

An update to the fatty acid profiles of bovine retail milk in the United Kingdom: implications for nutrition in different age and gender groups

Article

Accepted Version

Creative Commons: Attribution-Noncommercial-No Derivative Works 4.0

Stergiadis, S. ORCID: https://orcid.org/0000-0002-7293-182X, Berlitz, C. B., Hunt, B., Garg, S., Givens, D. I. ORCID: https://orcid.org/0000-0002-6754-6935 and Kliem, K. E. ORCID: https://orcid.org/0000-0002-0058-8225 (2019) An update to the fatty acid profiles of bovine retail milk in the United Kingdom: implications for nutrition in different age and gender groups. Food Chemistry, 276. pp. 218-230. ISSN 0308-8146 doi: https://doi.org/10.1016/j.foodchem.2018.09.165 Available at https://centaur.reading.ac.uk/79463/

It is advisable to refer to the publisher's version if you intend to cite from the work. See <u>Guidance on citing</u>.

To link to this article DOI: http://dx.doi.org/10.1016/j.foodchem.2018.09.165

Publisher: Elsevier

All outputs in CentAUR are protected by Intellectual Property Rights law, including copyright law. Copyright and IPR is retained by the creators or other copyright holders. Terms and conditions for use of this material are defined in



the End User Agreement.

www.reading.ac.uk/centaur

CentAUR

Central Archive at the University of Reading

Reading's research outputs online

An update to the fatty acid profiles of bovine retail milk in the United Kingdom: implications for nutrition in different age and gender groups

Running title: Fatty acid profile of UK retail milk and dietary intake implications

Sokratis Stergiadis^{*a}, Carolina B. Berlitz^{a,b}, Benjamin Hunt^a, Sneha Garg^a, D. Ian Givens^c, Kirsty E. Kliem^{a, c}

^a University of Reading, Animal, Dairy and Food Chain Sciences, School of Agriculture, Policy and Development, PO Box 237, Earley Gate, Reading, RG6 6AR, United Kingdom

^b Federal University of Rio Grande do Sul, Department of Animal Science, Av Bento Gonçalves, 7712, Porto Alegre, RS, 91540-000, Brazil.

^c University of Reading, Institute for Food, Nutrition and Health, PO Box 237, Earley Gate, Reading, RG6 6AR, United Kingdom

* Corresponding author: Sokratis Stergiadis, <u>s.stergiadis@reading.ac.uk</u>

1

Abstract

2 This study investigated the effect of UK dairy production system, month, and their interaction, 3 on retail milk fatty acid (FA) profile throughout the year. Milk samples (n=120) from four conventional (CON), four organic (ORG) and two free-range (FR) brands were collected 4 5 monthly. ORG milk had more nutritionally-desirable polyunsaturated FA, including rumenic 6 acid and the omega-3 PUFA α -linolenic, eicosapentaenoic and docosapentaenoic acids, and 7 less of the nutritionally-undesirable palmitic acid. Milk FA profile was similar between FR and 8 CON, but FR milk had less SFA and/or palmitic acid, and/or greater α-linolenic and rumenic 9 acids in certain months within the peak-grazing season. According to the measured milk FA 10 profiles and UK milk fat intakes, milk and dairy products contribute around one-third of the 11 maximum recommended saturated FA intake. A small increased intake of beneficial PUFA 12 may be expected by consuming ORG milk but human health implications from such differences 13 are unknown.

14

Keywords: milk; dairy management; dietary intakes; fatty acids; free-range; human health;
omega-3; organic

17

1. Introduction

Milk and dairy products provide a range of beneficial nutrients for human health, including 18 19 fatty acids (FA), proteins, bioactive peptides, minerals, carotenoids and vitamins (Haug, 20 Hostmark, & Harstad, 2007; Pereira, 2014; Thorning, et al., 2017). However, milk and dairy 21 products are dietary sources of saturated fatty acids (SFA), such as C12:0, C14:0 and C16:0, 22 elevated consumption of which may increase the risk of cardiovascular disease (CVD) (EFSA, 23 2010; FAO, 2010). These concerns and the increased incidence of lifestyle-related diseases, 24 such as obesity and CVD, may have contributed to the reduction in whole milk consumption 25 in developed countries, including UK, Denmark, France, USA, Canada and Germany (Kliem 26 & Givens, 2011). In the UK, whole milk consumption has decreased 5-fold compared with 27 1970s' levels, and despite the simultaneous increase in semi-skimmed milk consumption, the overall milk intake has declined (Kliem, et al., 2011). In contrast, milk is also rich in FA with 28 29 potentially beneficial effects on human health (see reviews from (Barcelo-Goblijn & Murphy, 30 2009; Dilzer & Park, 2012; Field, Blewett, Proctor, & Vine, 2009; Haug, et al., 2007; Swanson, Block, & Mousa, 2012)), such as the monounsaturated FA (MUFA) t11 C18:1 (VA, vaccenic 31 acid) and c9 C18:1 (OA, oleic acid), the polyunsaturated FA (PUFA) c9c12c15 C18:3 (ALNA, 32 33 α-linolenic acid), c5c8c11c14c17 C20:5 (eicosapentaenoic, EPA), c7c10c13c16c19 C22:5 34 (docosapentaenoic, DPA) and c4c7c10c13c16c19 C22:6 (docosahexaenoic acid, DHA), which 35 are omega-3 PUFA (n-3), the c9c12 C18:2 (LA, linoleic acid), which is an omega-6 PUFA (n-36 6), and the conjugated FA c9t11 C18:2 (RA, rumenic acid) (Kliem & Shingfield, 2016; Pereira, 37 2014).

Current nutritional recommendations are to reduce SFA consumption (as low as possible and
not exceeding 10% of total energy intake) and substitute dietary SFA with MUFA and/or PUFA
(EFSA, 2010; FAO, 2010). Previous research has shown that dairy management, and especially
cow diet, influence milk FA profiles; for example, cows with increased fresh grass intake,

42 higher dietary forage:concentrate ratio, and/or diets supplemented with plant oils, oilseeds or 43 protected lipids may produce milk with a FA profile that contains less SFA and more n-3 and 44 RA (Chilliard, Glasser, Ferlay, Bernard, Rouel, & Doreau, 2007; Elgersma, 2015; Kliem, et 45 al., 2016). Therefore, potential differences between different dairy production systems, which 46 involve differences in cow nutrition, may reflect on milk FA composition. In the UK, organic 47 milk contained greater concentrations of ALNA, EPA and n-3 PUFA all year round, and less 48 SFA in milk fat, including C16:0, during summer, when compared with conventional milk 49 (Butler, Stergiadis, Seal, Eyre, & Leifert, 2011b; Stergiadis, et al., 2012). A seasonal effect on 50 milk FA composition has been previously demonstrated in UK retail milk (Kliem, Shingfield, 51 Livingstone, & Givens, 2013), which also influences the extent of the compositional 52 differences between organic and conventional milk (Butler, et al., 2011b). However, the 53 interaction between production system and season has been assessed only during January and 54 July (Butler, et al., 2011b), which are potentially among the months with the highest difference 55 in pasture intake in UK dairy systems (Stergiadis, et al., 2012), so a more detailed assessment 56 throughout the year is required.

57 Fresh grass intake strongly influences n-3 PUFA content of milk fat, as recently highlighted in 58 several multivariate redundancy analyses (Stergiadis, et al., 2015a; Stergiadis, et al., 2015b; 59 Stergiadis, et al., 2012). Bulk tank milk from conventional extensive pasture-based farms, (pasture intake contributing more than 90% of cow dry matter intake), contained more of the 60 61 potentially nutritionally beneficial, when replacing SFA in human diets, MUFA and/or PUFA 62 and less SFA when compared with conventional and/or organic milk, although differences were 63 not consistent throughout the year or in all studies (Butler, et al., 2008; Stergiadis, et al., 2015b). 64 Recently, free-range milk, certified on farms where cows have access to pasture for a minimum 65 of 180 days/year and being outdoors for a minimum of 23 hours/day during the grazing season, reached the UK market. In the Netherlands, retail milk from dairy farms under a similar 66

certification scheme, but with less mandatory access to pasture (minimum 120 days/year at
pasture and 6 hours/day), had a similar FA profile to retail conventional milk (Capuano,
Gravink, Boerrigter-Eenling, & van Ruth, 2015) but potential differences under the UK dairy
management practices have not yet been investigated.

This study therefore aimed to (i) investigate the effect of production system (conventional, organic and, for the first time in the UK, free-range), month (March through to February) and their interaction, on retail milk FA profile throughout the year, and (ii) assess the potential implications on the intakes of FA which are relevant to human health.

75

2. Materials and methods

76 2.1 Experiment/survey design

All milk samples (n=120) in the present study were collected from retail outlets in England. 77 78 The survey lasted for 12 months and samples were collected monthly between March 2016 and 79 February 2017. Four brands of conventional milk and four brands of organic milk were sampled 80 monthly from four retail outlets within a 8 km radius of the University of Reading. The only 81 two brands of free-range-certified milk available to UK consumers during the period of this 82 study were obtained monthly from dairies in Lancashire and Gloucestershire. All retail milk 83 samples were whole, pasteurized and homogenized, while conventional and free-range milk 84 had also their fat content standardized to approximately 3.5 and 3.7 g/100g milk, respectively. 85 Milk samples were collected to represent the latest "best before" date, available at the day of sampling, to ensure minimum storage time at retail outlet. Milk samples in commercial 86 87 packaging were immediately transferred to the laboratories of the University of Reading, and 88 aliquoted into 30-ml sterile polypropylene screw-top containers and were frozen at -20°C until 89 analysis.

90 2.2 Milk analysis

91 Concentrations of fat, protein, casein, and lactose were analysed using a Milkoscan FT6000 (Foss Electric, Hillerod, Denmark), while somatic cell count (SCC) was analysed by a 92 Fossomatic (Foss Electric, Hillerod, Denmark), in the National Milk Laboratories 93 94 (Wolverhampton, UK). Milk FA profiles were analysed by GC flame ionisation detection (Bruker 350 GC, Bruker, Germany) according to previously described methods of esterification 95 96 and methylation (Chilliard, Martin, Rouel, & Doreau, 2009), and techniques of peak identification and quantification (Kliem, et al., 2013). A combined correction factor, to account 97 98 for carbon deficiency in the response of flame ionization detector for FA methyl esters with 4-99 10 atoms of carbon was used (Ulberth, Gabernig, & Schrammel, 1999).

100 2.3 Statistical analysis

Analysis of variance (ANOVA), derived from linear mixed effects models (residual maximum 101 102 likelihood analysis; REML) (Gilmour, Thompson, & Cullis, 1995) in GenStat (VSN International, 17th Edition, Hempstead, UK), by considering management (Conventional, 103 104 CON; Organic, ORG; Free-Range, FR) and month (March, April, May, June, July, August, 105 September, October, November, December, January, February), and their interaction, as fixed factors and milk ID (which was unique for each combination of brand/retailer and 106 107 management) as a random factor. Significant effect of the main treatments was declared when 108 P < 0.05 and tendencies were declared when 0.05 < P < 0.10. The residual diagnostics of the 109 final model were assessed using normality plots, with no data showing deviation from 110 normality except for SCC which were log-transformed prior to ANOVA. Pairwise comparisons 111 of means (P < 0.05) were performed using Fisher's Least Significant Difference test. Milk FA 112 profiles are reported as g/kg milk fat. Atherogenicity index (AI), thrombogenicity index (TI), 113 as markers to indicate potential risk of CVD, were calculated according to Srednicka-Tober et 114 al. (2016), as follows:

•
$$AI = (C12:0 + 4 \times C14:0 + C16:0) / (MUFA + PUFA),$$

116 117	• $TI = (C14:0 + C16:0 + C18:0) / [(0.5 \times MUFA) + (0.5 \times n-6) + (3 \times n-3) + (n-3/n-6)].$ Δ^9 -desaturase activity index (Δ^9I) was calculated according to Kay et al. (2004) as:
118	• $\Delta^9 I = (c9 C14:1+c9 C16:1+OA+RA) / (c9 C14:1+c9 C16:1+OA+RA+C14:0+C16:1+OA+RA+C14:0+C16:1+OA+RA+C14:0+C16:1+OA+RA+C14:0+C16:1+OA+RA+C14:0+C16:1+OA+RA+C14:0+C16:1+C16:1+C16+C16+C16+C16+C16+C16+C16+C16+C16+C1$
119	C16:0 + C18:0 + VA)
120	For the purposes of the intake calculations, this study assumes that all dairy products produced
121	in the UK have the same FA profile as the whole milk analysed. Intakes of individual FA or
122	FA groups, for males/females/all for the age groups of 4-10/11-18/19-64/65+ were estimated
123	separately as:
124	FA intake (g/d) = fat intake (g/d) (Bates, et al., 2014) × contribution of fat from milk and dairy
125	products (% of total fat intake) (Bates, et al., 2014) \times 0.933 (correction factor representing %
126	of FA in total milk fat) (Kliem, et al., 2013) \times milk FA concentration (% of total FA).
127	3. Results
128	All differences discussed in the Results section were statistically significant ($P < 0.05$) unless
129	otherwise stated.
130	3.1 Milk basic composition
131	3.1.1 Effect of production system
132	Significant effect of production system was identified for milk concentrations of fat and lactose
133	(Table 1). Compared with CON and FR milk, respectively, ORG milk contained more fat and
134	less lactose (Table 1). There were no significant differences in milk composition between CON
135	and FR milk (Table 1).
136	3.1.2 Effect of month
137	Significant effects of month were identified for milk concentrations of all basic composition

138 parameters (Table 2). Milk contained less fat during May-September and December than in

139 March-April, with the remaining months showing intermediate values, mostly without being 140 significantly different (Table 2). Protein concentrations in milk were higher in May, October 141 and November (highest) than in June-September, December and February, with the remaining 142 months showing intermediate values, mostly without being significantly different (Table 2). Casein concentrations in milk were highest in October-November when compared with all 143 144 other months, although the difference with May was not statistically significant (Table 2). 145 Highest lactose concentrations were observed in March-May and lowest in July, September, 146 October and December with intermediate values being observed during the other months, 147 mostly without being significantly different (Table 2). Milk had higher SCC during winter 148 (December-February) than in March-November, although the difference with January was not 149 statistically significant (Table 2).

150

3.1.3 Effect of the production system \times month interaction

151 Significant effects of the production system × month interaction were identified for milk lactose
152 concentrations (Appendix; Figure A1); CON milk had more lactose than ORG and FR milk in
153 June, September and November, and less lactose than FR milk in July.

154 3.2 Milk FA profile

155 *3.2.1 Effect of production system*

156 Significant effect of the production system was identified for milk concentrations of C16:0,

157 RA, ALNA, EPA, DPA, PUFA, n-3, and *trans* FA and the ratios of n-3/n-6, TI, C14:1/C14:0,

158 C16:1/C16:0 and OA/C18:0 (Table 1). Compared with CON and FR milk, respectively, ORG

159 milk had lower concentrations of C16:0 and higher concentrations of RA, ALNA, EPA, DPA,

160 trans MUFA, PUFA, cis PUFA, trans PUFA, cis/trans plus trans/cis PUFA, n-3 and trans FA

161 (Table 1). ORG milk had a higher ratio of n-3/n-6 and lower ratio of TI, C14:1/C14:0,

162 C16:1/C16:0 and OA/C18:0 than CON and FR milk although the difference between ORG and

FR milk for OA/C18:0 ratio was not statistically significant (Table 1). There were no
significant differences in FA profile between CON and FR milk (Table 1). The effect of
production system in the full FA profile of milk (80 individual FA) is shown in the Appendix
(Table A1).

3.2.2 Effect of month

Significant effects of month were identified for milk concentrations of all individual FA (except 168 169 DHA) and FA groups and indices (Table 2). Concentrations of C12:0 in milk fat were lower in 170 June-October than in March-May and November-February, with numerically smaller 171 significant differences between months within these periods also being observed (Table 2). 172 Concentrations of C14:0 and SFA and the AI and TI in milk were lower in May-October than 173 in March and November-February, with numerically smaller significant differences between months within these periods also being observed; their values were intermediate in April and 174 lower when compared with March and November-February (Table 2). Milk contained more 175 176 C16:0 in March and October-February than in May-August and had intermediate 177 concentrations in April and September, which were also lower when compared with October-178 February (Table 2). Milk contained more C18:0 in May-September than in March and 179 November-February, and had intermediate concentrations in April and October (Table 2).

180 Concentrations of VA, ALNA and EPA and n-3/n-6 in milk were higher in May-October than 181 in March-April and November-February, with numerically smaller significant differences 182 between months within these periods also being observed (Table 2). OA concentrations and 183 Δ^9 I in milk were higher in May-October than in March and November-February, with 184 numerically smaller significant differences between months within these periods also being 185 observed; their values were intermediate in April (Table 2). Milk contained more LA in March-May and October, than in July and November-February, with the remaining months showing 186 intermediate values, mostly without being significantly different (Table 2). RA and trans FA 187

188 concentrations in milk were highest in May-September, lowest in March and November-189 February, and showed intermediate values in April and October, with numerically smaller 190 significant differences between months within these periods also being observed (Table 2). 191 DPA concentrations in milk were higher in May-November than in March-April and 192 December-February, with numerically smaller significant differences between months within 193 these periods also being observed; the means for these parameters were intermediate in 194 January-February and higher when compared with March-April (Table 2).

195 Concentrations of MUFA, cis MUFA, trans MUFA, PUFA, cis PUFA, trans PUFA, cis/trans 196 plus trans/cis PUFA and n-3 in milk were higher in May-October than in March and 197 November-February, with numerically smaller significant differences between months within 198 these periods also being observed; the means for these parameters were intermediate in April 199 (Table 2). Milk contained more n-6 in March-May and September-October, than in July and 200 November-January, with the remaining months showing intermediate values, mostly without 201 being significantly different (Table 2). When excluding VA, trans FA concentrations in milk 202 were highest in May-July, lowest in September-November, and showed intermediate values in 203 March-April, August and December-February; numerically smaller significant differences 204 between months within these periods were also observed (Table 2).

205 Ratio of C14:1/C14:0 was higher in September-November than in March-June and February, 206 with numerically smaller significant differences between months within these periods also 207 being observed; the means for these parameters were intermediate in July-August and 208 December-January and higher when compared with March-May (Table 2). Ratio of 209 C16:1/C16:0 in milk was highest in May-October, lowest in March and December-February, 210 and showed intermediate values in April and November, with numerically smaller significant 211 differences between months within these periods also being observed (Table 2). Ratio of OA/C18:0 in milk was highest in September-November, lowest in April and December-212

February, and showed intermediate values in March and May-August, although the difference between March and April and December-February was not statistically significant; numerically smaller significant differences between months within these periods were also observed (Table 2). Milk had higher ratio of RA/VA in June-August and October-December than in May, with the remaining months showing intermediate values, mostly without being significantly different (Table 2). The effect of month in the full FA profile of milk (80 individual FA) is shown in the Appendix (Table A2).

220 *3.2.3 Effect of the production system* × *month interaction*

221 Significant effects of the production system \times month interaction were identified for milk 222 concentrations of C12:0, C16:0, LA, RA, ALNA (Figure 1), SFA, MUFA, PUFA, n-6, trans 223 FA (total or by excluding VA), n-3/n-6 ratio and AI (Figure 2), Δ^9 I, C14:1/C14:0, C16:1/C16:0, 224 OA/C18:0 and VA/RA (Appendix, Figure A2). C12:0 concentrations in ORG milk were lower 225 than in CON and FR milk in March, but higher than in FR milk in May; C12:0 concentrations 226 in FR milk were higher than in ORG and CON milk in July and December-February (Figure 227 1a). C16:0 concentrations were lower in ORG milk than in CON milk in April-November, and 228 compared with FR milk in February; concentrations in FR milk were also lower than in CON 229 milk in May and August-September (Figure 1b).

LA concentrations were higher in ORG milk than in FR milk in April-May and higher in CON milk than in FR milk in December (Figure 1c). ORG milk had higher RA concentrations when compared with FR and CON milk throughout the year, although differences were not statistically significant between ORG and FR milk in August-September and between ORG and CON milk in January-February; FR milk had higher RA concentrations than CON milk in September (Figure 1d). ORG milk had higher ALNA concentrations when compared with FR and CON milk throughout the year; FR milk had higher ALNA concentrations than CON milk

237 in May and August-September (Figure 1e).

238 SFA concentrations were lower in ORG milk than in FR milk in April, October and February, 239 and when compared with CON milk in April-May; when compared with CON milk, FR milk 240 had less SFA in May but more SFA in February (Figure 2a). FR milk had lower MUFA 241 concentrations than ORG milk in May and CON milk in February, and higher MUFA 242 concentrations than CON milk in June (Figure 2b). ORG milk had higher trans MUFA 243 concentrations than FR and CON milk in April-November but differences in April-May and 244 November were not significant when compared with FR and CON milk, respectively (Figure 245 2c). ORG milk had higher PUFA concentrations when compared with FR and CON milk 246 throughout the year; CON milk had higher PUFA concentrations than FR milk in November-247 February (Figure 2d). ORG milk had higher concentrations of cis PUFA than FR milk in April-248 July and October-February, and when compared with CON milk in April-May, July, 249 November-December and February; CON milk also contained more cis PUFA than FR milk in 250 November-December and February (Figure 2e). FR milk had lower n-6 concentrations 251 compared with ORG milk in May and with CON milk in November-December and February; 252 CON milk had higher LA concentrations than ORG milk in September (Figure 2f). ORG milk 253 had higher concentrations of *trans* FA in April-July and October, than CON and FR milk 254 (Figure 2g). When excluding VA from *trans* FA, ORG milk had higher concentrations than FR 255 milk in April, and when compared with CON milk in July; FR milk contained less trans FA 256 (excluding VA) than CON milk in April, November and December (Figure 2h). ORG milk had 257 higher n-3/n-6 when compared with FR and CON milk in June-February, and when compared 258 with CON milk in April-May; FR milk also had higher n-3/n-6 than CON milk in May (Figure 259 2i). AI was higher in FR milk than in ORG milk in April and February and higher than in CON 260 milk in February; ORG milk had lower AI than CON milk in April (Figure 2j).

261 *3.3 Estimated fatty acid intakes*

262	When the effect of production system on the estimated FA intakes (according to the National
263	Diet and Nutrition survey (Bates, et al., 2014) and milk FA profiles measured in the present
264	study) was assessed, significant effects were identified for the intakes of PUFA, n-3, ALNA,
265	EPA+DHA, and <i>trans</i> FA across all age groups and genders (Table 3). In male children 4-10
266	years old (yo), estimated intakes from ORG milk fat were higher for PUFA (+67 and +85
267	mg/d), n-3 (+51 and +41 mg/d), ALNA (+31 and +26 mg/d), EPA+DHA (+3 and +2 mg/d),
268	and trans FA (+57 and +70 mg/d), when compared with CON and FR milk fat, respectively.
269	In male teenagers 11-18 yo, estimated intakes from ORG milk fat were higher for PUFA (+60
270	and +77 mg/d), n-3 (+46 and +47 mg/d), ALNA (+28 and +24 mg/d), EPA+DHA (+2 and +2
271	mg/d), and trans FA (+52 and +63 mg/d), when compared with CON and FR milk fat,
272	respectively. In adult males 19-64 yo, estimated intakes from ORG milk were higher for PUFA
273	(+61 and +77 mg/d), n-3 (+46 and +37 mg/d), ALNA (+28 and +24 mg/d), EPA+DHA (+2 and
274	+2 mg/d), and trans FA (+52 and +64 mg/d), when compared with CON and FR milk fat,
275	respectively. In adult males over 65 yo, estimated intakes from ORG milk were higher for
276	PUFA (+77 and +99 mg/d), n-3 (+58 and +47 mg/d), ALNA (+36 and +30 mg/d), EPA+DHA
277	(+3 and +3 mg/d), and <i>trans</i> FA (+66 and +81 mg/d), when compared with CON and FR milk
278	fat, respectively. In female children 4-10 yo, estimated intakes from ORG milk were higher for
279	PUFA (+63 and +81 mg/d), n-3 (+48 and +39 mg/d), ALNA (+30 and +25 mg/d), EPA+DHA
280	(+3 and +2 mg/d), and <i>trans</i> FA (+54 and +66 mg/d), when compared with CON and FR milk
281	fat, respectively. In female teenagers 11-18 yo, estimated intakes from ORG milk fat were
282	higher for PUFA (+49 and +62 mg/d), n-3 (+37 and +30 mg/d), ALNA (+23 and +19 mg/d),
283	EPA+DHA (+2 and +1 mg/d), and trans FA (+42 and +51 mg/d), when compared with CON
284	and FR milk fat, respectively. In adult females 19-64 yo, estimated intakes from ORG milk
285	were higher for PUFA (+42 and +54 mg/d), n-3 (+32 and +26 mg/d), ALNA (+20 and +17
286	mg/d), EPA+DHA (+2 and +1 mg/d), and trans FA (+36 and +44 mg/d), when compared with

CON and FR milk, respectively. In adult females over 65 yo, estimated intakes from ORG milk
were higher for PUFA (+66 and +84 mg/d), n-3 (+50 and +40 mg/d), ALNA (+30 and +25
mg/d), EPA+DHA (+3 and +2 mg/d), and *trans* FA (+56 and +68 mg/d), when compared with
CON and FR milk fat, respectively.

291

4. Discussion

292 4.1 Milk basic composition

293 ORG milk contained more fat than CON milk (as in previous UK retail studies; (Butler, et al., 294 2011b)), and FR milk. This may be an effect of fat standardisation at processing plants rather 295 than an effect of production system, as this is common practice in CON and FR, but not in 296 ORG, supply chain. Although the effect of production system on lactose content was 297 significant, the numerical differences were marginal (0.4 g/kg less in ORG than in CON and 298 FR milk) and potential relevance to consumer health is small. This difference may be due to 299 fat standardisation in CON and FR milk; removing fat from whole milk decreases the dilution 300 factor for remaining milk solids, so could increase milk lactose concentration when expressed 301 as g/kg of milk. A significant effect of production system was not observed, for milk protein, 302 casein and SCC contents, in agreement with previous reports for UK milk (Butler, et al., 2011b; 303 Stergiadis, et al., 2012).

304 *4.2 Milk fatty acid profile*

305 *4.2.1 Organic milk*

The finding that ORG milk contains similar concentrations of SFA to CON milk is consistent with previous UK retail (Butler, et al., 2011b) and farm (Ellis, et al., 2006; Stergiadis, et al., 2012) surveys. In the current study, ORG milk had lower SFA concentrations during the period when cows are turned out to graze (spring) in UK, but also in October and December. However,

310 the principal SFA in milk fat, C16:0, which is considered undesirable in human nutrition, was 311 found in lowest concentrations in ORG milk, in line with a previous UK retail study (Butler, et 312 al., 2011b). Milk C16:0 originates both from diet and endogenous synthesis by the mammary 313 gland (Chilliard, et al., 2007). Concentration of C16:0 in ORG milk may reflect that lipids in 314 ORG cow diets (rich in fresh grass and with a high forage:concentrate ratio) may contain 315 proportionately less C16:0 than conventional cow diets; and/or cause a potential modification 316 in the amounts of the C16:0 substrates in the rumen, which are used for its *de novo* synthesis 317 in the mammary gland (Chilliard, et al., 2007). Multivariate analyses in other studies 318 (Stergiadis, et al., 2015a; Stergiadis, et al., 2015b; Stergiadis, et al., 2012) have shown a 319 negative relationship between fresh forage intake (which is expected to be higher in ORG 320 systems), and milk C16:0 concentration. Conversely, other studies reported a higher 16:0 321 concentration (USA; (O'Donnell, Spatny, Vicini, & Bauman, 2010)) or no difference (the 322 Netherlands; (Capuano, et al., 2015)) between organic and conventional milk. As cow diet is a 323 major driver of milk FA profile (Stergiadis, et al., 2015a; Stergiadis, et al., 2015b; Stergiadis, 324 et al., 2012), these discrepancies probably arise due to variations in diets used in different 325 countries as result of contrasting soil, climate, tradition and legislation (Butler, et al., 2011a). 326 Milk MUFA concentrations, mainly characterised by the nutritionally-desirable OA which 327 represented 73.1% of total MUFA in the present work, were similar between ORG and CON 328 milk, thus agreeing with previous UK studies (Butler, et al., 2011b; Stergiadis, et al., 2012). 329 The concentrations of OA may be affected by dietary supply, extent of rumen biohydrogenation (RBH), but also its synthesis from C18:0 by Δ^9 -desaturase action in the mammary gland 330 (Destaillats, Trottier, Galvez, & Angers, 2005). Differences between ORG and CON systems 331 332 in these factors may have been either small or showing a counteracting effect, thus resulting in 333 similar concentrations of OA in ORG and CON milk.

The finding that ORG milk contains more VA and RA than CON milk is in line with previous 334 335 retail surveys (Butler, et al., 2011b; Capuano, et al., 2015; O'Donnell, et al., 2010), although 336 other UK farm surveys have shown either no differences (Ellis, et al., 2006) or a significant 337 difference only when ORG was compared with intensive CON production systems during summer (Stergiadis, et al., 2012). VA is an intermediate product of RBH of dietary PUFA, and 338 339 in particular ALNA (Chilliard, et al., 2007; Destaillats, et al., 2005). Upon absorption and 340 delivery to the mammary gland, part of VA is converted to RA, under the effect of mammary 341 Δ^9 -desaturase (Chilliard, et al., 2007; Destaillats, et al., 2005). Therefore, cow diets rich in 342 ALNA, such as those of high pasture intake characterizing ORG systems, will increase the 343 availability of substrate for higher VA production in the rumen and the subsequent RA 344 synthesis in the mammary gland (Chilliard, et al., 2007; Elgersma, 2015). Pasture intake is 345 potentially the main driver for milk VA and RA concentrations in the current study because 346 differences in RA concentrations between ORG and CON were not significant during the 347 period that pasture was not available in the UK (December to February).

348 The higher concentrations of ALNA, EPA and DPA, and consequently the n-3, in ORG than 349 in CON milk is in line with other retail surveys (Butler, et al., 2011b; Capuano, et al., 2015; 350 O'Donnell, et al., 2010). However, for DPA, retail surveys may show only a tendency for higher 351 concentrations in ORG (Butler, et al., 2011b) or inconsistent results between summer and 352 winter (Capuano, et al., 2015). In a previous UK farm survey, ORG milk contained more DPA 353 only when compared with milk from highly-intensive CON production systems (Stergiadis, et 354 al., 2012). The higher concentrations of ALNA in ORG milk were observed throughout the year. In summer, milk ALNA concentrations can be enhanced by (i) higher pasture intake in 355 356 ORG systems, due to the higher ALNA supply from fresh forage than conserved forage and/or 357 concentrates (Elgersma, 2015), and (ii) clover contribution to the grazing swards, potentially 358 due to the increased transfer rates of dietary ALNA when fresh clover substitutes fresh grass

359 in cow diets (Stergiadis, et al., 2018). In winter, grass/clover silage commonly used in ORG 360 systems when pasture is not available, has been found to increase ALNA concentrations when 361 compared with grass or grass/maize silage (Dewhurst, Fisher, Tweed, & Wilkins, 2003; 362 Wiking, Theil, Nielsen, & Sorensen, 2010), commonly used in the CON systems. This may be explained by the slower DM degradation (Dewhurst, Evans, Scollan, Moorby, Merry, & 363 364 Wilkins, 2003) and rates of RBH of clover compared with grass (Lejonklev, Storm, Larsen, 365 Mortensen, & Weisbjerg, 2013), which may increase rumen passage rates, reduce RBH of 366 ALNA, and eventually increase transfer rates of dietary ALNA to milk. In humans and animals, 367 including cattle, ALNA is used as substrate for the synthesis of EPA and DPA by various enzymes, including elongases, Δ^5 -desaturase and Δ^6 -desaturase (Barcelo-Goblijn, et al., 2009). 368 369 Therefore, a higher supply of ALNA in the mammary gland of cows in the ORG systems, as a 370 consequence of the combined effect of high pasture and clover intake, may have increased the 371 substrate available for EPA and DPA synthesis.

Factors affecting Δ^9 -desaturase activity, potentially including, animal genetics, production 372 373 stage and diet, are not well understood. Transition from winter diets (relying on conserved 374 forages and concentrates) to summer diets (including substantial amounts of pasture) is likely 375 to (i) alter the supply of FA, transferred directly to milk or acting as substrate for conversion 376 to other milk FA, and (ii) exert metabolic changes in the rumen and/or the cow, possibly 377 altering the activity of nutritionally-sensitive enzymes responsible for *de novo* synthesis of 378 short and medium chain SFA or desaturation of FA in the mammary gland (Lock & 379 Garnsworthy, 2003). Higher fresh grass intakes, which increase water-soluble carbohydrate intakes and the subsequent insulin levels, may increase Δ^9 -desaturase activity (Lock, et al., 380 381 2003), but the opposite was observed in the ORG milk in the current study. ORG dairy herds 382 in the UK extensively use crossbred cows (most typically crosses between Holstein, 383 British/New Zealand Friesian, Jersey and/or Scandinavian Red) while CON herds rely almost entirely in Holstein cows. Provided the well documented substantial effect of breed and individual differences on Δ^9 -desaturase activity, such as the lower activity in Jersey and Holstein × Jersey crosses than in pure Holstein cows (Palladino, Buckley, Prendiville, Murphy, Callan, & Kenny, 2010), it is possible that the effect of breed may have overridden any potential effect of diet.

389

4.2.2 Potential impact of organic milk on the fatty acid intakes of UK consumers

390 Department of Health and Social Care in the UK has set a maximum reference nutrient intakes 391 (RNI) of SFA and trans FA at 11% and 2% of food energy intake, respectively, and 392 recommended intakes of PUFA at 6.5% of food energy intake (Department of Health, 1991). 393 According to the current average requirements for energy for the different age groups and 394 genders, and an energy content of fat at 37 kJ/g (SACN, 2011), under the current dairy fat 395 intakes in the UK (Bates, et al., 2014), milk fat contributes 24-37% of the maximum 396 recommended intakes of SFA intakes in adults (being maximum for men over 65 yo, which is 397 the group with the maximum milk fat intakes) and 20-43% of the maximum recommended 398 intakes in children. Overall intakes of SFA will not be affected by switching between milks 399 from different production systems because the effect of production system on milk SFA 400 concentrations was not significant. Even within months that the difference between ORG and 401 CON was maximised and was statistically significant (April; milk SFA concentrations of 680.4 402 and 705.2 g/kg FA respectively), consumption of ORG milk would minimally reduce SFA 403 intake by 0.250 g/d in men over 65 yo (contributing 36.5% RNI than 37.4% RNI) and 0.214 404 g/d in women (contributing 36.7% RNI than 37.6% RNI), when compared with CON milk. 405 Previous work has suggested that in order to benefit public health and reduce health care costs, 406 a reduction on milk SFA should be at the level of 150 g/kg FA (Kliem, et al., 2013). According 407 to these, any potential public health impact from the occasional reduction in milk SFA intakes 408 via consumption of ORG milk cannot be claimed by the results of the present work.

409 Previous work has highlighted that some milk SFA included in the calculations of total milk 410 SFA (e.g. C4:0, C8:0, C10:0), may have beneficial implications to human health (Haug, et al., 411 2007). Therefore, focusing on SFA which, if excessively consumed, increase CVD risk (C12:0, 412 C14:0 and C16:0; (FAO, 2010)), may be more accurate when discussing milk fat profile. 413 According to the results of this survey and current dairy food consumption in the UK (Bates, 414 et al., 2014), switching from CON to ORG milk fat will reduce consumption of C16:0 by 0.206 415 g/d (from 4.0 to 3.8 g/d) in children 4-10 yo, by 0.164 g/d (from 3.2 to 3.0 g/d) in children 11-416 18 yo, by 181 mg/d (from 3.5 to 3.3 g/d) in adults 19-64 yo, and by 243 mg/d (from 4.7 to 4.5 417 g/d) in adults over 65 yo. Nutritional recommendations are provided only for total SFA 418 (Department of Health, 1991), so it is not possible to estimate C16:0 contribution from dairy 419 foods.

420 According to the results of the present survey and the current dairy fat intakes in the UK (Bates, 421 et al., 2014), switching from CON to ORG milk fat will increase the contribution of PUFA, 422 expressed as % RNI, from 4.0% to 4.6% in children 4-10 yo, from 2.1% to 2.7% in children 423 11-18 yo, from 2.4% to 2.9% of adults 19-64 yo, and from 3.6% to 4.4% in adults over 65 yo. 424 The maximum benefit to PUFA intakes from switching from CON to ORG milk was observed 425 in May (milk PUFA concentrations 52.1 g/kg FA for ORG and 43.7 g/kg FA for CON) when 426 intakes were increased by 131 mg/d in men over 65 yo (contributing 4.0% RNI than 4.7% 427 RNI), and 111 mg/d in women over 65 yo (contributing 4.0% RNI than 4.8% RNI). Although 428 the potential health effects by these changes have not been investigated in the current study, 429 these differences are rather small and are unlikely to be associated with reduced chronic disease 430 risk within a whole diet.

A switch to ORG milk would increase the intakes of *trans* FA, expressed as % RNI, from
10.4% to 11.8% in children 4-10 yo, from 6.0% to 6.8% in children 11-18 yo, from 6.7% to
7.6% of adults 19-64 yo, and from 10.1% to 11.4% in adults over 65 yo. However, in this study

434 43% of *trans* FA in milk fat was VA, which is associated with positive effects in human health
435 (Field, et al., 2009). If concentrations of VA are not included in the calculation of *trans* FA,
436 the overall intakes will not be affected by switching between milks from different production
437 systems because the effect of production system on milk *trans* FA (excluding VA)
438 concentrations was not statistically significant.

439 The European Food and Safety Authority set the adequate intake (ADI) for ALNA at 0.5% 440 energy intake (EFSA, 2010). According to the results of the present survey and the current 441 dairy fat intakes in the UK (Bates, et al., 2014), switching from CON to ORG milk fat will 442 increase the intakes of ALNA from 5.8% to 8.9% ADI in children 4-10 yo, from 3.0% to 4.6% 443 ADI in children 11-18 yo, from 3.4% to 5.2% ADI in adults 19-64 yo and from 5.2% to 7.9% 444 ADI in adults over 65 yo. The maximum potential benefit in intakes of ALNA was observed 445 in September (milk ALNA concentrations 7.8 g/kg FA for ORG and 4.7 g/kg FA for CON) 446 when intakes could have been increased by 48 mg/d in males over 65 yo (contributing 9.2% 447 RNI than 5.6% ADI), and 40.8 mg/d in females over 65 yo (contributing 9.3% ADI than 5.6% 448 ADI), when compared with CON milk.

449 The European Food and Safety Authority set the adequate intake (ADI) for EPA+DHA of 250 mg/d in adults and children over 24 months old (EFSA, 2010). According to the results of this 450 451 survey and the current milk intakes in the UK (Bates, et al., 2014), switching from CON to 452 ORG milk fat will increase the intakes of EPA+DHA from 2.7% ADI to 3.6% ADI in children 453 4-10 yo, from 2.1% ADI to 2.9% ADI in children 11-18 yo, from 2.7% to 3.2% ADI in adults 454 19-64 yo, and from 3.2% to 4.3% ADI in adults over 65 yo. The maximum potential benefit in 455 intakes of EPA+DHA was observed in June (milk EPA+DHA concentrations 0.84 g/kg FA for 456 ORG and 0.78 g/kg FA for CON) when intakes could have been increased by 4.0 mg/d in males 457 over 65 yo (contributing 5.2% ADI than 3.6% ADI), and 3.4 mg/d in females over 65 yo (contributing 4.4% ADI than 3.1% ADI), when compared with CON milk. Part of dietary 458

ALNA in humans is converted to EPA, DPA and DHA, but with conversion efficiencies being
lower than 0.2% (Barcelo-Goblijn, et al., 2009). An additional supply of EPA+DHA may
therefore be expected, as a result of milk ALNA metabolism, but the low conversion
efficiencies, will result in minimal additional supply of EPA+DHA from ORG than CON milk
(less than 1 mg/d).

464 According to the results of this survey and the current milk intakes in the UK (Bates, et al., 2014), switching from CON to ORG milk fat will increase RA intake by 24.7 mg/d (from 71.5 465 466 to 96.2 mg/d) in children 4-10 yo, by 19.6 mg/d (from 56.9 to 76.5 mg/d) in children 11-18 yo, 467 by 21.6 mg/d (from 62.6 to 84.2 mg/d) in adults 19-64 yo, and by 29.1 mg/d (from 84.2 to 113.3 g/d) in adults over 65 yo. Given that on average 19 % of VA is also endogenously 468 469 converted to RA in the human body (Field, et al., 2009), consumption of ORG milk may also 470 increase the available RA via endogenous synthesis, because of its higher VA concentrations. 471 RA has been previously associated with a number of health benefits in humans (Dilzer, et al., 472 2012). However, nutritional recommendations for RA are not currently developed, so it is not 473 possible to estimate RA contribution from dairy foods.

474 Overall, organic milk could be considered desirable from a human nutrition perspective, in terms of lower SFA and higher PUFA, n-3, ALNA, RA contents, as well as a higher ratio of n-475 476 3/n-6 and lower TI (which however refer to the whole diet), thus aligning with current 477 nutritional recommendations (EFSA, 2010; FAO, 2010; Givens, 2017). A switch from CON to 478 ORG milk will influence the intakes of these FA, but any implications for human health cannot 479 be drawn in the present study, because these changed intakes are relatively small. The nutritional recommendations (Department of Health, 1991; EFSA, 2010; SACN, 2011) for 480 481 individual FA or FA groups refer to the total diet rather than a single food, and although the 482 current study estimates the potential changes on FA intakes from dairy products, any potential 483 effect on human health will be influenced by FA intakes from other foods. Current evidence 484 suggests there is no positive association between intake of milk and dairy products and the risk 485 of CVD and type-2 diabetes, while consumption of cheese and yoghurt also showed a negative 486 association (Kliem, et al., 2011; Thorning, et al., 2017). This may be an effect of interactions 487 between milk matrix components and enhances the necessity for future research on the effect 488 of milk and dairy products as whole foods, alongside investigations of the nutritional role of 489 their individual components (Kliem, et al., 2011; Thorning, et al., 2017).

490

4.2.3 Free-range milk

491 The lack of differences between the two conventional systems (CON, FR), one representing 492 typical CON UK dairy management and the other FR practices, is in line with recent results 493 from the Netherlands (Capuano, et al., 2015). Provided that diet, and in particular fresh grass 494 intake, is the major driver for milk FA profiles (Elgersma, 2015; Stergiadis, et al., 2015b; 495 Stergiadis, et al., 2012), the similarities between CON and FR milk may potentially reflect 496 small differences in cow nutrition between the two production systems. Previous studies that 497 showed substantial differences in the FA profile of milk between pasture-based, ORG and CON 498 milk at farm level had investigated low-input pasture-based farms where the average pasture 499 intake was more than 95% of total cow diet (Butler, et al., 2008; Stergiadis, et al., 2015b). 500 Provided that FR certification refers to access to outdoors/pasture but without setting minimum 501 requirements for pasture intake, lower contribution of pasture in cow diet when compared with 502 the low-input farms assessed in other studies (Butler, et al., 2008; Stergiadis, et al., 2015b), 503 may potentially explain the lack of effect on milk FA profile. In addition, allowing cows access 504 to pasture for six months is a typical practice in UK CON dairy systems, although maybe at a 505 lesser extent than in FR systems, and this may further contribute to the similarities in cow diets 506 and the subsequent FA profile between CON and FR milk. Other potential reasons for this 507 observations that have been provided in the Netherlands was (i) that not all farms provide 508 access to pasture the same time, thus diluting the effect of pasture intake when bulking milk at the dairy, and (ii) the contribution of fresh-cut grass in indoor CON systems, which wouldreduce the differences between grazing and indoor cows (Capuano, et al., 2015).

511 Despite the overall similarities between FR and CON milk, significant differences were 512 observed in specific months within the grazing season, potentially as an effect of the higher 513 pasture intake in FR herds than in CON herds during these months. For example, FR milk had 514 a more preferable FA profile than CON milk in May, August and/or September, by containing 515 less of the nutritionally-undesirable C16:0, more of the nutritionally-desirable ALNA and RA 516 and by having a higher n-3/n-6 ratio; thus representing a favourable effect of substituting SFA 517 with MUFA and PUFA, in line with current recommendations (EFSA, 2010; FAO, 2010; 518 Givens, 2017). FR milk also contained less SFA and more MUFA in May, but this relationship 519 was reversed in February. The lower PUFA in FR than CON milk during the indoor period 520 mainly reflects the lower concentrations of n-6 and LA, which may be a result of lower use of 521 maize silage, a main driver for milk n-6 concentrations (Chilliard, et al., 2007; Stergiadis, et 522 al., 2015b), in FR than in CON herds. Inconsistency in the differences between FR and CON 523 milk throughout the year may have also been a consequence of the small number of farms 524 contributing the FR milk at the dairies. Because of that, the effect of cow diet in individual FR 525 farms has a proportionately higher impact to the final product.

526

4.2.4 Seasonal variation

The effect of season on the FA profile of UK retail milk has been extensively investigated in other surveys (Butler, et al., 2011b; Kliem, et al., 2013). In agreement with the present work, these previous studies highlighted that concentrations of milk total SFA and individual SFA (C12:0, C14:0, C16:0) are lower, and those of VA, OA, RA, ALNA, EPA, n-3, PUFA, EPA are higher, during the grazing season. In the UK, in dairy systems where cows have access to pasture, and in line with local climate, animals are housed in winter (December-February), turn out to pasture at March-April, have a period where grazing is maximised (May-September) and 534 then gradually return to indoor diets rich in conserved forages and concentrates late in autumn 535 (October-November). Fresh grass is rich in PUFA, including ALNA, and therefore its 536 potentially higher dietary intakes may result in (i) higher milk ALNA concentrations, via direct 537 transfer to milk (Elgersma, 2015), (ii) higher OA, via rumen synthesis of C18:0 (end product 538 of RBH of FA) and the subsequent increased C18:0 supply in the mammary gland for OA 539 synthesis (Destaillats, et al., 2005), (iii) higher milk VA and RA concentrations, via rumen 540 synthesis of VA and the subsequent increase in VA supply in the mammary gland for RA 541 synthesis (Chilliard, et al., 2007; Destaillats, et al., 2005), (iv) higher EPA and DPA, potentially 542 via the higher supply of their substrate ALNA (Barcelo-Goblijn, et al., 2009), and eventually 543 (v) lower total SFA and individual SFA (C12:0, C14:0, C16:0) concentrations. Given that 60% 544 of the samples in the present study were from ORG and FR farms, which are expected to 545 provide access to pasture during the grazing season, the effect of pasture intake appears to be 546 the most possible explanation of the seasonal variation. The fact that pasture intake is among the strongest drivers of milk FA profiles in the UK, has been previously demonstrated in 547 548 multivariate redundancy analyses in data collected from dairy farms (Stergiadis, et al., 2015b; 549 Stergiadis, et al., 2012).

550

5. Conclusions

551 Organic retail milk showed a more favourable FA profile, containing more nutritionally-552 desirable FA, less C16:0, and a higher n-3/n-6 ratio than conventional milk. During specific 553 months, organic milk also had less total SFA. The free-range milk had similar FA profile to 554 conventional milk, but contained less SFA (including C16:0) and more ALNA and RA, in 555 specific months within the outdoor/grazing season. Although background information on dairy 556 management practices was not available, it is highly likely that differences in milk FA profiles 557 resulted from contrasting cow diets, and in particular the intakes of pasture, clover and 558 forage:concentrate ratio. Based on measured milk FA profiles across the production systems, dairy fat contributes approximately one-third of the maximum recommended intake of SFA in adult consumer diets. Consuming organic dairy products would increase intakes of nutritionally-desirable PUFA, and reduce consumption of nutritionally-undesirable SFA. However, when compared with conventional milk in terms of daily recommended intakes of these FA, there would be relatively little difference. Therefore, any implications to human health cannot be drawn from the present study.

565

Acknowledgements

The authors gratefully acknowledge financial support from the University of Reading. CBB was in receipt of a Science Without Borders scholarship from the Brazilian Federal Government. The work of SG in the project was funded by the Undergraduate Research Opportunities Programme of the University of Reading. Authors would also like to acknowledge the help of dairies working in collaboration with the Free Range Dairy Network with the collection and postage of free-range retail milk samples. 572 References 573 Barcelo-Goblijn, G., & Murphy, E. J. (2009). Alpha-linolenic acid and its conversion to longer 574 chain n-3 fatty acids: benefits for human health and a role in maintaining tissue n-3 575 fatty acid levels. Progress in Lipid Research, 48, 355-374. 576 Bates, B., Lennox, A., Prentice, A., Bates, C., Page, P., Nicholson, S., & Swan, G. (2014). 577 National Diet and Nutrition Survey. Results from Years 1-4 (combined) of the rolling 578 programme (2008/09-2011/12) (Revised February 2017). In 579 https://www.gov.uk/government/statistics/national-diet-and-nutrition-survey-results-580 from-years-1-to-4-combined-of-the-rolling-programme-for-2008-and-2009-to-2011-581 and-2012 (Ed.), (pp. 139): Public Health England. 582 Butler, G., Nielsen, J. H., Larsen, M. K., Rehberger, B., Stergiadis, S., Canever, A., & Leifert, 583 C. (2011a). The effects of dairy management and processing on quality characteristics 584 of milk and dairy products. NJAS Wageningen Journal of Life Science, 58, 97-102. 585 Butler, G., Nielsen, J. H., Slots, T., Seal, C. J., Eyre, M. D., Sanderson, R., & Leifert, C. (2008). 586 Fatty acid and fat-soluble antioxidant concentrations in milk from high- and low-input 587 conventional and organic systems: seasonal variation. Journal of Science of Food and 588 Agriculture, 88, 1431-1441. 589 Butler, G., Stergiadis, S., Seal, C. J., Eyre, M. D., & Leifert, C. (2011b). Fat composition of 590 organic and conventional retail milk in northeast England. Journal of Dairy Science, 591 94, 24-36. 592 Capuano, E., Gravink, R., Boerrigter-Eenling, R., & van Ruth, S. M. (2015). Fatty acid and 593 triglycerides profiling of retail organic, conventional and pasture milk: Implications for

26

health and authenticity. International Dairy Journal, 42, 58-63.

- 595 Chilliard, Y., Glasser, F., Ferlay, A., Bernard, L., Rouel, J., & Doreau, M. (2007). Diet, rumen
 596 biohydrogenation and nutritional quality of cow and goat milk fat. *European Journal*597 *of Lipid Science and Technology*, *109*, 828-855.
- 598 Chilliard, Y., Martin, C., Rouel, J., & Doreau, M. (2009). Milk fatty acids in dairy cows fed
 599 whole crude linseed, extruded linseed, or linseed oil, and their relationship with
 600 methane output. *Journal of Dairy Science*, *92*, 5199-5211.
- Department of Health. (1991). *Dietary reference values for food energy and nutrients for the United Kingdom*. London, UK: H.M. Stationery Office.
- Destaillats, F., Trottier, J. P., Galvez, J. M. G., & Angers, P. (2005). Analysis of α-linolenic
 acid biohydrogenation intermediates in milk fat with emphasis on conjugated linolenic
 acids. *Journal of Dairy Science*, 88, 3231-3239.
- Dewhurst, R. J., Evans, R. T., Scollan, N. D., Moorby, J. M., Merry, R. J., & Wilkins, R. J.
 (2003). Comparison of grass and legume silages for milk production. 2. In vivo and in
 sacco evaluations of rumen function. *Journal of Dairy Science*, 86(8), 2612-2621.
- Dewhurst, R. J., Fisher, W. J., Tweed, J. K. S., & Wilkins, R. J. (2003). Comparison of grass
 and legume silages for milk production. 1. Production responses with different levels
 of concentrate. *Journal of Dairy Science*, 86(8), 2598-2611.
- Dilzer, A., & Park, Y. (2012). Implication of Conjugated Linoleic Acid (CLA) in Human
 Health. *Critical Reviews in Food Science and Nutrition*, 52(6), 488-513.
- EFSA. (2010). Scientific Opinion on Dietary Reference Values for fats, including saturated
 fatty acids, polyunsaturated fatty acids, monounsaturated fatty acids, trans fatty acids,
 and cholesterol. *EFSA Journal*, 8, 1461.
- Elgersma, A. (2015). Grazing increases the unsaturated fatty acid concentration of milk from
 grass-fed cows: A review of the contributing factors, challenges and future
 perspectives. *European Journal of Lipid Science and Technology*, *117*, 1345-1369.

- 620 Ellis, K. A., Innocent, G., Grove-White, D., Cripps, P., McLean, W. G., Howard, C. V., &
- Mihm, M. (2006). Comparing the fatty acid composition of organic and conventional
 milk. *Journal of Dairy Science*, 89, 1938-1950.
- FAO. (2010). *Fats and fatty acids in human nutrition Report of an expert consultation*. Rome,
 Italy: Food and Agriculture Organization of the United Nations.
- Field, C. J., Blewett, H. H., Proctor, S., & Vine, D. (2009). Human health benefits of vaccenic
 acid. *Applied Physiology, Nutrition, and Metabolism, 34*, 979-991.
- Gilmour, A. R., Thompson, R., & Cullis, B. R. (1995). Average information REML: an
 efficient algorithm for variance parameter estimation in linear-mixed models. *Biometrics*, *51*, 1440-1450.
- Givens, D. I. (2017). Saturated fats, dairy foods and health: A curious paradox? *Nutrition Bulletin*, 42, 274-282.
- Haug, A., Hostmark, A. T., & Harstad, O. M. (2007). Bovine milk in human nutrition. *Lipids in Health and Disease*, *6*, 25.
- 634 Kay, J. K., Mackle, T. R., Auldist, M. J., Thomson, N. A., & Bauman, D. E. (2004).
- Endogenous synthesis of cis-9, trans-11 conjugated linoleic acid in dairy cows fed fresh
 pasture. *Journal of Dairy Science*, 87, 369-378.
- Kliem, K. E., & Givens, D. I. (2011). Dairy products in the food production chain: their impact
 on health. *Annual Review of Food Science and Technology*, *2*, 21-36.
- Kliem, K. E., & Shingfield, K. J. (2016). Manipulation of milk fatty acid composition in
 lactating cows: Opportunities and challenges. *European Journal of Lipid Science and Technology*, *118*, 1661-1683.
- Kliem, K. E., Shingfield, K. J., Livingstone, K. M., & Givens, D. I. (2013). Seasonal variation
 in the fatty acid composition of milk available at retail in the United Kingdom and
 implications for dietary intake. *Food Chemistry*, *141*, 274-281.

- Lejonklev, J., Storm, A. C., Larsen, M. K., Mortensen, G., & Weisbjerg, M. R. (2013).
 Differences in rate of ruminal hydrogenation of C18 fatty acids in clover and ryegrass. *Animal*, 7(10), 1607-1613.
- 648 Lock, A., & Garnsworthy, P. (2003). Seasonal variation in milk conjugated linoleic acid and 649 Δ^9 -desaturase activity in dairy cows. *Livestock Production Science*, *79*, 47-59.
- 650 O'Donnell, A. M., Spatny, K. P., Vicini, J. L., & Bauman, D. E. (2010). Survey of the fatty
- acid composition of retail milk differing in label claims based on production
 management practices. *J Dairy Sci*, 93, 1918-1925.
- Palladino, R. A., Buckley, F., Prendiville, R., Murphy, J. J., Callan, J., & Kenny, D. A. (2010).
- A comparison between Holstein-Friesian and Jersey dairy cows and their F1 hybrid on
 milk fatty acid composition under grazing conditions. *Journal of Dairy Science*, *93*,
 2176-2184.
- 657 Pereira, P. C. (2014). Milk nutritional composition and its role in human health. *Nutrition*, *30*,
 658 619-627.
- 659 SACN. (2011). Dietary reference values for energy. In Scientific Advisory Committee in
 660 Nutrition (Ed.)). London, UK: Crown Copyright.
- 661 Srednicka-Tober, D., Baranski, M., Seal, C., Sanderson, R., Benbrook, C., Steinshamn, H.,
- 662 Gromadzka-Ostrowska, J., Rembialkowska, E., Skwarlo-Sonta, K., Eyre, M., Cozzi,
- 663 G., Larsen, M. K., Jordon, T., Niggli, U., Sakowski, T., Calder, P. C., Burdge, G. C.,
- 664 Sotiraki, S., Stefanakis, A., Yolcu, H., Stergiadis, S., Chatzidimitriou, E., Butler, G.,
- 665 Stewart, G., & Leifert, C. (2016). Composition differences between organic and
- 666 conventional meat: a systematic literature review and meta-analysis. *British Journal of* 667 *Nutrition*, *115*, 994-1011.
- 668 Stergiadis, S., Bieber, A., Franceschin, E., Isensee, A., Eyre, M. D., Maurer, V.,
 669 Chatzidimitriou, E., Cozzi, G., Bapst, B., Stewart, G., Gordon, A., & Butler, G. (2015a).

- 670 Impact of Brown Swiss genetics on milk quality from low-input herds in Switzerland:
 671 interactions with grazing intake and pasture type. *Food Chemistry*, 175, 609-618.
- Stergiadis, S., Hynes, D. N., Thomson, A. L., Kliem, K. E., Berlitz, C. G. B., Gunal, M., &
 Yan, T. (2018). Effect of substituting fresh-cut perennial ryegrass with fresh-cut white
 clover on bovine milk fatty acid profile. *Journal of the Science of Food and Agriculture, under review.*
- Stergiadis, S., Leifert, C., Seal, C. J., Eyre, M. D., Larsen, M. K., Slots, T., Nielsen, J. H., &
 Butler, G. (2015b). A 2-year study on milk quality from three pasture-based dairy
 systems of contrasting production intensities in Wales. *Journal of Agricultural Science*, *153*(4), 708-731.
- Stergiadis, S., Leifert, C., Seal, C. J., Eyre, M. D., Nielsen, J. H., Larsen, M. K., Slots, T.,
 Steinshamn, H., & Butler, G. (2012). Effect of feeding intensity and milking system on
 nutritionally relevant milk components in dairy farming systems in the north east of
 England. *Journal of Agricultural and Food Chemistry*, 60, 7270-7281.
- 684 Swanson, D., Block, R., & Mousa, S. (2012). Omega-3 fatty acids EPA and DHA: health
 685 benefits throughout life. *Advances in Nutrition*, *3*, 1-7.
- 686 Thorning, T. K., Bertram, H. C., Bonjour, J.-P., de Groot, L., Dupont, D., Feeney, E., Ipsen,
- 687 R., Lecerf, J. M., Mackie, A., McKinley, M. C., Michalski, M.-C., Rémond, D., Risérus,
- 688 U., Soedamah-Muthu, S. S., Tholstrup, T., Weaver, C., Astrup, A., & Givens, I. (2017).

689 Whole dairy matrix or single nutrients in assessment of health effects: current evidence 690 and knowledge gaps. *The American Journal of Clinical Nutrition*, *105*(5), 1033-1045.

- 691 Ulberth, F., Gabernig, R. G., & Schrammel, F. (1999). Flame-ionization detector response to
- 692 methyl, ethyl, propyl and butyl esters of fatty acids. Journal of the American Oil
- 693 *Chemists Society*, 76, 263-266.

- 694 Wiking, L., Theil, P. K., Nielsen, J. H., & Sorensen, M. T. (2010). Effect of grazing fresh
- 695 legumes or feeding silage on fatty acids and enzymes involved in the synthesis of milk
- fat in dairy cows. *Journal of Dairy Research*, 77(3), 337-342.

Table 1

Means (and average SE) and ANOVA P-values for the effect of production system (conventional, CON; organic, ORG; free-range, FR) on the basic composition and fatty acid (FA) profile (g/kg total FA) of milk collected from retail outlets during the year

	CON	ORG	FR		ANOVA
Parameters assessed	n=48	n=48	$n=24^{a}$	SE	P-values ^b
Basic composition					-
Fat (g/kg milk)	34.9 ^B	40.0 ^A	37.0 ^B	0.60	***
Protein (g/kg milk)	32.7	32.4	32.7	0.49	ns
Casein (g/kg milk)	25.5	25.2	25.4	0.41	ns
Lactose (g/kg milk)	45.2 ^A	44.8 ^B	45.2 ^A	0.10	*
SCC ^{c} (x 10 ³ /ml milk)	38	137	58	30	ns
Individual FA					
SFA ^d					
C12:0	33.4	32.4	34.8	0.67	ns
C14:0	111	114	114	1.2	ns
C16:0	331 ^A	314 ^B	325 ^A	2.0	***
C18:0	99.5	106.1	103.3	2.36	ns
MUFA ^e					
VA	12.2 ^B	17 1 ^A	12.3 ^B	0.68	**
0A	200	197	199	2.6	ns
PUFA f	200	177	177	2.0	115
LA	17.1	16.6	153	0.75	ns
RA	5 91 ^B	7 95 ^A	6.06 ^B	0 259	***
ALNA	4 39 ^B	6 71 ^A	4.76^{B}	0.124	***
FPΔ	0 484 ^B	0.674 ^A	0 551 ^B	0.0226	***
DPA	0.404	1.024 ^A	0.834 ^B	0.0220	***
DHA	0.067	0.078	0.054	0.0279	ns
FA groups	0.007	0.070	0.000	0.0100	115
SFA	688	684	692	2.6	ns
MUFA	273	272	270	2.0	ns
cis MUFA ^g	243	272	240	2.1	ns
trans MUFA h	31 0 ^B	35 0 ^A	30 0 ^B	0.84	***
PLIFA	39.7 ^B	44 7 ^A	38 3 ^B	0.01	***
cis PUFA ⁱ	25.9 ^B	28 0 ^A	$24 4^{B}$	0.42	*
trans PLIFA j	0 34 ^B	0.52 ^A	0 37 ^B	0.033	*
$cis/trans + trans/cis PUFA^{k}$	13 4 ^B	16 1 ^A	13.5 ^B	0.035	**
$n_{-3}l$	7 93 ^B	11 60 ^A	8 66 ^B	0.42	***
$n-6^{m}$	20.9	20.0	18.9	0.332	ns
n-3/n-6	0.39 ^B	0.59A	0.46^{B}	0.031	**
trans $\mathbf{F} \mathbf{\Delta}^n$	3 13 ^B	3 55A	3 03 ^B	0.031	**
trans FA (ever VA)	1 00	1.84	1.81	0.000	ne
Indices	1.90	1.04	1.01	0.043	115
Human health related					
	2.60	2 56	2 69	0.047	ne
	2.00 3.13A	2.50 2.80B	2.09 3.15 ^A	0.047	***
11^{4}	5.15	2.09	5.15	0.027	
$\Delta - aestimate activity$	0.207	0.206	0.206	0.0026	20
ΔI $C14 \cdot 1/C14 \cdot 0$	0.297	0.290 0.080B	0.290	0.0020	11S **
$C_{14.1}/C_{14.0}$	0.064	0.000°	0.004	0.0007	*
OA/C18.0	2.036°	1.050°	1.020A	0.0000	*
	2.011	0.460	0.501	0.03/1	*
KA/VA	0.491	0.409	0.301	0.0085	

^{*a*} In September, there was a missing sample of free-range milk in the analysis of basic composition and the mean on this set of parameters was calculated from 23 samples

^b Significances were declared at ***, P < 0.001; **, P < 0.01; *, P < 0.05; \neq , 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 Fisher's Least Significant

Difference test (P < 0.05)

^c Somatic cell count

^d Saturated FA: C4:0, C5:0, C6:0, C7:0, C8:0, C9:0, C10:0, C11:0, C12:0, C13:0, C13:0*iso*, C13:0*anteiso*, C13:0, C14:0*iso*, C14:0, C15:0*anteiso*, C15:0, C16:0*iso*, C16:0, C17:0*iso*, C17:0, C18:0*iso*, C18:0, C20:0, C22:0, C24:0

^{*e*} Monounsaturated FA (MUFA): *c*9 C10:1, *c*10 C11:1, *c*9 C12:1, *c*9 C13:1, *t*9 14:1, *c*9 C14:1, *c*10 C15:1, *t*7+t8 C16:1, *t*9 C16:1, *t*11+*t*12+*t*13 C16:1, *c*9 C16:1 (co-elutes with C17:0*anteiso*), *c*11 C16:1, *c*13 C16:1, *t*10 C17:1, *c*9 C17:1, *t*4 C18:1, *t*5 C18:1, *t*6+*t*7+*t*8 C18:1, *t*9 C18:1, *t*10 C18:1, *t*11 C18:1 (VA), *c*6+*t*12 C18:1, *c*9 C18:1 (OA), *t*15 C18:1, *c*11 C18:1, *c*12 C18:1, *c*13 C18:1, *t*16 + *c*14 C18:1, *c*15 C18:1 (co-elutes with C19:0), *c*16 C18:1, *c*5 C20:1, *c*8 C20:1, *c*11 C20:1, *c*13 C22:1, *c*15 C24:1

^f Polyunsaturated FA (PUFA): *t*11*t*15 C18:2, *t*9*t*12 C18:2, *c*9*t*13 C18:2, *c*10*t*14 C18:2, *c*9*t*14 C18:2, *c*9*t*12 C18:2, *t*9*c*12 C18:2, *t*9*c*12 C18:2, *t*9*c*12 C18:2, *c*9*c*12 C18:2 (LA), *t*12*c*15 C18:2 (co-elutes with *c*9 C19:1), *c*6*c*9*c*12 C18:3, *c*9*c*12*c*15 C18:3 (ALNA), *c*9*c*11 C18:2 conjugated (RA) (co-elutes with *t*7*c*9+*t*8*c*10+*t*6*c*8 C18:2), other C18:2 conjugated FA of unknown isomerism, *c*11*c*14 C20:2, *c*8*c*11*c*14 C20:3, *c*11*c*14*c*17 C20:3, *c*5*c*8*c*11*c*14 C20:4, *c*13*c*16 C22:2, EPA, *c*13*c*16*c*19 C22:3, DPA, DHA

^{*g*} *cis* MUFA: *c*9 C10:1, *c*10 C11:1, *c*9 C12:1, *c*9 C13:1, *c*9 C14:1, *c*10 C15:1, *c*9 C16:1 (coelutes with C17:0*anteiso*), *c*11 C16:1, *c*13 C16:1, *c*9 C17:1, *c*6 C18:1 (co-elutes with *t*12 C18:1), OA, *c*11 C18:1, *c*12 C18:1, *c*13 C18:1, *c*14 C18:1 (co-elutes with *t*16 C18:1), *c*15 C18:1 (co-elutes with C19:0), *c*16 C18:1, *c*5 C20:1, *c*8 C20:1, *c*11 C20:1, *c*13 C22:1, *c*15 C24:1

^{*h*} *trans* MUFA: *t*9 14:1, *t*7+t8 C16:1, *t*9 C16:1, *t*11+*t*12+*t*13 C16:1, *t*10 C17:1, *t*4 C18:1, *t*5 C18:1, *t*6+*t*7+*t*8 C18:1, *t*9 C18:1, *t*10 C18:1, VA, *t*12 C18:1 co-elutes with *c*6 C18:1), *t*15 C18:1, *t*16 C18:1 (co-elutes with *c*14 C18:1)

^{*i*} *cis* PUFA: LA, ALNA, c11c14 C20:2, c8c11c14 C20:3, c11c14c17 C20:3, c5c8c11c14 C20:4, c13c16 C22:2, EPA, c13c16c19 C22:3, DPA, DHA

^{*j*} trans PUFA: t11t15 C18:2, t9t12 C18:2

^k cis/trans + trans/cis PUFA: c9t13 C18:2, c10t14 C18:2, c9t14 C18:2, c9t12 C18:2, t9c12 C18:2, t11c15 C18:2, t12c15 C18:2 (co-elutes with c9 C19:1), RA (co-elutes with t7c9+t8c10+t6c8 C18:2), other C18:2 conjugated FA of unknown isomerism

¹ omega-3 PUFA (n-3): *t*11*t*15 C18:2, *t*11*c*15 C18:2, *t*12*c*15 C18:2 (co-elutes with *c*9 C19:1), ALNA, *c*11*c*14*c*17 C20:3, EPA, *c*13*c*16*c*19 C22:3, DPA, DHA

^{*m*} omega-6 PUFA (n-6): *t9t*12 C18:2, *c9t*12 C18:2, *t9c*12 C18:2, LA, *c6c*9*c*12 C18:3, *c*11*c*14 C20:2, *c8c*11*c*14 C20:3, *c5c*8*c*11*c*14 C20:4, *c*13*c*16 C22:2, *c7c*10*c*13*c*16 C22:4

^{*n*} *trans* FA: *t*9 C14:1, *t*9 C16:1, *t*11+*t*12+*t*13 C16:1, *t*10 C17:1, *t*4 C18:1, *t*5 C18:1, *t*6+*t*7+*t*8 C18:1, *t*9 C18:1, *t*10 C18:1, VA, *t*12 C18:1, *t*15 C18:1, *t*16 C18:1, *t*11*t*15 C18:2, *t*9*t*12 C18:2, *t*12*t*15 C18:2

^{*o*} Atherogenicity index = (C12:0 + 4 x C14:0 + C16:0) / (MUFA + PUFA), as described in Srednicka-Tober et al. (2016)

^{*p*} Thrombogenicity index = (C14:0 + C16:0 + C18:0) / [(0.5 x MUFA) + (0.5 x n-6) + (3 x n-3) + (n-3/n-6)], as described in Srednicka-Tober et al. (2016)

 $^{r} \Delta^{9}$ -desaturase activity index = (c9 C14:1+c9 C16:1+OA+RA)/(c9 C14:1+c9 C16:1+OA+RA+C14:0+C16:0+C18:0+VA), as proposed by Kay et al. (2004)

 Table 2

 Means (and average SE) and ANOVA P-values for the effect of month on the basic composition and fatty acid (FA) profile (g/kg total FA) of milk collected from retail outlets

 during the year

							Month							
	March	April	May	June	July	August	September	October	November	December	January	February		ANOVA
Parameters assessed	n=10	n=10	n=10	n=10	n=10	n=10	$n=10^a$	n=10	n=10	n=10	n=10	n=10	SE	P-values ^b
Basic composition														
Fat (g/kg milk)	38.0 ^{AB}	38.0 ^{AB}	36.8 ^D	36.9 ^{CD}	36.6 ^D	36.9 ^D	37.2 ^{CD}	37.2 ^{BCD}	38.5 ^A	36.9 ^D	37.7 ^{ABC}	37.7 ^{ABC}	0.49	***
Protein (g/kg milk)	32.8 ^{BCE}	32.4^{CDEF}	33.2 ^B	32.2 ^{DEF}	31.8 ^F	32.1 ^{CDEF}	32.4 ^{EF}	33.2 ^B	34.0 ^A	32.1 ^{DEF}	32.5 ^{CDEF}	32.1 ^F	0.36	***
Casein (g/kg milk)	25.2 ^{CD}	25.0 ^D	25.7 ^{BC}	24.8 ^D	24.9 ^D	25.3 ^{CD}	25.3 ^{CD}	26.0 ^{AB}	26.6 ^A	24.8 ^D	25.3 ^{CD}	25.0 ^D	0.32	***
Lactose (g/kg milk)	45.5 ^{ABC}	45.6 ^A	45.7 ^{AB}	45.2 ^{BCD}	44.4^{EF}	44.9 ^{DE}	44.2 ^F	44.3 ^F	45.4^{ABCD}	44.5^{EF}	45.2 ^{CD}	45.4^{ABCD}	0.19	***
SCC c (x 10 ³)	52 ^C	48 ^C	49 ^c	30 ^C	40 ^C	38 ^C	44 ^C	38 ^c	35 ^C	219 ^{AB}	117 ^{BC}	270 ^A	48	***
Individual FA														
SFA ^d														
C12:0	35.9 ^B	34.8 ^{BCD}	34.2 ^D	31.1 ^{ef}	31.4 ^E	31.1 ^{EF}	27.5 ^G	29.9 ^F	37.7 ^A	35.7 ^{BC}	35.6 ^{BC}	34.3 ^{CD}	0.61	***
C14:0	116 ^B	113 ^C	109 ^{DE}	107 ^E	109 ^D	109 ^{DE}	105 ^F	110 ^D	123 ^A	117 ^B	117 ^B	116 ^B	1.1	***
C16:0	341 ^A	321 ^c	289 ^E	297 ^D	299 ^D	305 ^D	319 ^C	329 ^B	346 ^A	348 ^A	344 ^A	342 ^A	2.7	***
C18:0	97.2 ^D	104.0 ^C	108.1 ^B	110.1 ^{AB}	108.7 ^B	107.7 ^B	111.3 ^A	104.1 ^c	90.4 ^E	96.6 ^D	97.3 ^D	99.3 ^D	1.80	***
MUFA ^e														
VA	10.1 ^E	12.2 ^D	20.9 ^A	15.9 ^C	16.1 ^C	17.2 ^{BC}	18.6 ^B	17.7 ^{BC}	10.9 ^{DE}	9.9 ^E	10.2^{E}	10.5^{DE}	0.72	***
OA	186 ^D	195 ^C	209 ^B	212 ^B	211 ^B	211 ^B	223 ^A	213 ^B	183 ^{DE}	178^{E}	179 ^E	184^{DE}	2.5	***
PUFA ^f														
LA	17.4 ^{AB}	18.1 ^A	17.2 ^{ABC}	16.8 ^{BCD}	15.5^{EF}	16.4 ^{CDE}	16.9 ^{CD}	17.2 ^{ABC}	15.7 ^{EF}	15.6 ^F	15.5 ^F	16.1 ^{DEF}	0.55	***
ALNA	4.76 ^E	5.65 ^C	8.83 ^A	8.31 ^{AB}	8.52 ^{AB}	8.44^{AB}	8.36 ^{AB}	7.97 ^B	5.62 ^{CD}	4.76 ^E	4.92 ^{DE}	4.93 ^{DE}	0.286	***
RA	4.69 ^D	5.20 ^C	6.54 ^A	6.09 ^B	6.07 ^B	5.99 ^B	6.11 ^B	5.51 ^C	4.60 ^D	4.52 ^D	4.63 ^D	4.72 ^D	0.127	***
EPA	0.502 ^{CD}	0.513 ^{CD}	0.636 ^A	0.641^{AB}	0.659 ^A	0.644^{AB}	0.647 ^A	0.611 ^B	0.535 ^C	0.515 ^{CD}	0.488^{D}	0.484 ^D	0.0187	***
DPA	0.786^{D}	0.787^{D}	0.878^{BC}	0.895 ^{BC}	0.934^{AB}	0.919 ^{ABC}	0.980 ^A	0.960 ^{AB}	0.937 ^{AB}	0.856 ^{CD}	0.918B ^C	0.881B ^C	0.0304	***
DHA	0.059	0.058	0.067	0.066	0.066	0.059	0.132	0.079	0.061	0.070	0.063	0.058	0.0174	ns
FA groups														
SFA	708 ^B	692 ^C	661 ^{EF}	665^{DE}	668 ^D	669 ^D	657 ^F	670 ^D	711 ^{AB}	718 ^A	715 ^{AB}	709 ^{AB}	3.0	***
MUFA	254 ^E	267 ^D	292 ^{AB}	290 ^{BC}	288 ^{BC}	287 ^{BC}	297 ^A	286 ^C	251 ^{EF}	246 ^E	248^{EF}	253 ^{EF}	2.7	***
cis MUFA ^g	227 ^D	236 ^C	250 ^B	252 ^B	252 ^B	251 ^B	264 ^A	254 ^B	225 ^D	218 ^E	219 ^E	224^{DE}	2.3	***
trans MUFA ^h	27.1 ^{FG}	31.1 ^{de}	41.2 ^A	38.0 ^B	36.5 ^B	36.5 ^B	34.0 ^C	31.9 ^D	26.7 ^G	28.0 ^{FG}	28.8 ^F	29.3 ^{EF}	1.02	***
PUFA	37.9 ^E	41.1 ^D	47.2 ^A	44.7 ^{BC}	43.6 ^C	44.3 ^{BC}	45.4 ^B	44.3 ^c	37.3 ^{EF}	36.5 ^F	36.9 ^F	37.6 ^{EF}	0.51	***
cis PUFA ⁱ	26.1 ^{CD}	27.5 ^{AB}	28.3 ^A	27.3 ^{AB}	26.0 ^D	26.9 ^{BC}	27.8 ^{AB}	27.3 ^{AB}	24.4 ^F	25.1 ^{EF}	25.1 ^{EF}	25.8^{DE}	0.66	***
trans PUFA ^j	0.26 ^F	0.40^{DE}	0.69 ^A	0.49 ^{CD}	0.54^{BC}	0.56^{BC}	0.61 ^{AB}	0.52 ^{BC}	0.29^{EF}	0.20 ^F	0.26 ^F	0.18 ^F	0.047	***

<i>cis/trans</i> + <i>trans/cis</i> PUFA ^{<i>k</i>}	11.5^{EF}	13.3 ^C	18.2 ^A	16.9 ^B	17.0 ^B	16.9 ^B	17.1 ^{AB}	16.5 ^B	12.6^{DE}	11.2 ^F	11.6 ^{EF}	11.6 ^{EF}	0.55	***
n-3 ^{<i>l</i>}	7.74^{F}	8.76 ^D	11.71 ^A	10.80 ^B	11.01 ^B	10.75 ^B	10.99 ^B	10.02 ^C	8.10^{EF}	8.20^{DEF}	8.41^{DEF}	8.48^{DE}	0.293	***
n-6 ^{<i>m</i>}	20.8^{AB}	21.7 ^A	20.9 ^{ABC}	20.3 ^{BCD}	18.9 ^{DE}	19.9^{BCDE}	20.6^{ABC}	20.9^{AB}	19.0 ^E	19.4 ^{DEF}	19.3 ^{DEF}	19.9 ^{CDEF}	0.60	***
n-3/n-6	0.38 ^D	0.40^{CD}	0.57^{A}	0.54^{A}	0.59 ^A	0.55 ^A	0.55 ^A	0.49^{B}	0.43 ^C	0.43B ^C	0.44^{BC}	0.43 ^C	0.025	***
trans FA ⁿ	2.71 ^{GH}	3.13 ^{DE}	4.17 ^A	3.83 ^B	3.68 ^{BC}	3.69 ^B	3.44 ^C	3.23 ^D	2.68^{H}	2.84^{FGH}	2.93^{EFG}	2.97^{EF}	0.105	***
trans FA (exc. VA)	1.70^{D}	1.92 ^C	2.09 ^B	2.24 ^A	2.08 ^B	1.97 ^C	1.58^{E}	1.46 ^F	1.59 ^E	1.85 ^C	1.91 ^C	1.92 ^C	0.046	***
Indices														
Human health-related														
AI ^o	2.90^{B}	2.63 ^C	2.24^{E}	2.26^{E}	2.32^{DE}	2.33 ^E	2.24^{E}	2.43 ^D	3.04 ^A	3.02 ^A	2.98^{AB}	2.89^{AB}	0.048	***
TI^{p}	3.38 ^A	3.09 ^B	2.58^{E}	2.67 ^D	2.69 ^D	2.73 ^D	2.72 ^D	2.90 ^C	3.42 ^A	3.48 ^A	3.43 ^A	3.36 ^A	0.042	***
Δ^9 -desaturase activity														
$\Delta^9 \mathbf{I} r$	0.279 ^D 0	.293 ^c ().317 ^A ().318 ^A ().317 ^A ().314 ^A 0.	319 ^A 0	.308 ^B 0	$.277^{\rm D}$ 0.2	$269^{\rm E}$ 0	.271 ^E 0	.276 ^{DE}	0.0028	***
C14:1/C14:0	0.080^{F}	0.077^{G}	0.075^{G}	0.081^{EF}	0.084 ^C	0.084^{CD}	0.086^{B}	0.087^{B}	0.089^{A}	0.083 ^{CD}	0.082^{DE}	0.081^{EF}	0.0008	***
C16:1/C16:0	0.054^{E}	0.056^{D}	0.063 ^A	0.062^{B}	0.062^{B}	0.060°	0.060°	0.059 ^c	0.056^{D}	0.052^{EF}	0.052^{F}	0.052^{F}	0.0006	***
OA/C18:0	1.918 ^{de}	1.887^{EF}	1.940 ^C	1.926 ^{CD}	1.945 ^{CD}	1.957 ^C	2.008 ^B	2.051 ^A	2.023 ^{AB}	1.841 ^G	1.838 ^G	1.853 ^{FG}	0.0250	***
RA/VA	0.475^{BC}	0.476^{BC}	0.433 ^D	0.527^{A}	0.537 ^A	0.491 ^B	0.449 ^{CD}	0.451 ^{CD}	0.528^{A}	0.483 ^B	0.489 ^B	0.474^{BC}	0.0113	***

^{*a*} In September, there was a missing sample of free-range milk in the analysis of basic composition and the mean on this set of parameters was calculated from nine samples ^{*b*} Significances were declared at ***, P < 0.001; **, P < 0.01; *, P < 0.05; \uparrow , 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 Fisher's Least Significant Difference test (P < 0.05)

^c Somatic cell count

^d Saturated FA: C4:0, C5:0, C6:0, C7:0, C8:0, C9:0, C10:0, C11:0, C12:0, C13:0, C13:0*iso*, C13:0*anteiso*, C13:0, C14:0*iso*, C14:0, C15:0*anteiso*, C15:0, C16:0*iso*, C16:0, C17:0*iso*, C17:0, C18:0*iso*, C18:0, C20:0, C22:0, C24:0

^{*e*} Monounsaturated FA: *c*9 C10:1, *c*10 C11:1, *c*9 C12:1, *c*9 C13:1, *t*9 14:1, *c*9 C14:1, *c*10 C15:1, *t*7+t8 C16:1, *t*9 C16:1, *t*11+*t*12+*t*13 C16:1, *c*9 C16:1 (co-elutes with C17:0*anteiso*), *c*11 C16:1, *c*13 C16:1, *t*10 C17:1, *c*9 C17:1, *t*4 C18:1, *t*5 C18:1, *t*6+*t*7+*t*8 C18:1, *t*9 C18:1, *t*10 C18:1, *t*11 C18:1 (VA), *c*6+*t*12 C18:1, *c*9 C18:1 (OA), *t*15 C18:1, *c*11 C18:1, *c*12 C18:1, *c*13 C18:1, *t*16+*c*14 C18:1, *c*15 C18:1 (co-elutes with C19:0), *c*16 C18:1, *c*5 C20:1, *c*8 C20:1, *c*11 C20:1, *c*13 C22:1, *c*15 C24:1

^{*f*} Polyunsaturated FA: *t*11*t*15 C18:2, *t*9*t*12 C18:2, *c*9*t*13 C18:2, *c*9*t*14 C18:2, *c*9*t*14 C18:2, *c*9*t*12 C18:2, *t*9*c*12 C18:2, *t*9*c*12 C18:2, *c*9*c*12 C18:2 (LA), *t*12*c*15 C18:2 (coelutes with *c*9 C19:1), *c*6*c*9*c*12 C18:3, *c*9*c*12*c*15 C18:3 (ALNA), *c*9*c*11 C18:2 conjugated (RA) (co-elutes with *t*7*c*9+*t*8*c*10+*t*6*c*8 C18:2), other C18:2 conjugated FA of unknown isomerism, *c*11*c*14 C20:2, *c*8*c*11*c*14 C20:3, *c*11*c*14*c*17 C20:3, *c*5*c*8*c*11*c*14 C20:4, *c*13*c*16 C22:2, EPA, *c*13*c*16*c*19 C22:3, DPA, DHA

^{*g*} *cis* MUFA: *c*9 C10:1, *c*10 C11:1, *c*9 C12:1, *c*9 C13:1, *c*9 C14:1, *c*10 C15:1, *c*9 C16:1 (co-elutes with C17:0*anteiso*), *c*11 C16:1, *c*13 C16:1, *c*9 C17:1, *c*6 C18:1 (co-elutes with *t*12 C18:1), OA, *c*11 C18:1, *c*12 C18:1, *c*13 C18:1, *c*14 C18:1 (co-elutes with *t*16 C18:1), *c*15 C18:1 (co-elutes with C19:0), *c*16 C18:1, *c*5 C20:1, *c*8 C20:1, *c*11 C20:1, *c*13 C22:1, *c*15 C24:1

^h trans MUFA: t9 14:1, t7+t8 C16:1, t9 C16:1, t11+t12+t13 C16:1, t10 C17:1, t4 C18:1, t5 C18:1, t6+t7+t8 C18:1, t9 C18:1, t10 C18:1, VA, t12 C18:1 co-elutes with c6 C18:1), t15 C18:1, t16 C18:1 (co-elutes with c14 C18:1)

^{*i*} *cis* PUFA: LA, ALNA, c11c14 C20:2, c8c11c14 C20:3, c11c14c17 C20:3, c5c8c11c14 C20:4, c13c16 C22:2, EPA, c13c16c19 C22:3, DPA, DHA ^{*j*} *trans* PUFA: *t*11*t*15 C18:2, *t*9*t*12 C18:2

^k cis/trans + trans/cis PUFA: c9t13 C18:2, c10t14 C18:2, c9t14 C18:2, c9t12 C18:2, t9c12 C18:2, t11c15 C18:2, t12c15 C18:2 (co-elutes with c9 C19:1), RA (co-elutes with t7c9+t8c10+t6c8 C18:2), other C18:2 conjugated FA of unknown isomerism

¹ omega-3 PUFA (n-3): *t*11*t*15 C18:2, *t*11*c*15 C18:2, *t*12*c*15 C18:2 (co-elutes with *c*9 C19:1), ALNA, *c*11*c*14*c*17 C20:3, EPA, *c*13*c*16*c*19 C22:3, DPA, DHA ^{*m*} omega-6 PUFA (n-6): *t*9*t*12 C18:2, *c*9*t*12 C18:2, *t*9*c*12 C18:2, LA, *c*6*c*9*c*12 C18:3, *c*11*c*14 C20:2, *c*8*c*11*c*14 C20:4, *c*13*c*16 C22:2, *c*7*c*10*c*13*c*16 C22:4 ^{*n*} trans FA: t9 C14:1, t9 C16:1, t11+t12+t13 C16:1, t10 C17:1, t4 C18:1, t5 C18:1, t6+t7+t8 C18:1, t9 C18:1, t10 C18:1, VA, t12 C18:1, t15 C18:1, t16 C18:1, t11t15 C18:2, t9t12 C18:2, t12t15 C18:2

^o Atherogenicity index = $(C12:0 + 4 \times C14:0 + C16:0) / (MUFA + PUFA)$, as described in Srednicka-Tober et al. (2016)

^{*p*} Thrombogenicity index = $(C14:0 + C16:0 + C18:0) / [(0.5 \times MUFA) + (0.5 \times n-6) + (3 \times n-3) + (n-3/n-6)]$, as described in Srednicka-Tober et al. (2016)

 $^{r}\Delta^{9}$ -desaturase activity index = (c9 C14:1+c9 C16:1+OA+RA)/(c9 C14:1+c9 C16:1+OA+RA+C14:0+C16:0+C18:0+VA), as proposed by Kay et al. (2004)

Table 3

Means (and average SE) and ANOVA P-values for the effect of production system (conventional, CON; organic, ORG; free-range, FR) on the estimated intakes of fatty acid (FA) intakes from dairy products, using milk FA profiles measured in this study.

Intakes	Age group			Male	ales			Females					All				
Fat (g/d) ^a	4-10			58.4					56.2					57.4			
	11-18			73.8					59.8					67.0			
	19-64			77.7					60.1				68.8				
	65+			74.1				57.8					65.0				
Milk fat ^a	4-10			22.2					23.0					22.6			
(% fat)	11-18			15.7	5.7			15.1						15.4			
	19-64			15.8					17.2					16.5			
	65+			22.4			24.4							23.5			
				Male	s		Females							All			
				ANOVA		ANOVA					ANOVA						
Age group	p FA intakes (g/d)	CON	ORG	FR	SE	P-value ^b	CON	ORG	FR	SE	P-value ^b	CON	ORG	FR	SE	P-value ^b	
4-10	$SFA^{c}(g/d)$	8.3	8.3	8.4	0.05	ns	8.3	8.2	8.3	0.05	ns	8.3	8.3	8.4	0.05	ns	
	$MUFA^{d}$ (g/d)	3.3	3.3	3.3	0.04	ns	3.3	3.3	3.3	0.04	ns	3.3	3.3	3.3	0.04	ns	
	PUFA $e (mg/d)$	480^{B}	540 ^A	464 ^B	8.5	***	479 ^в	539 ^A	462 ^B	8.5	***	480 ^B	³ 540 ^A	464 ^B	8.5	***	
	$n-3^{f}$ (mg/d)	96 ^B	141 ^A	105 ^B	3.3	***	96 ^B	141 ^A	105 ^B	3.3	***	96 ^в	³ 142 ^A	105 ^B	3.3	***	
	ALNA (mg/d)	53 ^B	81 ^A	58 ^B	1.7	***	53 ^B	81 ^A	57 ^в	1.7	***	53 ^B	8 81 ^A	58 ^B	1.7	***	
	EPA+DHA (mg/d)	7 ^B	9 ^A	7 ^B	0.2	***	7 ^в	9 ^A	7 ^в	0.2	***	7 ^в	9 ^A	7 ^в	0.2	***	
	trans FA ^g (mg/d)	378 ^B	429 ^A	367 ^в	10.4	***	377 ^в	428 ^A	366 ^B	10.4	***	378 ^B	³ 430 ^A	367 ^в	10.4	***	
	trans FA ^g (exc. VA) (mg/d)	230	223	218	5.2	ns	230	222	218	5.2	ns	230	223	218	5.2	ns	
11-18	$SFA^{c}(g/d)$	7.4	7.4	7.5	0.05	ns	5.8	5.8	5.8	0.04	ns	6.6	6.6	6.7	0.04	ns	
	$MUFA^{d}$ (g/d)	2.9	2.9	2.9	0.04	ns	2.3	2.3	2.3	0.03	ns	2.6	5 2.6	2.6	0.03	ns	
	PUFA e (mg/d)	429 ^в	483 ^A	414 ^B	7.6	***	334 ^в	376 ^A	323 ^B	5.9	***	382 ^B	³ 430 ^A	369 ^B	6.8	***	
	$n-3^{f}$ (mg/d)	86 ^B	126 ^A	94 ^b	3.0	***	67 ^в	99 ^A	73 ^в	2.3	***	76 ^в	³ 113 ^A	83 ^B	2.6	***	
	ALNA (mg/d)	47 ^в	73 ^A	51 ^B	1.5	***	37 ^в	57 ^A	40^{B}	1.2	***	42 ^B	65 ^A	46 ^B	1.4	***	
	EPA+DHA (mg/d)	6 ^B	8 ^A	7 ^в	0.2	***	5 ^в	6 ^A	5 ^в	0.1	***	5 ^в	5 7 ^A	6 ^B	0.2	***	
	trans FA g (mg/d)	338 ^B	384 ^A	328 ^B	9.3	***	263 ^B	299 ^A	255 ^B	7.2	***	301 ^B	³ 342 ^A	292 ^B	8.3	***	
	trans FA ^g (exc. VA) (mg/d)	206	199	195	4.7	ns	160	155	152	3.6	ns	183	177	174	4.2	ns	
19-64	$SFA^{c}(g/d)$	7.9	7.8	7.9	0.05	ns	6.6	6.6	6.7	0.04	ns	7.3	7.2	7.3	0.05	ns	
	$MUFA^{d}$ (g/d)	3.1	3.1	3.1	0.04	ns	2.6	2.6	2.6	0.03	ns	2.9	2.9	2.9	0.04	ns	
	PUFA $e (mg/d)$	455 ^B	511 ^A	439 ^B	8.0	***	383 ^B	431 ^A	370 ^B	6.8	***	420 ^B	473 ^A	406^{B}	7.4	***	
	$n-3^f$ (mg/d)	91 ^B	134 ^A	99 ^b	3.1	***	76 ^в	113 ^A	84 ^B	2.6	***	84 ^B	³ 124 ^A	92 ^B	2.9	***	
	ALNA	50 ^B	77 ^A	55 ^B	1.6	***	42 ^B	65 ^A	46 ^B	1.4	***	46 ^B	⁸ 71 ^A	50 ^B	1.5	***	

	EPA+DHA	6 ^B	9 ^A	7 ^B	0.2	***	5 ^B	7 ^A	6 ^B	0.2	***	6 ^B	8 ^A	6 ^B	0.2	***
	trans FA ^g (mg/d)	358 ^B	407 ^A	347 ^B	9.8	***	301 ^b	342 ^A	292 ^B	8.3	***	331 ^b	376 ^A	321 ^b	9.1	***
	trans ^g FA (exc. VA) (mg/d)	218	211	207	5.0	ns	184	178	174	4.2	ns	202	195	191	4.6	ns
65+	$SFA^{c}(g/d)$	10.7	10.6	10.7	0.07	ns	9.0	9.0	9.1	0.06	ns	9.8	9.7	9.9	0.06	ns
	$MUFA^{d}$ (g/d)	4.2	4.2	4.2	0.06	ns	3.6	3.6	3.5	0.05	ns	3.9	3.9	3.8	0.05	ns
	$PUFA^{e}$ (mg/d)	615 ^B	692 ^A	593 ^B	10.9	***	522 ^B	588 ^A	504 ^B	9.2	***	566 ^B	636 ^A	546 ^B	10.0	***
	$n-3^{f}$ (mg/d)	123 ^B	181 ^A	134 ^b	4.2	***	104 ^B	154 ^A	114 ^B	3.6	***	113 ^b	167 ^A	124 ^B	3.9	***
	ALNA	68^{B}	104 ^A	74 ^B	2.2	***	58 ^B	88 ^A	63 ^B	1.9	***	63 ^B	96 ^A	68^{B}	2.0	***
	EPA+DHA	9 ^B	12 ^A	9 ^B	0.3	***	7^{B}	10 ^A	8^{B}	0.2	***	8^{B}	11 ^A	9 ^B	0.2	***
	trans FA ^g (mg/d)	484 ^B	550 ^A	469 ^B	13.3	***	411 ^B	467 ^A	399 ^b	11.3	***	445 ^B	506 ^A	432 ^B	12.3	***
	trans ^g FA (exc. VA) (mg/d)	295	285	280	6.7	ns	251	242	238	5.7	ns	271	262	257	6.2	ns

^{*a*} Intake data of fats and fatty acids of Year 1 of the National Diet and Nutrition Survey rolling programme 2008-2009, as presented by Bates et al. (2014). For the purposes of the intake calculations, this study assumes that all dairy products produced in the UK have the same FA profile as the whole milk analysed.

^b Significances were declared at ***, P < 0.001; **, P < 0.01; *, P < 0.05; t, 0.05 < P < 0.10 (trend); ns, P > 0.10 (non-significant). Means for production system within a row and gender with different upper case letters are significantly different according to Fisher's Least Significant Difference test (P < 0.05)

^d Saturated FA: C4:0, C5:0, C6:0, C7:0, C8:0, C9:0, C10:0, C11:0, C12:0, C13:0, C13:0*iso*, C13:0*anteiso*, C13:0, C14:0*iso*, C14:0, C15:0*anteiso*, C15:0, C16:0*iso*, C16:0, C17:0*iso*, C17:0, C18:0*iso*, C18:0, C20:0, C22:0, C24:0

^{*e*} Monounsaturated FA: *c*9 C10:1, *c*10 C11:1, *c*9 C12:1, *c*9 C13:1, *t*9 14:1, *c*9 C14:1, *c*10 C15:1, *t*7+t8 C16:1, *t*9 C16:1, *t*11+t12+t13 C16:1, *c*9 C16:1 (co-elutes with C17:0*anteiso*), *c*11 C16:1, *c*13 C16:1, *t*10 C17:1, *c*9 C17:1, *t*4 C18:1, *t*5 C18:1, *t*6+*t*7+*t*8 C18:1, *t*9 C18:1, *t*10 C18:1, *t*11 C18:1 (VA), *c*6+*t*12 C18:1, *c*9 C18:1 (OA), *t*15 C18:1, *c*11 C18:1, *c*12 C18:1, *c*13 C18:1, *t*16 + *c*14 C18:1, *c*15 C18:1 (co-elutes with C19:0), *c*16 C18:1, *c*5 C20:1, *c*8 C20:1, *c*11 C20:1, *c*13 C22:1, *c*15 C24:1

^{*f*} Polyunsaturated FA: *t*11*t*15 C18:2, *t*9*t*12 C18:2, *c*9*t*13 C18:2, *c*10*t*14 C18:2, *c*9*t*14 C18:2, *c*9*t*12 C18:2, *t*9*c*12 C18:2, *t*11*c*15 C18:2, *c*9*c*12 C18:2 (LA), *t*12*c*15 C18:2 (co-elutes with *c*9 C19:1), *c*6*c*9*c*12 C18:3, *c*9*c*12*c*15 C18:3 (ALNA), *c*9*c*11 C18:2 conjugated (RA) (co-elutes with *t*7*c*9+*t*8*c*10+*t*6*c*8 C18:2), other C18:2 conjugated FA of unknown isomerism, *c*11*c*14 C20:2, *c*8*c*11*c*14 C20:3, *c*11*c*14*c*17 C20:3, *c*5*c*8*c*11*c*14 C20:4, *c*13*c*16 C22:2, EPA, *c*13*c*16*c*19 C22:3, DPA, DHA

^{*g*} omega-3 PUFA (n-6): *t*11*t*15 C18:2, *t*11*c*15 C18:2, *t*12*c*15 C18:2 (co-elutes with *c*9 C19:1), ALNA, *c*11*c*14*c*17 C20:3, EPA, *c*13*c*16*c*19 C22:3, DPA, DHA

^h omega-6 PUFA (n-6): *t9t*12 C18:2, *c9t*12 C18:2, *t9c*12 C18:2, LA, *c6c9c*12 C18:3, *c*11*c*14 C20:2, *c8c*11*c*14 C20:3, *c5c8c*11*c*14 C20:4, *c*13*c*16 C22:2, *c7c*10*c*13*c*16 C22:4

^{*i*} trans FA: t9 C14:1, t9 C16:1, t11+t12+t13 C16:1, t10 C17:1, t4 C18:1, t5 C18:1, t6+t7+t8 C18:1, t9 C18:1, t10 C18:1, VA, t12 C18:1, t15 C18:1, t16 C18:1, t11t15 C18:2, t9t12 C18:2, t12t15 C18:2



Figure 1. Interaction means \pm SE (error bars) for the effects of production system (conventional, CON; organic, ORG; free-range, FR) and month (in order of appearance from left to right in Axis Y: M, March; A, April; M, May; J, June; J, July; A, August; S, September; O, October; N, November; D, December; J, January; F, February) on the concentrations of (a) C12:0, (b) C16:0, (c) linoleic acid (LA), (d) rumenic acid (RA) and (e) α -linolenic acid (ALNA) of milk collected from retail outlets during the year. P represents the ANOVA P-value for the interaction. Means for production system and within a month with different upper case letters are significantly different according to Fisher's Least Significant Difference test (P < 0.05). 704







Figure 2. Interaction means \pm SE (error bars) for the effects of production system (conventional, CON; organic, ORG; free-range, FR) and month (in order of appearance from left to right in Axis Y: M, March; A, April; M, May; J, June; J, July; A, August; S, September; O, October; N, November; D, December; J, January; F, February) on the concentrations of (a) saturated fatty acids (SFA), (b) monounsaturated fatty acids (MUFA), (c) *trans* MUFA, (d) polyunsaturated fatty acids (PUFA), (e) *cis* PUFA, (f) omega-6 PUFA (n-6), (g) *trans* FA, *trans* FA (h) excluding VA, (i) the ratio of omega-3 PUFA/omega-6 PUFA (n-3/n-6) and (j) the atherogenicity index (as proposed by Srednicka-Tober et al. (2016)) of milk collected from retail outlets during the year. P represents the ANOVA P-value for the interaction. Means for production system and within a month with different upper case letters are significantly different according to Fisher's Least Significant Difference test (P < 0.05). 706



Figure A1. Interaction means \pm SE (error bars) for the effects of production system (conventional, organic, free-range) and month (in order of appearance from left to right in Axis Y: M, March; A, April; M, May; J, June; J, July; A, August; S, September; O, October; N, November; D, December; J, January; F, February) on the concentrations of lactose of milk collected from retail outlets during the year. P represents the ANOVA P-value for the interaction. Means for production system and within a month with different upper case letters are significantly different according to Fisher's Least Significant Difference test (P < 0.05).



Figure A2. Interaction means \pm SE (error bars) for the effects of production system (conventional, organic, free-range) and month (in order of appearance from left to right in Axis Y: M, March; A, April; M, May; J, June; J, July; A, August; S, September; O, October; N, November; D, December; J, January; F, February) on the Δ^9 -desaturase activity indices (Δ^9 I as proposed by Kay et al. (2004), and ratios of C14:1/C14:0, C16:1/C16:0, oleic acid (OA)/C18:0 and vaccenic acid (VA)/rumenic acid (RA)) of milk collected from retail outlets during the year. P represents the ANOVA P-value for the interaction. Means for production system and within a month with different upper case letters are significantly different according to Fisher's Least Significant Difference test (P < 0.05). 710

APPENDIX

Table A1

Means (and average SE) and ANOVA P-values for the effect of production system (conventional, CON; organic, ORG; free-range, FR) on the fatty acid profile (g/kg total fatty acids) of milk collected from retail outlets during the year

Production System												
	CON	ORG	FR		ANOVA							
Parameters assessed	n=48	n=48	$n=24^{a}$	SE	P-values ^b							
C4:0	20.7	21.2	20.9	0.54	†							
C5:0	0.207	0.230	0.194	0.0189	ns							
C6:0	15.3 ^в	15.9 ^A	15.5 ^{AB}	0.38	*							
C7:0	0.218 ^A	0.174^{B}	0.206^{AB}	0.0123	*							
C8:0	10.0	10.3	10.1	0.23	ns							
C9:0	0.288	0.244	0.270	0.0122	ns							
C10:0	25.1	25.7	25.5	0.55	ns							
c9 C10:1	2.44	2.55	2.52	0.045	ns							
C11:0	0.547	0.459	0.509	0.0228	ns							
C12:0	33.4	32.4	34.8	0.60	ns							
C13:0 iso	0.271 ^B	0.334 ^A	0.291 ^B	0.0104	**							
C13:0 anteiso	0.145	0.110	0.116	0.0087	†							
c9 C12:1	0.830	0.799	0.861	0.0173	ns							
C13:0	0.908	0.870	0.848	0.0265	ns							
C14:0 iso	0.812 ^B	1.037 ^A	0.836 ^B	0.0180	***							
C14:0	111	114	114	1.1	ns							
t9 C14:1	2.16 ^C	2.56 ^A	2.37 ^B	0.042	***							
C15:0 anteiso	4.23 ^B	4.74 ^A	4.65 ^A	0.098	**							
c9 C14:1	9.31	9.09	9.56	0.138	ns							
C15:0	10.3 ^B	11.1 ^A	10.6^{B}	0.13	**							
C16:0 iso	1.99 ^B	2.26 ^A	1.97 ^B	0.028	***							
C16:0	331 ^A	314 ^B	325 ^A	37	***							
t6+t7+t8 C16·1	0 316 ^A	0.293^{B}	0.290^{B}	0.0071	*							
t9 C16:1	0.092	0.081	0.083	0.0030	ns							
C17.0 iso	3.84^{B}	4 28 ^A	4.09^{AB}	0 1 1 0	*							
t11+t12+t13 C16·1	1 96	1.20	1.02	0.047	ns							
c9 C16:1 + C17 anteiso	19 1 ^A	17.5 ^B	18 8 ^A	0.014	***							
c11 C16:1	1 43	1 48	1 89	0.053	+							
c13 C16:1	1.40	1 34	1.09	0.055	/ ns							
C17:0	1.40 ∕1.87 ^B	5 69 ^A	5 02 ^B	0.052	***							
t10 c17:1	-4.07	0.551 ^A	0.477 ^C	0.070	***							
C18:0 iso	0.501	0.551	0.477	0.0195	ne							
c9 C17·1	2.11^{B}	0.030 2.27A	2 15 ^{AB}	0.0175	*							
C18·0	100	106	103	0.055	ns							
t4 C18·1	0 158 ^A	0.128 ^B	0.120^{B}	0.00/0	**							
t5 C18·1	0.150 0.111A	0.120 0.088 ^B	0.002AB	0.0042	*							
15×10.1 t6+t7+t8 C18·1	2.76	2 55	2.40	0.0037	*							
t9 C18·1	1 93 ^A	1.66 ^B	1 72 ^{AB}	0.048	/ *							
t10 C18:1	3 70	3 /0	3.40	0.0+3 0.241	ne							
t11 C18:1	12.70	17 1 ^A	12 3 ^B	0.241	***							
$c6 \pm t12 C18.1$	2 8 3 A	2.30^{B}	12.3 2 50 ^B	0.71	**							
c0 - 112 - 0.1	2.85	2.30	2.50	2.0	ne							
+15 C18.1	200 2.08A	1 80 ^B	1 07AB	0.146	*							
c11 C19:1	2.08 5.80A	1.67 4.61 ^B	1.97 4.00 ^B	0.140	*							
c11 C10.1	2.09	4.01	4.90	0.099								
c12 C18.1	2.24 0.012A	1.94 0.910B	1.90	0.037	*							
$c_{13} c_{10,1}$	2.915	2 20	0.004	0.0100								
$110 \pm 014 \cup 10.1$	5.23 1.27	3.29 1.25	3.27 1.24	0.049	118 -4							
C13 C10.1 + C19.0	1.27 0.260B	1.55	1.54 0.214B	0.039	<i>T</i>							
11113 U18:2	0.2095	0.444.1	0.5145	0.0278	-1-							
19112 018:2	0.071	0.072	0.058	0.0072	ns							

c9t13 C18:2	2.10	2.00	1.99	0.052	ns
c10t14 C18:2	1.10	1.08	1.00	0.024	ns
c9t14 C18:2	1.25 ^A	1.14^{B}	1.18 ^{AB}	0.022	*
c9t12 C18:2	0.620 ^A	0.596 ^B	0.614^{AB}	0.0087	*
c16 C18:1	0.336	0.371	0.320	0.0166	ns
t11c15 C18:2	1.50 ^B	2.23 ^A	1.71 ^B	0.101	**
t9c12 C18:2	0.194 ^A	0.118 ^C	0.157 ^B	0.0125	***
c9c12 C18:2	17.1	16.6	15.3	0.28	ns
t12c15 C18:2 + c9 C19:1	0.430	0.480	0.480	0.0461	ns
C20:0	1.40^{B}	1.60 ^A	1.39 ^B	0.025	**
c6c9c12 C18:3	0.258	0.229	0.226	0.0069	ns
c8 C20:1	1.02 ^B	1.13 ^A	1.01 ^B	0.014	**
c11 C20:1	0.393	0.346	0.347	0.0295	†
c9c12c15 C18:3	4.39 ^B	6.71 ^A	4.76 ^B	0.141	***
c9t11 C18:2	5.91 ^B	7.95 ^A	6.06^{B}	0.315	***
Unknown C18:2 conjugated	0.286^{B}	0.344 ^A	0.277 ^B	0.0172	*
Unknown C18:2 conjugated	0.265^{B}	0.413 ^A	0.288 ^B	0.0134	***
c11c14 C20:2	0.196 ^B	0.220^{A}	0.196 ^B	0.0156	*
C22:0	0.556^{B}	0.735 ^A	0.578^{B}	0.0118	***
c8c11c14 C20:3	0.809	0.704	0.713	0.0105	†
c13 C22:1	0.160	0.182	0.093	0.0129	ns
c11c14c17 C20:3	0.105^{B}	0.186 ^A	0.102 ^B	0.0102	***
c5c8c11c14 C20:4	1.06	0.95	0.94	0.016	†
c13c16 C22:2	0.397	0.452	0.495	0.0138	ns
c5c8c11c14c17 C20:5	0.484^{B}	0.674^{A}	0.551 ^B	0.0143	***
C24:0	0.345 ^B	0.457 ^A	0.362 ^B	0.0063	***
c13c16c19 C22:3	0.104	0.114	0.096	0.0070	†
c7c10c13c16 C22:4	0.170^{A}	0.132 ^B	0.114 ^B	0.0113	*
c7c10c13c16c19 C22:5	0.795^{B}	1.024 ^A	0.834 ^B	0.0161	***
c4c7c10c13c16c19 C22:6	0.067	0.078	0.060	0.0051	ns

^{*a*} In September, there was a missing sample of free-range milk in the analysis of basic composition and the mean on this set of parameters was calculated from 23 samples ^{*b*} 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 Fisher's Least Significant Difference test (P < 0.05)

						• 1	Month		,					
	March	April	May	June	July	August	September	October	November	December	January	February		ANOVA
Parameters assessed	n=10	n=10	n=10	n=10	n=10	n=10	$n=10^a$	n=10	n=10	n=10	n=10	n=10	SE	P-values ^b
C4:0	20.8 ^D	22.4 ^{BC}	22.7 ^B	23.2 ^A	23.5 ^A	22.4 ^{BC}	14.7 ^F	14.3 ^I	⁷ 18.3 ^E	23.3 ^A	23.6 ^A	22.0 ^C	0.19	***
C5:0	0.224	0.215	0.215	0.174	0.169	0.172	0.125	0.140	0.202	0.235	0.296	0.396	0.0258	ns
C6:0	16.5 ^{BC}	16.7 ^B	16.5 ^C	16.6 ^{BC}	16.5 ^C	16.1 ^D	10.8 ^F	10.9 ^I	⁷ 15.1 ^E	17.3 ^A	17.4 ^A	16.7 ^B	0.14	***
C7:0	0.231 ^{BC}	0.223 ^{BC}	0.219 ^C	0.163 ^D	0.151^{DE}	0.155 ^D	0.115 ^E	0.141^{DE}	e 0.208 ^c	0.245^{BC}	0.274 ^A	0.253 ^B	0.0136	***
C8:0	10.9 ^{AB}	11.0 ^{AB}	11.0 ^{AB}	10.6 ^{CD}	10.4^{DE}	10.2^{E}	7.1 ^G	7.4 ^I	⁷ 10.4 ^{DE}	11.1 ^A	11.0 ^A	10.7 ^{BC}	0.13	***
C9:0	0.317 ^{AB}	0.303 ^B	0.311 ^B	0.217 ^C	0.210 ^C	0.215 ^C	0.173 ^D	0.2080	0.308 ^{AB}	0.318 ^{AB}	0.326 ^A	0.297 ^B	0.0138	***
C10:0	27.6 ^A	27.5^{AB}	27.5^{AB}	25.3 ^C	24.9 ^c	24.5 ^c	18.9 ^E	20.3 ^r	27.7 ^A	27.3 ^{AB}	27.2 ^{AB}	26.4 ^B	0.44	***
c9 C10:1	2.62^{BCD}	2.50^{EF}	2.43 ^G	2.46^{FG}	2.56^{DEF}	2.54^{DEF}	1.99 ¹	2.10 ^H	^I 2.85 ^A	2.69 ^B	2.65 ^{BC}	2.58^{CDE}	0.041	***
C11:0	0.587^{A}	0.552 ^A	0.584 ^A	0.394 ^{bc}	0.381 ^C	0.391 ^{bC}	0.348 ^C	0.429 ^E	³ 0.607 ^A	0.610 ^A	0.594 ^A	0.573 ^A	0.0267	***
C12:0	35.9 ^B	34.8 ^{BCD}	34.2 ^D	31.1^{EF}	31.4 ^E	31.1^{EF}	27.5 ^G	29.9 ¹	⁷ 37.7 ^A	35.7 ^{BC}	35.6 ^{BC}	34.3 ^{CD}	0.62	***
C13:0 iso	0.280^{E}	0.280^{E}	0.306 ^C	0.377 ^A	0.380 ^A	0.380 ^A	0.332 ^B	0.300 ^{CE}	0.288 ^{DE}	0.221 ^F	0.229 ^F	0.227 ^F	0.0125	***
C13:0 anteiso	0.113 ^B	0.105 ^B	0.098 ^B	0.111 ^B	0.118 ^B	0.291 ^A	0.096 ^B	0.098^{E}	^B 0.116 ^B	0.115 ^B	0.120 ^B	0.123 ^B	0.0105	***
c9 C12:1	0.868^{B}	0.812 ^{CD}	0.812 ^{CDE}	0.759^{E}	0.796 ^{CDE}	0.787^{CDE}	0.697^{F}	0.773^{DE}	E 0.995 ^A	0.881 ^B	0.876^{B}	0.827^{BC}	0.0210	***
C13:0	0.976^{ABC}	0.927 ^{CD}	0.935^{DE}	0.752^{F}	0.745^{F}	0.768^{F}	0.713 ^F	0.824^{H}	E 1.026 ^A	0.970 ^{ABCD}	1.001^{AB}	0.933 ^{BCD}	0.0316	***
C14:0 iso	0.837^{DE}	0.818^{E}	0.846^{CDE}	1.008^{A}	1.048^{A}	1.004^{A}	0.907^{B}	0.882^{BCE}	0.913 ^{BC}	0.864^{CDE}	0.863 ^{CDE}	0.891^{BCDE}	0.0416	***
C14:0	116 ^B	113 ^C	109 ^{DE}	107 ^E	109 ^D	109 ^{DE}	105 ^F	110 ^r	^o 123 ^A	117 ^B	117 ^B	116 ^B	1.1	***
t9 C14:1	2.23^{DEF}	2.23^{DE}	2.40^{BC}	2.65 ^A	2.74 ^A	2.63 ^A	2.41 ^B	2.31 ^{CE}	2.27 ^{DE}	2.10 ^F	2.17^{EF}	2.21^{DEF}	0.072	***
C15:0 anteiso	4.26 ^{BCD}	4.36 ^{BCD}	5.07 ^A	5.14 ^A	5.23 ^A	4.50 ^B	4.52 ^{BC}	4.48^{BC}	4.34 ^{BCD}	4.00^{D}	4.15 ^{CD}	4.18^{BCD}	0.120	***
c9 C14:1	9.29^{DE}	8.64 ^F	8.17 ^G	8.66 ^F	9.18 ^{DE}	9.08 ^E	8.99 ^E	9.61 ^E	³ 10.99 ^A	9.73 ^B	9.57 ^{BC}	9.32 ^{CD}	0.136	***
C15:0	10.8 ^{CD}	10.1 ^E	10.0^{EF}	9.8 ^F	10.1^{EF}	10.1 ^E	10.6^{E}	11.0 ^{BC}	^c 11.9 ^A	11.4 ^B	11.4 ^B	11.1 ^B	0.17	***
C16:0 iso	2.00^{EFG}	2.02^{EFG}	2.04^{DE}	2.26^{AB}	2.32 ^A	2.23 ^B	2.11^{E}	2.10 ^{CE}	2.02 ^{EFG}	1.98 ^{FG}	1.96 ^G	2.06^{EF}	0.056	***
C16:0	341 ^A	321 ^C	289 ^E	297 ^D	299 ^D	305 ^D	319 ^c	329 ^E	³ 346 ^A	348 ^A	344 ^A	342 ^A	3.6	***
t6+t7+t8 C16:1	0.309^{CDE}	0.303^{DE}	0.318 ^{BCD}	0.322 ^{BC}	0.333 ^{AB}	0.339 ^A	0.337 ^A	0.341 ^A	0.300 ^E	0.233 ^F	0.239 ^F	0.245 ^F	0.0068	***
t9 C16:1	0.077 ^C	0.089 ^{BC}	0.105 ^A	0.094^{AB}	0.086^{BC}	0.098^{AB}	0.081 ^C	0.077^{CE}	0.096 ^{AB}	0.063 ^D	0.077 ^C	0.089^{BC}	0.0050	***
C17:0 iso	3.50 ^F	3.76 ^E	4.78^{AB}	4.82^{AB}	4.91 ^A	4.65 ^{BC}	4.49 ^C	4.18 ^t	9 3.52 ^F	3.34 ^F	3.37 ^F	3.46 ^F	0.095	***
t11+t12+t13 C16:1	1.72 ^F	1.87 ^E	2.32 ^A	2.18 ^{BC}	2.19 ^{BC}	2.18 ^B	2.09 ^C	1.97 ^r) 1.72 ^F	1.56 ^G	1.58 ^G	1.61 ^G	0.037	***
c9 C16:1 + C17 anteiso	18.3 ^{CDE}	17.9^{EFG}	18.2 ^{DC}	18.4 ^{DC}	18.6 ^C	18.2^{CDEF}	19.1 ^B	19.3 ^A	19.3 ^{AB}	18.1^{DEF}	17.8^{FG}	17.7 ^G	0.31	***
c11 C16:1	1.66^{BC}	1.71 ^{AB}	1.66 ^{BC}	1.42^{EF}	1.54^{DE}	1.58 ^{CD}	1.82 ^A	1.70 ^{BC}	1.56 ^{CD}	1.25 ^F	1.32 ^F	1.28^{F}	0.097	***

Means (and average SE) and ANOVA P-values for the effect of month on the fatty acid profile (g/kg total fatty acids) of milk collected from retail outlets during the year

Table A2

c13 C16:1	1.45 ^{BC}	1.32 ^{BC}	1.30 ^{BC}	1.15 ^C	1.24 ^{BC}	1.17 ^C	1.22 ^{BC}	1.39 ^{BC}	1.87 ^A	1.50 ^B	1.47 ^B	1.40 ^{BC}	0.069	***
C17:0	5.21 ^{AB}	5.17 ^{AB}	5.38 ^{AB}	5.56 ^A	5.48^{AB}	5.36 ^{AB}	5.41 ^{AB}	5.34 ^{AB}	4.57 ^C	5.07 ^{BC}	5.07 ^{BC}	5.10 ^{AB}	0.160	***
t10 c17:1	0.529^{E}	0.546^{DE}	0.589 ^A	0.590 ^{AB}	0.582^{ABC}	0.561^{BCD}	0.566^{BCD}	0.561 ^{CD}	0.499 ^F	0.409^{G}	0.368^{H}	0.395 ^{GH}	0.0126	***
C18:0 iso	0.582^{EFG}	0.609 ^{CDE}	0.726 ^A	0.711 ^{AB}	0.753 ^A	0.658^{BC}	0.646^{BCD}	0.608^{DEF}	0.560^{FG}	0.549^{FG}	0.543 ^G	0.547^{G}	0.0325	***
c9 C17:1	2.15 ^D	2.15 ^D	2.38 ^A	2.37 ^A	2.38 ^A	2.27 ^{BC}	2.35 ^{AB}	2.25 ^C	2.11 ^D	1.94 ^E	1.92^{E}	1.92^{E}	0.041	***
C18:0	97 ^D	104 ^C	108 ^B	110 ^{AB}	109 ^B	108 ^B	111 ^A	104 ^C	90 ^E	97 ^D	97 ^D	99 ^D	1.8	***
t4 C18:1	0.129^{DEF}	0.130 ^{DEF}	0.141 ^{BCD}	0.125^{DEF}	0.110^{EF}	0.134^{CDE}	0.135^{CDE}	0.149^{BCD}	0.105 ^F	0.166 ^{AB}	0.157 ^{ABC}	0.179 ^A	0.0099	***
t5 C18:1	0.102^{CDE}	0.110^{BCD}	0.116 ^{AB}	0.112 ^{ABC}	0.095^{E}	0.100^{DE}	0.064^{G}	0.062^{G}	0.080^{F}	0.117^{AB}	0.095^{E}	0.122 ^A	0.0055	***
t6+t7+t8 C18:1	2.42 ^D	2.65 ^{BC}	2.83 ^A	2.76^{AB}	2.48 ^{CD}	2.65 ^B	2.86 ^A	2.92 ^A	2.46 ^{CD}	2.33 ^D	2.37 ^D	2.50^{CD}	0.083	***
t9 C18:1	1.60 ^E	1.82^{CDE}	2.01 ^A	1.87^{ABC}	1.80^{BCD}	1.83 ^{BCD}	1.44 ^F	1.37 ^F	1.68^{DE}	1.94^{ABC}	1.96 ^{AB}	2.04 ^A	0.074	***
t10 C18:1	2.94 ^E	4.07 ^{BC}	4.10 ^B	6.03 ^A	5.29 ^A	3.88 ^{BCD}	1.97 ^F	1.66 ^F	3.00 ^E	3.17^{DE}	3.31^{CDE}	3.25^{CDE}	0.288	***
t11 C18:1	10.1 ^E	12.2 ^D	20.9 ^A	15.9 ^C	16.1 ^C	17.2 ^{BC}	18.6 ^B	17.7 ^{BC}	10.9^{DE}	9.9 ^E	10.2^{E}	10.5^{DE}	1.00	***
c6 + t12 C18:1	2.91 ^B	3.02 ^B	2.96 ^B	3.05 ^B	2.42 ^C	2.46 ^C	1.49 ^E	1.23 ^E	1.83 ^D	2.91 ^B	3.32 ^A	3.00^{AB}	0.134	***
c9 C18:1	186 ^D	195 ^C	209 ^B	212 ^B	211 ^B	211 ^B	223 ^A	213 ^B	183 ^{DE}	178 ^E	179 ^E	184^{DE}	2.3	***
t15 C18:1	1.84 ^B	1.87 ^B	2.07 ^B	2.07 ^B	1.82 ^B	1.98 ^B	0.95 ^C	0.58^{D}	1.27 ^C	3.07 ^A	3.13 ^A	3.12 ^A	0.111	***
c11 C18:1	4.82^{FG}	5.36 ^{BCD}	5.75 ^A	5.43 ^B	5.08^{CDEF}	5.14^{CDEF}	5.38 ^{BC}	5.41 ^B	4.58 ^G	4.95^{EF}	5.03^{DEF}	5.21^{BCDE}	0.251	***
c12 C18:1	2.23 ^{AB}	2.27^{AB}	1.91 ^C	1.93 ^C	1.66 ^D	1.87 ^C	1.88 ^C	1.95 ^C	1.93 ^C	2.26 ^B	2.30 ^{AB}	2.43 ^A	0.100	***
c13 C18:1	0.809^{EF}	0.902^{BCD}	1.019 ^A	0.886^{BC}	0.824^{DE}	0.897^{BC}	0.939 ^B	0.901 ^B	0.750^{F}	0.833 ^{CDE}	0.785^{EF}	0.837^{CDE}	0.0250	***
t16 + c14 C18:1	3.16 ^{DE}	3.51 ^B	3.72 ^A	3.44 ^{BC}	3.33 ^{CD}	3.39 ^{BC}	3.52 ^B	3.28 ^D	2.85^{G}	2.93 ^{FG}	3.04^{EF}	3.06^{EF}	0.049	***
c15 C18:1 + C19:0	1.43 ^B	1.50^{AB}	1.59^{AB}	1.59 ^{AB}	1.58^{AB}	1.41 ^B	1.63 ^A	1.55^{AB}	1.18 ^C	0.75^{D}	0.78^{D}	0.77^{D}	0.042	***
t11t15 C18:2	0.215^{EF}	0.330 ^D	0.583 ^A	0.435 ^{BC}	0.479^{BC}	0.474^{BC}	0.493 ^{AB}	0.418 ^C	0.242^{DE}	0.155^{EF}	0.199 ^{EF}	0.150 ^F	0.0387	***
t9t12 C18:2	0.042^{E}	0.073^{BCDE}	0.110^{AB}	0.057^{DE}	0.064^{BCDE}	0.085^{ABCD}	0.114 ^A	0.098^{ABC}	0.048^{DE}	0.042^{DE}	0.060^{CDE}	0.035^{E}	0.0124	***
c9t13 C18:2	1.84 ^D	2.07 ^C	2.58 ^A	2.17^{BC}	2.16 ^{BC}	2.22 ^B	2.30 ^B	2.22 ^B	1.85 ^D	1.61 ^E	1.68^{DE}	1.74^{DE}	0.052	***
c10t14 C18:2	1.13 ^{AB}	1.18 ^A	1.14^{ABC}	1.07^{BCD}	1.03 ^{BCD}	1.04 ^D	1.04 ^D	1.12^{AB}	1.04^{BCD}	1.01 ^D	1.03 ^D	1.03 ^{CD}	0.040	***
c9t14 C18:2	1.13 ^E	1.22 ^D	1.31 ^{AB}	1.25^{BCD}	1.23 ^{CD}	1.27 ^{BC}	1.35 ^A	1.31 ^{AB}	1.14^{E}	0.98^{G}	1.04 ^F	1.04^{FG}	0.026	***
c9t12 C18:2	0.579^{DEF}	0.656^{AB}	0.679 ^A	0.629 ^{BC}	0.590^{DE}	0.610 ^{CD}	0.669 ^A	0.639 ^{BC}	0.555^{F}	0.551 ^F	0.569 ^{EF}	0.586^{DEF}	0.0121	***
c16 C18:1	0.256 ^D	0.352 ^C	0.500^{A}	0.427 ^B	0.404^{B}	0.393 ^B	0.424 ^B	0.413 ^B	0.312 ^C	0.209^{E}	0.239 ^{DE}	0.232^{DE}	0.0173	***
t11c15 C18:2	1.22^{E}	1.56 ^D	2.64 ^A	2.29 ^{BC}	2.44 ^B	2.31 ^B	2.25 ^{BC}	2.09 ^C	1.44^{DE}	1.26 ^E	1.28^{E}	1.22^{E}	0.143	***
t9c12 C18:2	0.224 ^A	0.250 ^A	0.140 ^{BC}	0.154 ^B	0.109 ^{CD}	0.113 ^{CD}	0.170 ^B	0.225^{A}	0.239 ^A	0.080^{D}	0.095 ^D	0.073 ^D	0.0180	***
c9c12 C18:2	17.4 ^{AB}	18.1 ^A	17.2 ^{ABC}	16.8 ^{BCD}	15.5^{EF}	16.4 ^{CDE}	16.9 ^{BC}	17.2^{ABC}	15.7^{EF}	15.6 ^F	15.5 ^F	16.1^{DEF}	0.56	***
t12c15 C18:2 + c9 C19:1	0.237 ^G	0.290^{DEF}	0.350 ^C	0.339 ^{CD}	0.330 ^{CDE}	0.275^{EFG}	0.319 ^{CDE}	0.312^{CDE}	0.253^{FG}	0.894^{B}	0.938 ^{AB}	0.986 ^A	0.0220	***
C20:0	1.54 ^B	1.44 ^{CD}	1.33 ^E	1.51 ^{BC}	1.42 ^D	1.30 ^E	1.48^{BCD}	1.48^{CD}	1.32 ^E	1.62 ^A	1.66 ^A	1.68 ^A	0.043	***
c6c9c12 C18:3	0.221 ^{BC}	0.239 ^{ABC}	0.306 ^A	0.244^{ABC}	0.230 ^{BC}	0.223 ^{BC}	0.227^{BC}	0.231 ^{BC}	0.196 ^C	0.274^{AB}	0.239 ^{ABC}	0.250 ^{ABC}	0.0120	*
c8 C20:1	1.03 ^{CD}	1.00 ^D	0.93 ^E	1.08^{BC}	1.09 ^B	1.07^{BC}	1.14 ^A	1.16 ^A	1.04^{BCD}	1.03 ^{BCD}	1.07^{BC}	1.10 ^B	0.026	***
c11 C20:1	0.428^{D}	0.471 ^{BC}	0.465 ^{BC}	0.478^{AB}	0.438 ^{CD}	0.446 ^{CD}	0.497^{AB}	0.516 ^A	0.442^{CD}	0.079^{E}	0.075^{E}	0.046^{E}	0.0147	***
c9c12c15 C18:3	4.69 ^D	5.20 ^C	6.54 ^A	6.09 ^B	6.07^{B}	5.99 ^B	6.11 ^B	5.51 ^C	4.60^{D}	4.52 ^D	4.63 ^D	4.72 ^D	0.380	***

c9t11 C18:2	4.76 ^E	5.65 ^C	8.83 ^A	8.31 ^{AB}	8.52 ^{AB}	8.44^{AB}	8.36 ^{AB}	7.97 ^B	5.62 ^{CD}	4.76^{E}	4.92^{DE}	4.93 ^{DE}	0.409	***
Unknown C18:2 conjugated	0.289 ^C	0.282 ^C	0.356 ^B	0.484^{A}	0.383 ^B	0.352 ^B	0.385 ^B	0.352 ^B	0.294 ^C	0.197 ^D	0.167^{DE}	0.146^{E}	0.0185	***
Unknown C18:2 conjugated	0.223 ^F	0.249^{EF}	0.372 ^{сва}	0.351 ^{BCD}	0.392 ^{AB}	0.384 ^A	0.385 ^{AB}	0.382^{ABC}	0.268^{EF}	0.275^{E}	0.324 ^D	0.344 ^{CD}	0.0283	***
c11c14 C20:2	0.103 ^F	0.116^{EF}	0.125^{E}	0.125^{EF}	0.126^{EF}	0.170^{D}	0.227 ^C	0.222 ^C	0.188 ^D	0.349 ^B	0.338 ^B	0.375 ^A	0.0091	***
C22:0	0.617^{DE}	0.622^{CD}	0.623 ^{CD}	0.678^{AB}	0.583^{EF}	0.536 ^G	0.709^{A}	0.712 ^A	0.569^{FG}	0.665 ^{BC}	0.618^{DE}	0.652^{BCD}	0.0320	***
c8c11c14 C20:3	0.764^{AB}	0.768^{AB}	0.753 ^{AB}	0.745^{ABC}	0.699 ^{CD}	0.729^{BC}	0.749^{AB}	0.772 ^A	0.685 ^D	0.764^{AB}	0.764^{AB}	0.782^{A}	0.0265	***
c13 C22:1	0.115	0.131	0.149	0.136	0.123	0.125	0.128	0.221	0.175	0.167	0.246	0.150	0.0266	†
c11c14c17 C20:3	0.078^{E}	0.089 ^{CDE}	0.111^{CD}	0.118 ^C	0.116 ^{CD}	0.109^{CDE}	0.117 ^C	0.101^{CDE}	0.083^{DE}	0.189 ^B	0.221 ^B	0.307 ^A	0.0155	***
c5c8c11c14 C20:4	0.98^{BC}	0.99 ^{BC}	0.97 ^{CD}	0.96 ^{CD}	0.91 ^D	0.95^{CD}	1.00^{BC}	1.03 ^{AB}	0.91 ^D	1.09 ^A	1.10 ^A	1.05^{AB}	0.033	***
c13c16 C22:2	0.364 ^{FG}	0.398^{EF}	0.521 ^A	0.470^{BCD}	0.493 ^{AB}	0.426^{DE}	0.458^{BC}	0.435^{CDE}	0.340^{G}	0.433 ^{CDE}	0.467^{BC}	0.459 ^{BC}	0.0233	***
c5c8c11c14c17 C20:5	0.502^{CD}	0.513 ^{CD}	0.636 ^A	0.641 ^{AB}	0.659 ^A	0.644^{AB}	0.647 ^A	0.611 ^B	0.535 ^C	0.515 ^{CD}	0.488 ^D	0.484 ^D	0.0327	***
C24:0	0.360 ^E	0.366 ^{DE}	0.377 ^{CDE}	0.428^{A}	0.411 ^{AB}	0.425^{A}	0.423 ^A	0.410^{AB}	0.333 ^F	0.409^{AB}	0.388 ^{CD}	0.394 ^{bc}	0.0192	***
c13c16c19 C22:3	0.071^{E}	0.078^{DE}	0.080^{DE}	0.091 ^{CDE}	0.079^{DE}	0.109 ^C	0.095^{CDE}	0.096 ^{CD}	0.075^{DE}	0.184 ^A	0.154 ^B	0.163 ^{AB}	0.0071	***
c7c10c13c16 C22:4	0.129 ^{CDE}	0.148^{CD}	0.124^{DE}	0.118^{DEF}	0.102^{EFG}	0.181 ^{BC}	0.081^{FG}	0.071^{G}	0.090 ^{FG}	0.231 ^A	0.242 ^A	0.205^{AB}	0.0155	***
c7c10c13c16c19 C22:5	0.786^{D}	0.787^{D}	0.878^{BC}	0.895 ^{BC}	0.934 ^{AB}	0.919 ^{ABC}	0.980^{A}	0.960^{AB}	0.937 ^{AB}	0.856 ^{CD}	0.918 ^{BC}	0.881 ^{BC}	0.0441	***
c4c7c10c13c16c19 C22:6	0.059	0.058	0.067	0.066	0.066	0.059	0.132	0.079	0.061	0.070	0.063	0.058	0.0087	ns

^{*a*} In September, there was a missing sample of free-range milk in the analysis of basic composition and the mean on this set of parameters was calculated from nine samples ^{*b*} 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 Fisher's Least Significant Difference test (P < 0.05)