and axis 2 a further 2.1%. Continuous variables, shown as arrows were (in order of contribution to the explained variation GRA ($p = 0.002$), MS ($p = 0.002$), TFOR ($p = 0.002$), NHOL ($p = 0.006$), CER ($p = 0.016$), DRY ($p = 0.048$), OIL ($p = 0.046$), MIN ($p = 0.074$), WC ($p = 0.096$), VIT ($p = 0.204$), HS ($p = 0.148$), GGCS ($p = 0.540$), MBP ($p = 0.724$), LS ($p = 0.872$).


p < 0.001
1600
1500
1400
1300
1200
1100
K (mg/kg milk)
J F M A M J J A S O N D
Month
p < 0.001
900
850
800
750
700
650
600
P (mg/kg milk)
J F M A M J J A S O N D
Month
p < 0.001
6
5
4
3
2
1
0
Al (mg/kg milk)
J F M A M J J A S O N D
Month
p < 0.001
9
8
7
6
5
4
3
2
1
0
Sn (µg/kg milk)
J F M A M J J A S O N D
Month
Conventional
Organic


p < 0.001
Cu (µg/kg milk)
0
20
40
60
80
100
120
140
J F M A M J J A S O N D
Month
p < 0.001
Fe (µg/kg milk)
0
5
10
15
J F M A M J J A S O N D
Month
p < 0.001
I (µg/kg milk)
200
300
400
500
600
J F M A M J J A S O N D
Month
p < 0.001
Mn (µg/kg milk)
0
20
40
60
80
100
120
140
J F M A M J J A S O N D
Month
p < 0.001
Mo (µg/kg milk)
40
60
80
100
120
140
160
J F M A M J J A S O N D
Month
p < 0.001
Zn (mg/kg milk)
2
4
6
8
10
J F M A M J J A S O N D
Month
Conventional
Organic

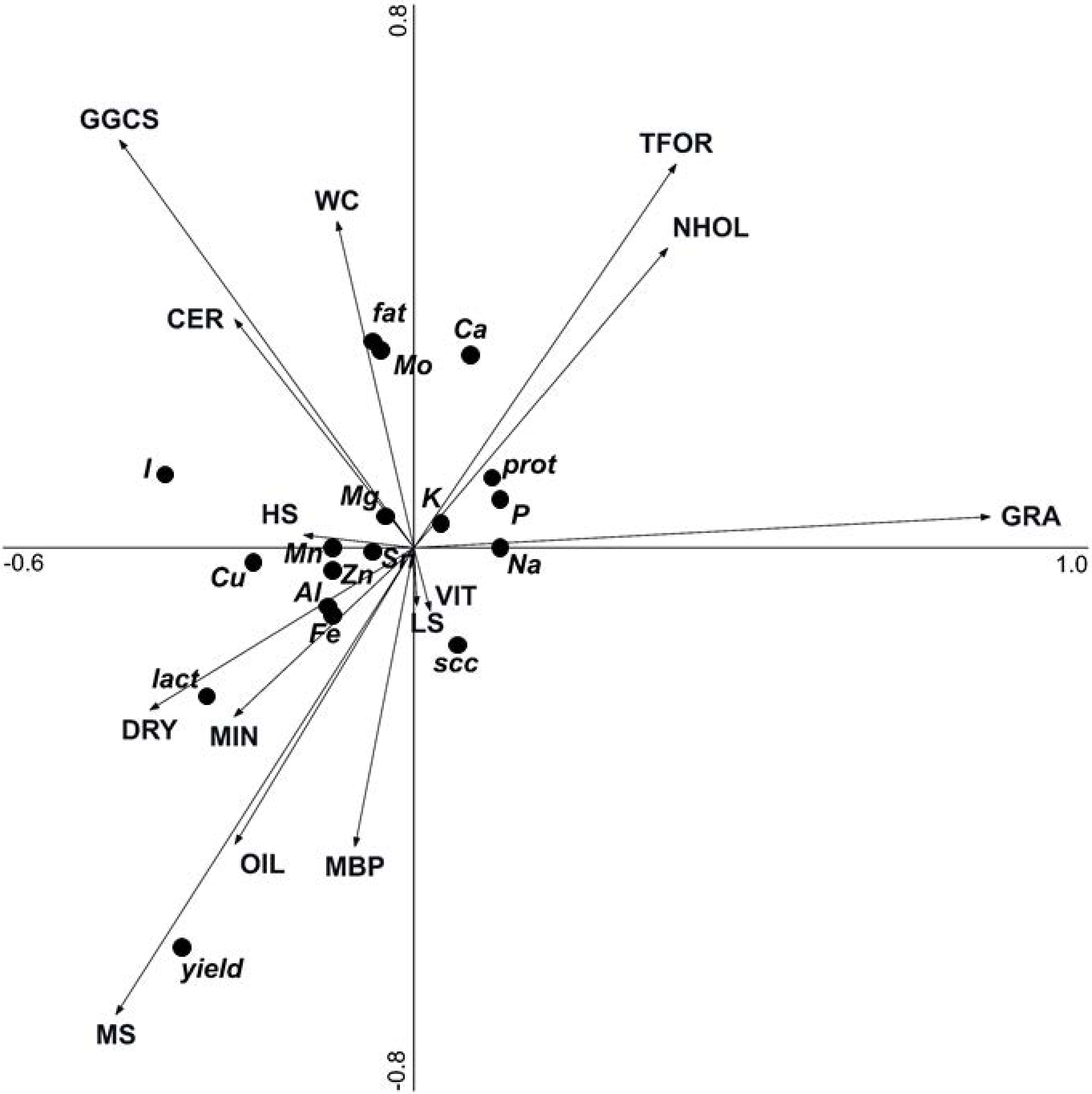

0.8
GGCS
TFOR
WC
NHOL
CER
fat
Ca
Mo
I
prot
Mg
K
P
GRA
HS
Mn
Sr
Na
-0.6
Cu
Zn
1.0
Al
VIT
LS
Fe
scc
lact
DRY
MIN
OIL
MBP
yield
MS
-0.8

Table S1. Average liveweight for each breed used to estimated total dry matter intake and pasture intake (by difference), as described by Butler et al. (2008)[a]

Breed	Liveweight (kg)	Source
Holstein	680	https://www.thecattlesite.com/breeds/dairy/22/holstein/
British Friesian	580	https://www.cows.ie/british-friesian-cattle/
New Zealand Friesian	468	https://www.crv4all-international.com/wp-content/uploads/2016/02/229-14-info-rassen-NZ-Genetics.pdf
Jersey	425	https://www.thecattlesite.com/breeds/dairy/23/jersey/
Scandinavian Red	548	http://www.thecattlesite.com/breeds/dairy/37/norwegian-red/
Shorthorn	600	https://www.vetstream.com/treat/bovis/breeds-pages/shorthorn
Ayrshire	544	https://www.thecattlesite.com/breeds/dairy/19/ayrshire/
Montbeliarde	685	https://www.thecattlesite.com/breeds/dairy/27/montbliarde/
Brown Swiss	590	https://www.thecattlesite.com/breeds/dairy/31/brown-swiss/
Guernsey	475	https://www.thecattlesite.com/breeds/dairy/21/guernsey/
Other breed	560	Average of all other breeds
Crossbreed	560	Average of all other breeds

[a] Butler, G., Nielsen, J., Slots, T., Seal, C., Eyre, M., Sanderson, R., & Leifert, C. (2008). Fatty acid and fat-soluble antioxidant concentrations in milk from high- and low-input conventional and organic systems: seasonal variation. Journal of Science of Food and Agriculture, 88, 1431-1441

Table S2. Changes of average milk yield, basic composition and macromineral and trace element composition of conventional farms over time.

Parameter	Mean[1]												SE	*p*-value[2]
	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec		
Yield and basic composition														
Milk yield (kg/cow/d)	30.4^{abc}	29.5^{cd}	30.5^{a}	30.1^{abc}	30.3^{ab}	29.6^{bcd}	29.9^{abcd}	29.2^{def}	28.7^{ef}	28.5^{f}	28.4^{f}	29.8^{cde}	0.67	<0.001
Fat (g/kg milk)	40.2^{abc}	39.0^{cd}	39.5^{bc}	38.9^{cd}	37.9^{e}	38.3^{de}	36.6^{f}	38.9^{cd}	39.0^{cd}	40.6^{a}	41.1^{a}	40.2^{ab}	0.40	<0.001
Protein (g/kg milk)	33.6^{b}	33.6^{c}	33.1^{d}	33.0^{d}	32.8^{e}	32.5^{ef}	32.3^{f}	32.6^{e}	33.4^{c}	34.5^{b}	35.0^{a}	34.3^{b}	0.18	<0.001
Lactose (g/kg milk)	45.5^{a}	45.3^{b}	45.5^{a}	45.3^{b}	45.3^{b}	45.2^{b}	45.2^{b}	44.9^{cd}	44.9^{d}	45.0^{cd}	45.0^{c}	45.1^{c}	0.09	<0.001
SCC (×1000/ml milk)	154.0^{abc}	131.5^{c}	157.7^{ab}	164.7^{ab}	150.2^{abc}	174.5^{abc}	149.0^{a}	161.6^{ab}	117.3^{d}	146.4^{abc}	148.4^{abc}	143.9^{bc}	7.50	<0.001
Macrominerals (mg/kg milk)														
Ca	951.0^{c}	969.4^{c}	1044.3^{ab}	950.0^{c}	971.6^{c}	911.5^{d}	1035.1^{ab}	971.0^{c}	1055.0^{ab}	1056.9^{a}	1007.6^{b}	1028.2^{ab}	12.76	<0.001
K	1244.6^{d}	1341.5^{c}	1406.8^{ab}	1331.1^{c}	1366.2^{bcd}	1260.2^{d}	1445.3^{a}	1348.1^{c}	1429.0^{a}	1427.4^{a}	1335.0^{c}	1415.8^{a}	16.29	<0.001
Mg	89.8^{f}	90.9^{ef}	101.2^{ab}	90.3^{f}	94.3^{d}	85.5^{g}	98.5^{bc}	93.9^{de}	102.8^{ab}	104.3^{a}	102.1^{ab}	94.9^{cd}	1.32	<0.001
Na	281.7^{g}	315.0^{ef}	383.1^{b}	354.7^{c}	330.8^{d}	309.9^{f}	405.8^{a}	327.3^{de}	351.8^{c}	362.9^{c}	349.3^{c}	351.3^{c}	6.14	<0.001
P	685.3^{g}	782.2^{de}	842.9^{a}	795.2^{cd}	810.9^{bcd}	729.3^{f}	829.7^{ab}	760.5^{e}	814.4^{bcd}	808.7^{bcd}	794.0^{cd}	858.5^{ab}	10.64	<0.001
Essential trace elements (µg/kg milk unless otherwise stated)														
Cu	84.2^{bc}	49.9^{d}	119.9^{a}	51.8^{d}	45.8^{de}	42.6^{ef}	74.2^{c}	79.6^{b}	37.6^{f}	44.8^{de}	52.0^{d}	45.2^{de}	3.76	<0.001
Fe (mg/kg milk)	6.85^{a}	0.68^{de}	2.97^{b}	0.47^{de}	0.22^{e}	0.27^{e}	9.55^{a}	1.71^{bc}	0.29^{de}	0.27^{e}	1.03^{cd}	0.37^{de}	0.589	<0.001
I	400.4^{a}	373.9^{abc}	426.4^{ab}	334.5^{cd}	269.1^{ef}	316.2^{cde}	269.4^{f}	295.5^{def}	326.6^{bcd}	367.0^{ab}	316.1^{de}	282.6^{ef}	34.31	<0.001
Mn	105.9^{a}	27.5^{c}	101.1^{a}	25.8^{cd}	20.0^{de}	19.6^{de}	104.4^{ab}	53.9^{b}	19.4^{e}	20.1^{cde}	24.6^{cde}	19.6^{de}	7.19	<0.001
Mo	67.7^{cd}	45.9^{gh}	106.0^{a}	51.3^{efg}	50.9^{ef}	49.3^{efgh}	70.4^{c}	88.9^{b}	44.1^{h}	50.0^{efgh}	55.8^{de}	47.9^{fgh}	2.72	<0.001
Zn (mg/kg milk)	5.87^{c}	4.03^{def}	8.81^{a}	4.22^{de}	3.93^{def}	4.39^{d}	6.07^{c}	7.04^{b}	3.61^{f}	3.96^{def}	4.32^{de}	3.80^{ef}	0.211	<0.001
Non-essential trace elements														
Al (mg/kg milk)	4.38^{a}	0.37^{cd}	3.57^{b}	0.32^{cd}	0.07^{d}	0.08^{d}	3.46^{b}	0.67^{c}	0.19^{cd}	0.14^{d}	0.40^{cd}	0.15^{cd}	0.368	<0.001
Sn (µg/kg milk)	4.26^{bc}	1.48^{ef}	4.73^{a}	2.11^{cde}	3.26^{bcd}	1.53^{ef}	3.26^{de}	2.55^{cde}	1.33^{f}	1.95^{ef}	1.64^{ef}	75.88^{ab}	6.070	<0.001

[1]The means of milk yield, fat, protein and lactose contents are the predicted means obtained from the fitted mixed linear model. The means of somatic cell count (SCC), macrominerals and trace elements are the arithmetic means of the measured values. The model included month as fixed factor and farm ID as random factor.

[2]The *p*-value of SCC was obtained from the fitted mixed linear model based on log(x)-transformed values. The *p*-values of macrominerals and trace elements were obtained from the fitted mixed linear model based on the log(x+1)-transformed values. Different letters indicate significant ($p < 0.05$) difference between months.

Table S3. Changes of average milk yield, basic composition and macromineral and trace element composition of organic farms over time.

Parameter	Mean[1]												SE	p-value[2]
	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec		
Yield and basic composition														
Milk yield (kg/cow/d)	21.8	21.9	21.1	23.7	23.8	22.6	21.6	21.6	21.8	21.8	22.3	22.5	0.95	0.073
Fat (g/kg milk)	40.8^{bcd}	40.0^{cd}	39.9^{d}	38.3^{e}	37.4^{ef}	38.2^{ef}	37.0^{f}	39.9^{cd}	41.3^{ab}	41.4^{ab}	42.1^{a}	41.0^{abc}	0.55	<0.001
Protein (g/kg milk)	33.0^{bc}	33.0^{bc}	32.5^{c}	33.2^{bc}	33.1^{bc}	33.0^{bc}	32.5^{c}	33.6^{bc}	33.3^{b}	33.6^{bc}	34.3^{a}	33.6^{bc}	0.39	<0.001
Lactose (g/kg milk)	45.1^{bcd}	45.1^{bcd}	45.4^{a}	45.3^{ab}	45.2^{abc}	44.9^{cd}	44.4^{ef}	44.1^{f}	44.3^{f}	44.7^{de}	44.9^{bcd}	44.9^{bcd}	0.17	<0.001
SCC (×1000/ml milk)	138.2^{ab}	121.4^{b}	152.4^{a}	129.1^{ab}	127.7^{ab}	150.3^{a}	146.9^{a}	156.8^{a}	109.8^{c}	137.2^{a}	133.3^{ab}	128.4^{ab}	11.67	<0.001
Macrominerals (mg/kg milk)														
Ca	974.1^{ef}	1005.4^{de}	1087.7^{b}	1019.4^{cd}	1009.7^{de}	948.7^{f}	1093.1^{b}	1060.4^{bc}	1091.8^{b}	1153.0^{a}	1088.2^{b}	1062.8^{b}	14.75	<0.001
K	1177.1^{g}	1308.2^{e}	1392.8^{cd}	1369.9^{d}	1388.7^{d}	1250.2^{f}	1488.6^{ab}	1425.2^{cd}	1445.1^{bc}	1532.0^{a}	1405.5^{cd}	1419.7^{cd}	19.97	<0.001
Mg	85.3^{e}	91.6^{d}	100.3^{bc}	89.9^{d}	92.1^{d}	84.9^{ef}	100.1^{bc}	97.1^{c}	99.1^{bc}	105.4^{a}	102.1^{ab}	92.4^{d}	1.49	<0.001
Na	265.6^{h}	323f^{g}	376.5^{b}	355.3^{bcd}	329.3^{ef}	309.4^{g}	416.6^{a}	349.8^{cde}	355.4^{bcd}	366.5^{bc}	346.5^{cde}	343.2^{de}	9.12	<0.001
P	644.5^{e}	772.7^{c}	842.2^{ab}	841.7^{ab}	835.6^{ab}	737.6^{d}	858.6^{a}	819.4^{b}	811.7^{b}	837.2^{ab}	816.5^{b}	863.6^{a}	11.70	<0.001
Essential trace elements (μg/kg milk unless otherwise stated)														
Cu	43.5^{cd}	106.4^{a}	100.0^{a}	47.8^{b}	36.0^{cd}	36.6^{cd}	31.1^{d}	42.5^{cd}	48.5^{b}	42.8^{cd}	49.2^{b}	48.7^{b}	4.15	<0.001
Fe (mg/kg milk)	1.69^{a}	1.16^{a}	0.83^{ab}	0.33^{cd}	0.20^{d}	0.24^{cd}	1.72^{ab}	0.59^{bc}	0.32^{cd}	0.22^{d}	0.33^{cd}	0.28^{cd}	0.181	<0.001
I	464.4^{a}	406.4^{ab}	485.1^{ab}	345.3^{de}	272.9^{fg}	277.6^{efg}	247.9^{g}	301.3^{ef}	346.2^{cd}	442.2^{ab}	400.2^{ab}	383.8^{bc}	72.39	<0.001
Mn	36.4^{c}	71.5^{a}	47.3^{b}	22.1^{d}	17.4^{e}	20.3^{de}	28.1^{d}	24.9^{d}	21.7^{de}	17.4^{e}	20.4^{de}	19.3^{de}	2.87	<0.001
Mo	56.9^{d}	142.0^{a}	105.2^{b}	67.3^{cd}	59.5^{d}	59.6^{d}	46.8^{e}	65.6^{cd}	66.3^{cd}	65.2^{cd}	76.2^{c}	74.0^{c}	5.31	<0.001
Zn (mg/kg milk)	3.65^{de}	8.86^{a}	7.29^{b}	3.95^{cd}	3.40^{de}	4.31^{c}	3.17^{e}	4.01^{cd}	4.39^{c}	3.53^{de}	3.92^{cd}	3.93^{cd}	0.242	<0.001
Non-essential trace elements														
Al (mg/kg milk)	0.76^{a}	0.77^{a}	0.61^{ab}	0.17^{cd}	0.06^{d}	0.10^{cd}	0.49^{bc}	0.22^{cd}	0.17^{cd}	0.12^{cd}	0.18^{cd}	0.13^{cd}	0.101	<0.001
Sn (μg/kg milk)	2.08^{bcd}	3.48^{a}	3.30^{ab}	2.04^{bcd}	2.51^{abc}	1.56^{de}	4.75^{e}	1.87^{cde}	2.57^{de}	1.37^{de}	1.72^{bcd}	4.86^{ab}	0.872	<0.001

[1]The means of milk yield, fat, protein and lactose contents are the predicted means obtained from the fitted mixed linear model. The means of somatic cell count (SCC), macrominerals and trace elements are the arithmetic means of the measured values. The model included month as fixed factor and farm ID as random factor.
[2]The p-value of SCC was obtained from the fitted mixed linear model based on log(x)-transformed values. The p-values of macrominerals and trace elements were obtained from the fitted mixed linear model based on the log(x+1)-transformed values. Different letters indicate significant ($p < 0.05$) difference between months.

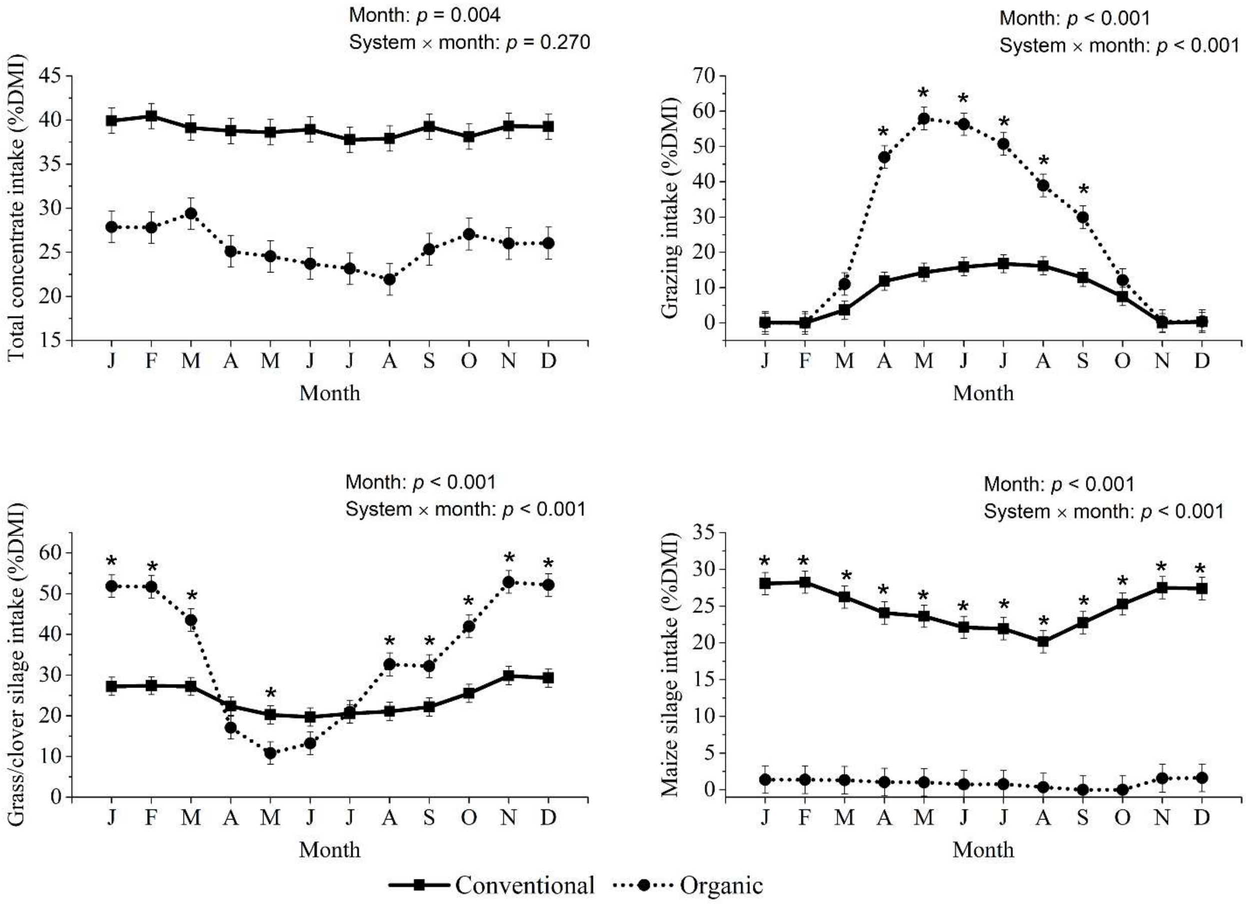


Figure S1. Intakes of concentrate, grazing, grass/clover silage, and maize silage in conventional and organic farms. Means are the predicted means from the fitted mixed linear model. The error bars represent the standard error obtained from the same linear model. *: significant difference between conventional and organic farms within the month ($p < 0.05$).

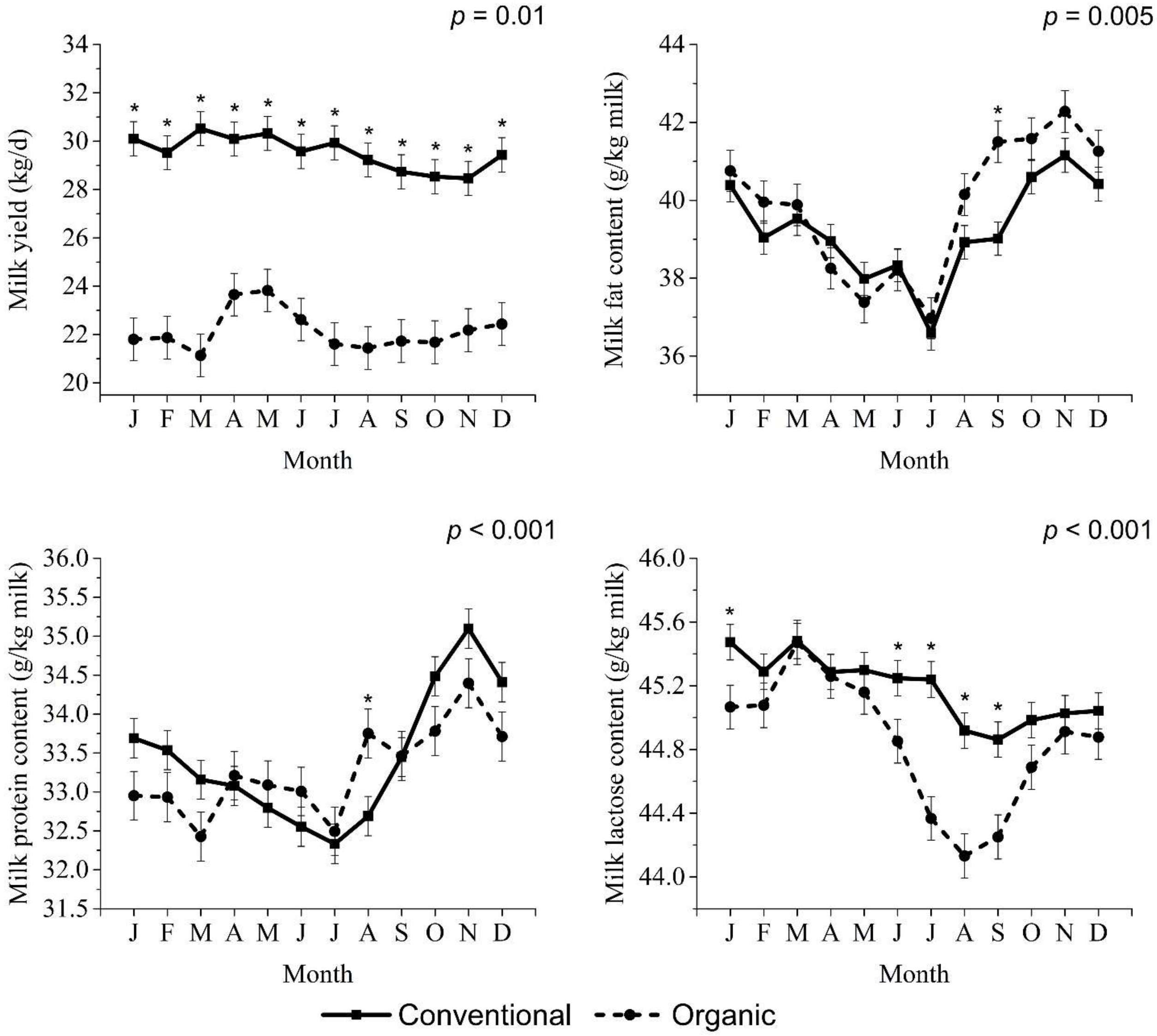


Figure S2. Significant interactions between production system and month of milk yield and composition. Means are the predicted means from the fitted mixed linear model. The error bars represent the standard error obtained from the same linear model. *: significant difference between conventional and organic farms within the month ($p < 0.05$).