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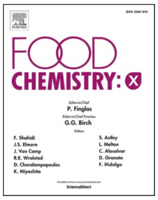

# Results of the BfR MEAL Study: The food type has a stronger impact on calcium, potassium and phosphorus levels than factors such as seasonality, regionality and type of production

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ABSTRACT

The BfR MEAL Study aims to provide representative levels of chemical substances in foods consumed by the population in Germany for dietary exposure assessment. Calcium, potassium and phosphorus (Ca, K, P) are essential to obtain physiological functions in humans. Levels were investigated in 356 foods. Foods were purchased representatively, prepared as typically consumed and pooled before analysis. High mean levels were found in milk, dairy products, legumes, nuts, oilseeds and spices as well as chia seeds (Ca, K, P), chewing gum (Ca) and cocoa powder (K). Different levels comparing organically and conventionally produced foods were determined among others in cereal cracker (puffed), olives and tofu. Higher K levels were found in fried compared to boiled potatoes. Similar P levels were mainly found in regionally and seasonally sampled foods. These data provide a substantially improved basis to address dietary exposure assessment of the population in Germany for Ca, K and P.

## Introduction

The minerals calcium (Ca), potassium (K), and phosphorus (P) are essential nutritional components for the human organism and must therefore be consumed with the diet.

The Ca content of foods varies widely. Main sources are milk and dairy products, and to some extent vegetables (e.g. kale, broccoli) and mineral waters. The absorption rate from food is approximately 20–60%, depending on various factors such as age, pregnancy, amount of food, Ca concentration in the intestinal lumen and vitamin D status (EFSA, European Food Safety Authority, 2015). The bioavailability of Ca can be reduced (absorption 5–8%) by some food ingredients such as phytic acid or oxalic acid (e.g. in cereals, rhubarb, or spinach) (Weaver, 2020).

Ca is mostly present in the human body as an extracellular component of bones and teeth (more than 99%). Together with other minerals such as phosphate, Ca stabilizes and strengthens the bones. In addition, bone tissue serves as a functional reservoir for the homeostatic

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*Abbreviations:* BfR, German Federal Institute for Risk Assessment; BLE, Federal Office for Agriculture and Food; BMEL, Federal Ministry of Food and Agriculture; Ca, Calcium; ICP-MS, Inductively coupled plasma mass spectrometry; K, Potassium; Kg, Kilogram; LOD, Limit of Quantification; LOQ, Limit of Detection; MEAL, Meals for exposure assessment and analysis of foods; Mg, Milligram; min, Minutes; ml, Milliliters; mLB, Modified lower bound; n, Number; NEMONIT, German National Nutrition Monitoring; NVS, German National Nutrition Survey; P, Phosphorus; p, P value for significance; SD, Standard deviation; TDS, Total diet study; UB, Upper bound; v/v, Volume percent; VELS, Verzehrsstudie zur Ermittlung der Lebensmittelaufnahme von Säuglingen und Kleinkindern (a nutrition survey for children); w/w, Weight by weight.

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regulation of Ca concentration in the blood (Peacock, 2010). Ca also acts as a second messenger in a variety of physiological processes (Berridge, Bootman, & Lipp, 1998).

K is found in a wide range of plant (e.g. vegetables, fruits and nuts) and animal foods (meat, poultry, fish, milk and dairy products) including beverages (e.g. fruit juices, coffee and tea) (Souci, Fachmann, & Kraut, 2016). Since K is water-soluble, losses of 20–50% might occur during processing of food (cooking, boiling) (Kimura & Itokawa, 1990).

K is present in all human tissues to maintain cell function. The absorption rate of dietary $K^+$ from the small intestine is about 90%. As the major cation of the intracellular space, it contributes to the maintenance of intracellular fluid volume and the transmembrane electrochemical gradient required for excitation transmission in neurons and contraction of muscles (Palmer, 2015). K is also important for numerous physiological functions (Palmer, 2015).

P is one of the most widespread nutrients in our diet. Protein-rich foods such as milk, meat, fish and eggs, but also nuts, seeds and legumes have frequently high P levels. The intestinal P absorption of ingested intake is about 55–80%, with the exception of plant seeds (e.g. beans, cereals, nuts), which contain P as largely indigestible phytic acid (inositol hexaphosphate), (Gutiérrez, 2020).

About 85% of the body's P reservoir is inorganic phosphate in hydroxyapatite together with Ca in bones and teeth. Organic P compounds are components of nucleic acids (DNA, RNA) and of membranes of cells and cell organelles. P is involved in numerous metabolic processes of the cell as they are regulated by phosphorylation reactions. Phosphate is also a component of intracellular messengers and involved to maintain the acid-base balance (pH) (Berndt & Kumar, 2009).

During food processing, a large number of Ca and K compounds as well as inorganic phosphates may be added to foods as food additives for technological and nutritional purposes (EC, European Commission, 2008). Inorganic phosphates particularly used in processed foods and carbonated soft drinks, are more readily and almost completely absorbed, compared to organic phosphates from foods naturally high in P. In vulnerable population groups, significant increase in dietary K and inorganic P may be associated with an increased risk of hyperkalemia and hyperphosphatemia, cardiovascular risk factors (Cupisti & Kalantar-Zadeh, 2013; Picard, Picard, Mager, & Richard, 2021).

In Germany, data to estimate chronic dietary exposure for chemical substances in foods are scarce. To improve the knowledge about levels of chemical substances in foods including Ca, K and P, the first German total diet study (TDS) called BfR MEAL Study (meals for exposure assessment and analysis of foods), was initiated in 2015 at the German Federal Institute for Risk Assessment (BfR) (Sarvan, Bürgelt, Lindtner, & Greiner, 2017). According to WHO recommendations, a TDS is the most cost-effective and reliable method for the analysis of chemical substances in foods for exposure assessment (EFSA, FAO, & WHO, 2011). The design of a TDS includes three criteria: i) representativeness of eating behaviour of a population, ii) foods are prepared as eaten and iii) similar foods are pooled to one sample. As domestic cooking techniques might affect the levels of substances in foods due to decomposition of chemical substances, the occurrence of breakdown products and the formation of new substances, different food preparation behaviours were taken into account (EFSA et al., 2011). In comparison to other TDS, the BfR MEAL Study is one of the most extensive study worldwide based on the combination of foods and chemical substances analysed.

The objective of this paper is to present levels of Ca, K and P in foods consumed by the population in Germany and analysed within the BfR MEAL Study. Moreover, the BfR MEAL Study is supposed to determine differences between regionally and seasonally sampled MEAL foods (P) as well as between organical and conventional type of production (Ca, K, P) as this information is scarce. The results described in this study are foreseen to be used for dietary risk assessment for the population in Germany by combining the levels of chemical substances in foods measured in the BfR MEAL Study with representative consumption data.

## Material and methods

### Selection, purchasing and preparation of foods

The design of the BfR MEAL Study followed international recommendations for TDS as described elsewhere (Sarvan et al., 2017). Most current and comprehensive consumption data were used from the National Nutrition Survey II (NVS II; n = 13,926; 24 h recalls; 14 to 80 years) (Heuer, Krems, Moon, Brombach, & Hoffmann, 2015) and consumption survey for children (VELS, n = 804; 24 h recalls; 0.5 to < 5 years) (Banasiak, Heseker, Sieke, Sommerfeld, & Vohmann, 2005) to establish the MEAL food list. Due to missing data, consumption habits of 5 to 13 years old children were not explicitly considered to derive the MEAL food list. The MEAL food list includes 356 MEAL foods which were assigned to 19 main food groups. Foods are processed and prepared as consumed and referred to as MEAL foods in the present study. At least 90% of the German diet was covered for different age groups and genders (Sarvan et al., 2017). Rarely consumed (<10%) but potentially highly contaminated foods were included in the MEAL food list as well.

Purchasing, preparation and homogenization of samples in order to represent German consumer behaviour were performed between 2017 and 2019 as described previously from our group (Hackethal, Kopp, Sarvan, Schwerdtle, & Lindtner, 2021; Sarvan, Kolbaum, Pabel, Buhrke, Greiner, & Lindtner, 2021). In total, 356 MEAL foods were analysed for Ca, K and P. If differences between regionally, seasonally and organically or conventionally produced MEAL foods were expected, foods were stratified accordingly.

For P, 869 pooled samples were analysed. If no differences were expected between regionality, seasonality or type of production, MEAL foods were sampled in the area Berlin. These MEAL foods were sampled throughout the year (not in special seasons) and the pooled samples of these foods could be composed of both organic and conventional food items (subsamples). If differences were expected, MEAL foods were sampled according to regionality (n = 70 for each region), seasonality (n = 58 for each season) as well as organic or conventional type of production (n = 105 for each type of production). Regionally sampled foods were purchased in four different regions of Germany (east, south, west, north) without considering their origin. Seasonally sampled foods were purchased at two different times of the year to integrate the influence of global food supply in the sampling of foods. Foods may originate from geographically different areas or may be influenced by various production conditions such as climate, soil and type of animal husbandry during the year. The composition of organically and conventionally pooled samples depends on market and consumption data as well as market availability. Thereof, pooled samples of organically and conventionally produced pooled samples might differ. For ingredients that constitute <5% of recipes (w/w), the top brand according to market share data was purchased regardless regionality, seasonality and type of production.

In total, 493 pooled samples were analysed for Ca and K, respectively. For Ca and K analysis, regionally and seasonally retrieved samples were aggregated to one individual pooled sample. Therefore, aggregated pooled samples consist of up to 120 subsamples. In contrast, as no differences were expected between regionality, seasonality or type of production, MEAL foods were sampled in the area Berlin. A distinction was made between 105 MEAL foods sampled according to organic and conventional type of production each.

To represent consumer's shopping behavior, pools without considering regionality, seasonality and type of production were composed of 20 subsamples. MEAL foods discriminated between regionality and seasonality or type of production were composed of at least 15 individual subsamples. The number of subsamples for each pooled sample is listed in Tables S3–S5. For the preparation of MEAL foods, drinking water from the MEAL study kitchen was used. It was sampled in the study kitchen at four different time points. Additionally, 29 different regions in Germany (29 pooled samples) with one subsample each were sampled.

### Determination of phosphorus

The analysis of P was undertaken by an accredited contract laboratory.

Samples were digested using a microwave according to DIN EN 13805:2014 (German Institute for Standardization e.V.; Berlin) and analysed as part of the multi-element method using an inductively coupled plasma–mass spectrometry (ICP-MS) as described (Hackethal et al., 2021) and summarized briefly in the supplementary document (Supplement material).

Due to low levels of P in samples of drinking water (tap water) which were close to the limit of quantification (LOQ), samples were additionally analysed photometrically. For photometric analysis, LCK 349 kit (Hach Lange GmbH, Düsseldorf, Germany) according to DIN EN ISO 6878:2004 (D) (German Institute for Standardization e.V.; Berlin) was used. Sample preparation and analysis were performed according to manufacturer's instructions. In brief, samples (2 ml; pH 2–10) were offset with peroxidsulfate and incubated at 100 °C for 60 min. After cooling to room temperature, 0.2 ml ascorbic acid and a solution containing antimony and molybdenum were added. After a 10 min incubation, levels of P were determined at a wavelength of 880 nanometer (nm) using a photometer (DR 3900, Hach Lange). Limit of detection (LOD) and LOQ were 0.020 mg $kg^{-1}$ and 0.050 mg $kg^{-1}$, respectively.

### Determination of calcium and potassium

The determination of Ca and K levels in samples was executed by an accredited contract laboratory. Sample digestion was adapted to the food matrix. Liquid and solid samples as well as fatty and oily samples were prepared as described elsewhere (Hackethal et al., 2021) and briefly summarized in the supplementary document (Supplement material).

Fatty or oily samples with higher sample weight of 2.0–5.0 g ± 0.1 g were incinerated in a microwave muffle furnace. Temperature-time-protocols followed the instructions of the German official collection of method of analysis according to §64 German Food and Feed Act (Federal Ministry of Justice and Consumer Protection, 2005). After cooling to room temperature, samples were dissolved in 5 ml $HNO_3$ (65% v/v) and diluted with purified water (Sartorius arium® comfort 2, 18.1 MΩ cm, Satorius Lab Instruments GmbH & Co, KG, Göttingen, Germany) up to a total volume of 25 ml.

Levels of Ca, K and P were quantified by external calibration measured daily. As internal standard, Niobium (P) or Indium (Ca, K) were used (Merck KGaA). Concentration of internal standards (Niobium, Indium) were prepared according to manufacturer's instructions (ICP-MS iCAP Q or X-Series II: 20 μg/l; ICP-MS 7800: 60 μg/l). For analytical quality control, blank solutions and reference material (NIST 1849; milk powder) were included in every run. The measurement uncertainties were 15% (Ca, P) and 8% (K).

Further information on instrumental parameters as well as LOD and LOQ are listed in Tables S1 and S2, respectively.

### Statistical methods

Left-censored data (results reported below LOD and/or LOQ) were substituted as described by WHO (WHO, World Health Organization, 2009). For the calculation of the modified lower bound (mLB), results below LOD or below LOQ were replaced by zero or the value reported as LOD, respectively. For the calculation of the upper bound (UB), results below LOD were replaced by the value reported as LOD and results below LOQ and above LOD by the value reported as LOQ.

In this paper, the results are presented and discussed according to the mLB approach. The result of each pooled sample according to the UB approach is available in supplementary Tables S3–S5.

Levels of Ca, K and P for each pooled sample analysed are listed in supplementary Tables S3–S5, respectively. Various substance levels resulting from the analysis were arithmetically averaged to obtain an mLB result and an UB result for each pooled sample. For each MEAL food, results of one to ten pooled samples were available to calculate statistical parameters for MEAL foods sampled according to regionality, seasonality or type of production.

To calculate mean Ca, K, and P levels for main food groups, results of pooled samples for each MEAL food were arithmetically averaged. Mean levels for each MEAL food of the respective main food group were arithmetically averaged.

Standard deviation (SD) determined on main food group level reflect the variability between different MEAL foods of the respective main food group and do not consider the variation covered by subsamples. Firstly, subsamples are aggregated to a pooled sample during sample preparation and secondly, pooled samples are aggregated to MEAL foods for the calculation of statistical parameters.

To calculate mean values, median (50th percentile, P50), SD as well as minimum (Min.) and maximum (Max.) levels Microsoft Excel 2016 was used.

Statistical analysis were carried out using IBM® SPSS® Statistics (version 26). Significant differences between substance levels for all MEAL foods sampled in different regions and by production type were investigated, respectively. Averaged results for each food per region or production type were used. First, data sets were tested for normal distribution using Kolmogorov-Smirnov test showing data sets were not normal distributed. Secondly, significant differences between all MEAL foods sampled by the two production types (organic and conventional) as well as for all MEAL foods sampled in four different regions were tested using the Mann-Whitney-*U* test or using Kruskal-Wallis test, respectively. The significance level was set to 0.05. Seasonal differences between all MEAL foods sampled in season 1 and season 2 were not evaluated as seasons vary between foods and include different months of the year.

## Results and discussion

### Levels of calcium, potassium and phosphorus in MEAL foods

Ca was quantified in 489 (99%) pooled samples with the exception of corn oil/maize-germ oil, olive oil and conventional sunflower seed oil (Table 1).

The highest mean levels of Ca were found in decreasing order in the main food groups "milk and dairy products" (2288 mg $kg^{-1}$), "sugar, confectionery and water-based sweet desserts" and "legumes, nuts, oilseeds and spices" (Table 1). Ca levels in the other main food groups ranged from 1060 mg $kg^{-1}$ (food products for infants and toddlers) to 52.6 mg $kg^{-1}$ (alcoholic beverages).

Of all 356 MEAL foods analysed, the 15 foods with highest Ca levels are shown in Fig. 1. Levels ranged from 19,230 to 2465 mg $kg^{-1}$ (Fig. 1). The highest levels of Ca were measured in chewing gum (19230 mg $kg^{-1}$). High Ca levels were also found in different types of cheese including Emmental cheese, firm/semi–hard cheese, soft ripened cheese, goat cheese, processed cheese and sheep cheese with levels ranging from 9210 to 2900 mg $kg^{-1}$. Chia seeds and spices showed high Ca levels as well (5760 and 4278 mg $kg^{-1}$, respectively). Additionally, high levels of Ca were measured in two foods for infants (powder) as well (instant milk cereals for reconstitution with liquids and infant formulae, follow-on formulae for reconstitution with liquids). In two almond based foods, almond paste and almonds (sweet), high levels of Ca were found with 3035 and 2639 mg $kg^{-1}$, respectively. High Ca levels were also measured in tofu and condensed milk (2690 and 2465 mg $kg^{-1}$).

From 493 pooled samples analysed, K was quantified in 490 (99%) pooled samples (Table 2). The pooled samples with levels below LOQ belong to the main food group "animal and vegetable fats and oils". In

**Table 1**
Levels of calcium in the main food groups (mLB) [mg kg$^{-1}$].

| Main food group | Pooled samples (n) | MEAL foods (n) | <LOD/LOQ (%) | Mean* | SD* | Median* | Min.* | Max.* |
|---|---|---|---|---|---|---|---|---|
| Alcoholic beverages | 11 | 8 | 0 | 52.6 | 25.8 | 51.6 | 0.978 | 82.4 |
| Animal and vegetable fats and oils | 12 | 8 | 33 | 82.4 | 105 | 25.5 | 0.200 | 309 |
| Coffee, cocoa, tea and infusions | 12 | 9 | 0 | 299 | 429 | 120 | 53.6 | 1468 |
| Composite dishes | 73 | 52 | 0 | 502 | 448 | 240 | 132 | 1647 |
| Eggs and egg products | 4 | 2 | 0 | 537 | 18.1 | 537 | 519 | 555 |
| Fish, seafood and invertebrates | 30 | 30 | 0 | 328 | 287 | 167 | 19.1 | 1081 |
| Food products for infants and toddlers | 15 | 11 | 0 | 1060 | 1504 | 274 | 69.2 | 4235 |
| Fruit and fruit products | 29 | 22 | 0 | 191 | 193 | 113 | 45.8 | 816 |
| Fruit and vegetable juices and nectars | 12 | 10 | 0 | 82.2 | 25.7 | 82.8 | 32.5 | 125 |
| Grains and grain-based products | 55 | 40 | 0 | 635 | 904 | 412 | 30.5 | 5760 |
| Legumes, nuts, oilseeds and spices | 24 | 20 | 0 | 1227 | 1004 | 846 | 302 | 4278 |
| Meat and meat products | 47 | 35 | 0 | 119 | 57.0 | 105 | 41.9 | 361 |
| Milk and dairy products | 30 | 23 | 0 | 2288 | 2245 | 1207 | 803 | 9210 |
| Products for non-standard diets and food imitates | 8 | 7 | 0 | 975 | 720 | 794 | 347 | 2690 |
| Seasoning, sauces and condiments | 16 | 16 | 0 | 633 | 511 | 420 | 134 | 2035 |
| Starchy roots or tubers and products thereof | 12 | 8 | 0 | 191 | 102 | 130 | 92 | 338 |
| Sugar, confectionery and water-based sweet desserts | 18 | 15 | 0 | 2008 | 4654 | 680 | 3.85 | 19,230 |
| Vegetables and vegetable products | 47 | 34 | 0 | 465 | 495 | 281 | 45.3 | 1911 |
| Water and water-based beverages | 38 | 6 | 0 | 68.7 | 34.7 | 74.4 | 19.7 | 110 |
| **Total** | **493** | **356** | **1** | **643** | **1384** | **267** | **0.200** | **19,230** |

*Left censored-data were analysed using the modified lower bound (mLB) approach. Results below LOD were replaced by zero and results above LOD and below LOQ were set to the value reported as the LOD.

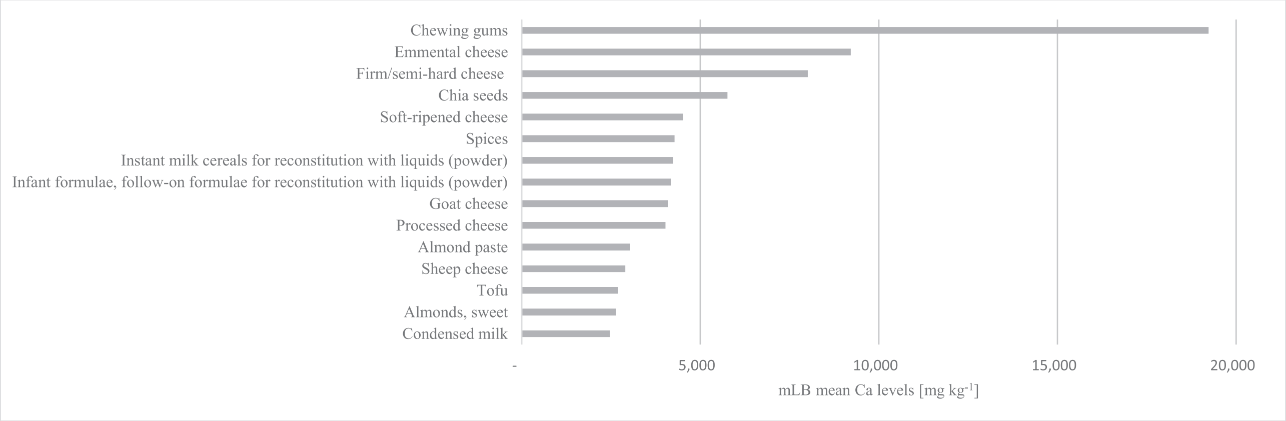

**Fig. 1.** Highest mean levels of calcium in 15 out of 356 MEAL foods analysed (mLB) [mg kg$^{-1}$].

**Table 2**
Levels of potassium in the main food groups (mLB) [mg kg$^{-1}$].

| Main food group | Pooled samples (n) | MEAL foods (n) | <LOD/LOQ (%) | Mean* | SD* | Median* | Min.* | Max.* |
|---|---|---|---|---|---|---|---|---|
| Alcoholic beverages | 11 | 8 | 0 | 478 | 284 | 478 | 2.08 | 984 |
| Animal and vegetable fats and oils | 12 | 8 | 25 | 195 | 239 | 195 | 0 | 607 |
| Coffee, cocoa, tea and infusions | 12 | 8 | 0 | 5449 | 11,509 | 5449 | 27.1 | 36,975 |
| Composite dishes | 73 | 52 | 0 | 2152 | 887 | 2152 | 752 | 4967 |
| Eggs and egg products | 4 | 2 | 0 | 1428 | 119 | 1428 | 1309 | 1547 |
| Fish, seafood and invertebrates | 30 | 30 | 0 | 2534 | 940 | 2534 | 423 | 4034 |
| Food products for infants and toddlers | 15 | 11 | 0 | 2453 | 1892 | 2453 | 139 | 6655 |
| Fruit and fruit products | 29 | 22 | 0 | 2497 | 1977 | 2497 | 980 | 8228 |
| Fruit and vegetable juices and nectars | 12 | 10 | 0 | 1131 | 431 | 1131 | 519 | 1895 |
| Grains and grain-based products | 55 | 40 | 0 | 2322 | 1474 | 2322 | 434 | 7118 |
| Legumes, nuts, oilseeds and spices | 24 | 20 | 0 | 6201 | 3280 | 6201 | 487 | 16,238 |
| Meat and meat products | 47 | 35 | 0 | 2897 | 759 | 2897 | 1703 | 4575 |
| Milk and dairy products | 30 | 24 | 0 | 1606 | 709 | 1606 | 608 | 3653 |
| Products for non-standard diets and food imitates | 8 | 7 | 0 | 2151 | 1701 | 2151 | 161 | 5995 |
| Seasoning, sauces and condiments | 16 | 16 | 0 | 2032 | 1185 | 2032 | 71.6 | 5065 |
| Starchy roots or tubers and products thereof | 12 | 8 | 0 | 4493 | 2671 | 4493 | 1755 | 10,840 |
| Sugar, confectionery and water-based sweet desserts | 18 | 15 | 0 | 2449 | 2102 | 2449 | 12.7 | 7225 |
| Vegetables and vegetable products | 47 | 34 | 0 | 2725 | 2305 | 2725 | 1168 | 15,030 |
| Water and water-based beverages | 38 | 6 | 0 | 122 | 199 | 122 | 2.13 | 562 |
| **Total** | **493** | **356** | **1** | **1990** | **2713** | **1990** | **0** | **36,975** |

*Left censored-data were analysed using the modified lower bound (mLB) approach. Results below LOD were replaced by zero and results above LOD and below LOQ were set to the value reported as the LOD.

this group, 25% of results were below LOD/LOQ.

High mean levels of K per main food group were found in “legumes, nuts, oilseeds and spices” (6201 mg $kg^{-1}$) followed by the groups “coffee, cocoa, tea and infusions” and “starchy roots or tubers and products thereof” (Table 2). Among the other main food groups, levels ranged from 2897 mg $kg^{-1}$ (meat and meat products) to 122 mg $kg^{-1}$ (water and water-based beverages).

Of all 356 MEAL foods analysed, the 15 foods with highest K levels are shown in Fig. 2. Levels ranged from 36,975 to 7090 mg $kg^{-1}$ (Fig. 2). By far, highest levels of K were measured in cocoa powder (36975 mg $kg^{-1}$), followed by spices (16238 mg $kg^{-1}$), vegetable crisps (15030 mg $kg^{-1}$) and potato crisps (10840 mg $kg^{-1}$). High K levels were found in MEAL foods of the main food group “legumes, nuts, oilseeds and spices” including pistachio nuts (9750 mg $kg^{-1}$), pumpkin seeds (9220 mg $kg^{-1}$), trail mix (tree nuts, oilseeds and raisins) (7495 mg $kg^{-1}$), almond paste (7265 mg $kg^{-1}$), hazelnut paste (7240 mg $kg^{-1}$) and sunflower seeds (7090 mg $kg^{-1}$). Cocoa beverage-preparation (instant, powder) also exhibited high K levels with 9515 mg $kg^{-1}$. In two fruit products (grapes and dried fruits), high levels of K were observed as well with 8228 and 7590 mg $kg^{-1}$, respectively. Similar levels of K were measured in dark chocolate and chia seeds (7225 and 7118 mg $kg^{-1}$).

From 869 pooled samples analysed, P was quantified in 841 (97%) pooled samples (Table 3). The highest percentage of results below LOD/LOQ was found in the main food group “water and water based beverages” (59%) followed by “animal and vegetables fats and oils” (23%) and “alcoholic beverages” (9%). In the remaining main food groups, all results were above LOQ.

High mean levels of P were found in “legumes, nuts, oilseeds and spices” (3593 mg $kg^{-1}$) followed by the groups “eggs and egg products” and “milk and dairy products” (Table 3). Among the other main food groups, levels ranged from 2047 mg $kg^{-1}$ (meat and meat products) to 8.85 mg $kg^{-1}$ (water and water-based beverages).

Of all 356 MEAL foods analysed, highest levels of P were measured in pumpkin seeds (11385 mg $kg^{-1}$), processed cheese (8433 mg $kg^{-1}$) as well as in chia seeds (7701 mg $kg^{-1}$) and sunflower seeds (7635 mg $kg^{-1}$) (Fig. 3). High levels of P were also found in nuts and seeds, ranging from 5450 to 4960 mg $kg^{-1}$ including pistachio nuts, cashew nuts and almonds (sweet). Additionally, almond paste contained P on high level as well (3772 mg $kg^{-1}$). In linseeds and oat flakes, levels of P were also high with 4647 and 3683 mg $kg^{-1}$, respectively. Among meat and meat products, high levels of P were measured in sheep liver with 3689 mg $kg^{-1}$.

Other European and international TDS as well as food databases reported similar levels of Ca, K and P in milk and dairy products (Ambrógi, Avegliano, & Maihara, 2016; Chekri et al., 2012; Food Composition Database Estonia, 2021; Food Composition Database Finland, 2021; Tanase, Griffin, Koski, Cooper, & Cockell, 2011). Milk is known as a food rich in Ca, K and P due to the high requirements for these elements by the neonate and suckling for growth and development (Shennan & Peaker, 2000).

In the present study, Ca and P levels in cheese were 3.7-fold to 3.8-fold higher than in milk (Table S3 and S5). Comparable results were reported by other studies (Chekri et al., 2012; Food Composition Database Estonia, 2021; Food Composition Database Finland, 2021). High levels of both elements in cheese can be attributed to the manufacturing process. During the processing of cheese, whey is separated from the thick milk resulting in the concentration of both elements (Belitz, Grosch, & Schieberle, 2008). Furthermore, Ca and P salts were added to improve coagulation properties (Lucey & Fox, 1993). Due to these additions of salts, Emmental cheese and firm/semi–hard cheese contain 50% more Ca and P compared to different types of soft-ripened cheese (Chekri et al., 2012; Food Composition Database Estonia, 2021; Food Composition Database Finland, 2021). To guarantee spreadability of processed cheese, P salts like diphosphate, sodium phosphate and polyphosphate were added during food processing. Therefore, high levels of P were found in processed cheese (Fig. 3).

For the main food group “legumes, nuts, oilseeds and spices”, similar results were reported by others (Ambrógi et al., 2016; Chekri et al., 2012; Food Composition Database Estonia, 2021; Food Composition Database Finland, 2021). In contrast, Tanase et al. (2011) reported lower mean levels of K in the food group “ingredients and sauces” (3599 mg $kg^{-1}$) including e.g. herbs and spices, mayonnaise and condiments (ketchup and mustard) in Canada whereas similar results were found for herbs and spices alone (15939 mg $kg^{-1}$) (Tanase et al., 2011). In France, Chekri et al. (2012) reported 1.8-fold lower levels of Ca in the product group “dried vegetables” including beans and lentils (539 to 295 mg $kg^{-1}$) (Chekri et al., 2012). The lower levels of Ca might be explained by a different composition of the product group analysed in both studies. In the French TDS, white beans ($n = 16$) and lentils ($n = 2$) were analysed whereas in the present study, kidney beans, chickpeas, lentils and canned pulses were analysed. Nevertheless, similar Ca levels were determined in beans comparing both studies. Differences in the composition of samples analysed might occur due to country specific consumption data serving as a basis for food selection.

High levels of Ca, K and P were found in the present study in chia seeds and nuts as reported in other studies as well (Figs. 1-3) (Food Composition Database Estonia, 2021; Food Composition Database Finland, 2021). Similar levels were reported in chia seeds by European studies and resulted from their origin. Chia seeds grow in tropical or subtropical climates and suppliers for Europe are mainly sourced from Paraguay, Nicaragua, Mexico and Bolivia (Kulczyński, Kobus-Cisowska, Taczanowski, Kmiecik, & Gramza-Michałowska, 2019). Due to drying, water content in chia seeds decreases and levels of Ca, K and P concentrate.

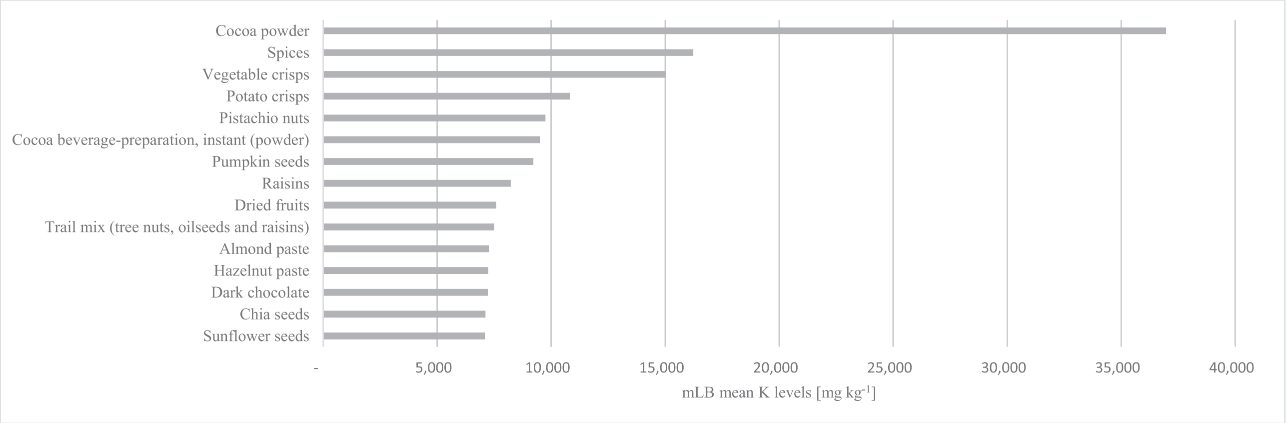

**Fig. 2.** Highest mean levels of potassium in 15 out of 356 MEAL foods analysed (mLB) [mg $kg^{-1}$].

**Table 3**
Levels of phosphorus in the main food groups (mLB) [mg $kg^{-1}$].

| Main food group | Pooled samples (n) | MEAL foods (n) | <LOD/LOQ (%) | Mean* | SD* | Median* | Min.* | Max.* |
|---|---|---|---|---|---|---|---|---|
| Alcoholic beverages | 11 | 8 | 9 | 130 | 69.3 | 130 | 0.300 | 262 |
| Animal and vegetable fats and oils | 13 | 8 | 23 | 82.7 | 102 | 29.9 | 0.300 | 269 |
| Coffee, cocoa, tea and infusions | 12 | 9 | 0 | 958 | 2049 | 46.6 | 3.48 | 6538 |
| Composite dishes | 170 | 52 | 0 | 938 | 522 | 836 | 118 | 2205 |
| Eggs and egg products | 10 | 2 | 0 | 2257 | 115 | 2257 | 2142 | 2372 |
| Fish, seafood and invertebrates | 39 | 30 | 0 | 1757 | 496 | 1676 | 682 | 2916 |
| Food products for infants and toddlers | 15 | 11 | 0 | 1305 | 1280 | 540 | 6.97 | 3662 |
| Fruit and fruit products | 64 | 22 | 0 | 245 | 193 | 183 | 82.7 | 887 |
| Fruit and vegetable juices and nectars | 12 | 10 | 0 | 92.9 | 40.6 | 89.5 | 31.0 | 177 |
| Grains and grain-based products | 97 | 40 | 0 | 1884 | 1325 | 1438 | 404 | 7701 |
| Legumes, nuts, oilseeds and spices | 24 | 20 | 0 | 3593 | 2536 | 3358 | 78.2 | 11,385 |
| Meat and meat products | 101 | 35 | 0 | 2047 | 600 | 2069 | 1007 | 3689 |
| Milk and dairy products | 37 | 23 | 0 | 2075 | 1970 | 1145 | 639 | 8433 |
| Products for non-standard diets and food imitates | 8 | 7 | 0 | 1055 | 567 | 960 | 250 | 1940 |
| Seasoning, sauces and condiments | 19 | 16 | 0 | 820 | 573 | 789 | 27.3 | 1828 |
| Starchy roots or tubers and products thereof | 26 | 8 | 0 | 564 | 195 | 481 | 368 | 920 |
| Sugar, confectionery and water-based sweet desserts | 18 | 15 | 0 | 1057 | 887 | 977 | 3.20 | 2390 |
| Vegetables and vegetable products | 152 | 34 | 0 | 420 | 302 | 318 | 135 | 1665 |
| Water and water-based beverages | 41 | 6 | 59 | 8.85 | 12.1 | 4.76 | 0.109 | 35.1 |
| **Total** | **869** | **356** | **3** | **1289** | **1379** | **970** | **0** | **11,385** |

*Left censored-data were analysed using the modified lower bound (mLB) approach. Results below LOD were replaced by zero and results above LOD and below LOQ were set to the value reported as the LOD.

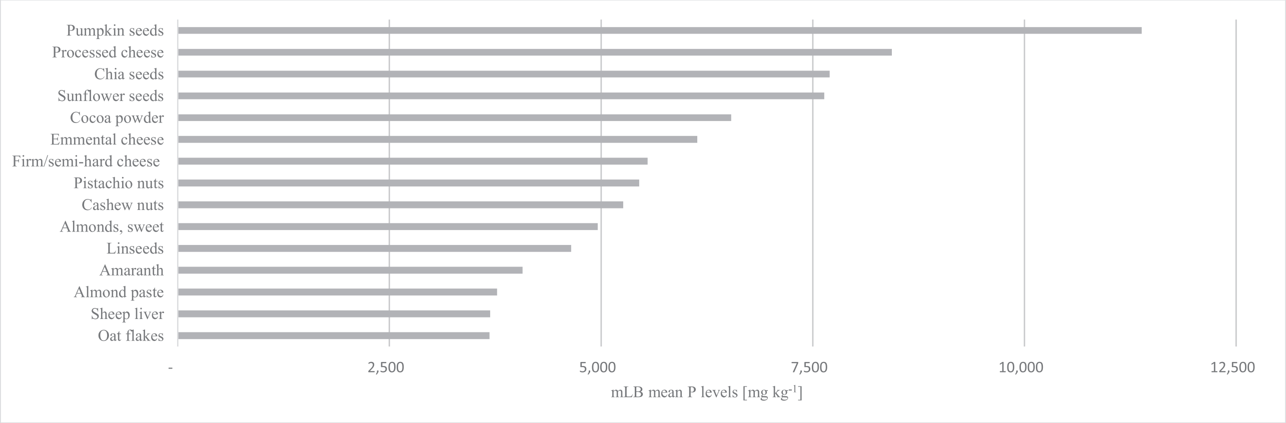

**Fig. 3.** Highest mean levels of phosphorus in 15 out of 356 MEAL foods analysed (mLB) [mg $kg^{-1}$].

In the present study, high Ca levels were measured in cheese and chewing gum as well (Fig. 1). For the latter, during manufacturing process, Ca-containing food additives (calcium carbonate and/or calcium phosphate) were added as coloring and textural agent (Jadhav & Mohite, 2014). As chewing gum is not a staple food in German households, it is not responsible for the daily Ca supply. Nevertheless, studies demonstrate anti–cariogenic effects of Ca and P supplemented sugar-free chewing gum (Reynolds, Cai, Shen, & Walker, 2003). In comparison to chewing gum, mean Ca levels in the other MEAL foods assigned to the main food group "sugar, confectionery and water-based sweet desserts" were lower ranging from 2380 (milk chocolate) to 3.85 mg $kg^{-1}$ (sugar). Therefore, high mean levels of this main food group can be primarily contributed to chewing gum.

As shown in the present study, high levels of K were found in unsweetened and sweetened cocoa powder. In line with data from the Finish food database, lower levels of K were determined in unsweetened compared to sweetened cocoa powder in both studies (Food Composition Database Finland, 2021). The high levels of K in unsweetened cocoa powder can be attributed to the use of potassium carbonate as declared on the packaging. In contrast, no K-containing food additives like potassium carbonate were declared on the packaging of the sweetened cocoa powder analysed. Furthermore, lower levels of K in sweetened cocoa powder can also be explained by its composition. Sugar is the main component of sweetened cocoa powder and consists of relatively low levels of K with 12.67 mg $kg^{-1}$ (Table S4).

Conclusively. the data presented in this study show that high levels of Ca, K and P were especially found in dry foods as well as in foods containing additives to increase quality (Table S3–S5).

### *Levels of calcium, potassium and phosphorus in MEAL foods sampled according to organic or conventional type of production*

In total, 105 MEAL foods were sampled according to organic or conventional type of production and analysed for Ca, K, and P. These MEAL foods are assigned to 16 main food groups of the BfR MEAL Study. The BfR MEAL Study is one of the first European TDS investigating differences between organically and conventionally produced foods. It is of special interest as the consumption of organically produced foods and thereof organic agriculture has considerably increased the last years. Different levels of Ca, K and P between organically and conventionally produced MEAL foods might be caused by various endogenous levels of these elements in the base products, as well as food processing, food supplementation and different composition of pooled samples.

No significant differences were found between all organically and conventionally sampled MEAL foods ($p \geq 0.05$) analysed for Ca, K and P, respectively (Fig. S2 A-C). Although not significantly different, varying levels of Ca and P were found in the main food group "products for non-standard diets and food imitates". In this main food group, only the MEAL food tofu was sampled according to organic and conventional type of production. In the organically produced tofu, higher levels of Ca and P were found compared to conventionally produced tofu (3645 to 1735 mg kg$^{-1}$ and 2397 to 1021 mg kg$^{-1}$, respectively) (Table S3 and S5). In line with our results, Paz and colleagues (2021) reported 2.0-fold higher Ca levels in organically produced tofu compared to conventionally produced tofu (Paz, Rubio, Gutiérrez, González-Weller, & Hardisson, 2021). Moreover, they also showed tofu originated from Europe has higher element content compared to tofu originated from China (Paz et al., 2021). This finding supports the observation obtained within the BfR MEAL Study because conventional subsamples originated mostly from China whereas organically produced tofu originated from Europe. The high levels of Ca and P in organically produced tofu can be attributed to three aspects: (i) A higher number of firm tofu containing more protein and less water compared to silken tofu resulting from different manufacturing processes. (ii) A higher number of smoked tofu. Due to thermal processing, mineral content is concentrated as a consequence of water loss in comparison to natural tofu. (iii) Country specific conditions such as the composition of the soil as well as temperature and climate influence endogenous mineral content in soybeans. In contrast, the usage of food additives like calcium sulfate or magnesium chloride do not explain the different levels of Ca and P between organically and conventionally produced tofu. Both additives function as coagulants and are added during processing of silken and firm tofu (Verhoeckx et al., 2015).

For the other main food groups, minor differences between organically and conventionally produced foods were found. These minor differences can be attributed to some foods of each main food group with different levels of Ca, K and P comparing organically and conventionally produced foods (Table S3–S5).

Varying mean levels of Ca, K and P were found in the main food group "legumes, nuts, oilseeds and spices" which can be attributed to the MEAL food olives. For Ca, higher levels were found in conventionally compared to organically produced olives (1030 to 390 mg kg$^{-1}$). For K and P, higher mean levels were found in organically compared to conventionally produced olives (763 to 212 mg kg$^{-1}$ and 101 to 55.4 mg kg$^{-1}$, respectively) (Table S3–S5). The 3-fold higher Ca levels in conventionally produced olives might be attributed to the application of calcium chloride as a food additive during fruit development and packaging to improve fruit quality and to increase preservability and texture (Morales-Sillero et al., 2021). In contrast, 3.6 to 1.8-fold higher levels of K and P were found in organically compared to conventionally produced olives, respectively (Table S4 and S5). Higher levels of K in organically produced olives can be explained by the addition of sea-salt as declared on the packaging of the foods analysed. In contrast, table salt was added to the sampled conventionally produced olives as declared on the packaging. Sea-salt contains higher levels of K because minerals like K were stripped during refining of table salt (Tan, Azlan, & Noh, 2016). Higher levels of P in organically produced olives might be due to the usage of phosphate rich fertilizers to maximize fruit load and optimize fruit quality and increase preservability (Erel, Dag, Ben-Gal, Yermiyahu, & Schwartz, 2011).

In the present study, mean P levels tend to be higher in organically compared to conventionally produced MEAL foods in the main food group "grains and grain-based products" which can be attributed to the MEAL food cereal cracker (puffed) (2710 to 450 mg kg$^{-1}$). For Ca and K, no differences were visible on main food group level. But levels of Ca and K in organically compared to conventionally produced cereal cracker (puffed) were higher (177 to 17.65 mg kg$^{-1}$ and 2897 to 821 mg kg$^{-1}$, respectively) (Table S3–S5). Higher levels in organically produced cereal cracker (puffed) might be explained by the high number of subsamples based on different types of grain such as corn, spelt as well as millet and the higher number of subsamples containing amaranth and linseeds. As shown in the present study, amaranth and linseeds contain high levels of Ca, K and P (Table S3–S5). In contrast, conventionally produced cereal cracker (puffed) were exclusively available on corn base without amaranth and linseeds.

For P, higher mean levels of organically produced foods in the main food group "food products for infant and toddlers" can be attributed to the MEAL food mixed cereal porridge and infant formulae for reconstitution with liquids (powder) (3025 to 2185 mg kg$^{-1}$) (Table S5). The higher levels in the organically produced food might be attributed to a different composition of the organic and conventional pooled samples, respectively.

In the MEAL food sunflower seed oil, higher mean levels of Ca and K were found in the organically compared to the conventionally produced sunflower seed oil (3.01 to 0.20 mg kg$^{-1}$ and 1.21 to 0.80 mg kg$^{-1}$, respectively) (Tables S3 and S4). The lower levels of Ca and K in conventionally produced sunflower seed oil might be attributed to the refining of conventional oil.

To compare organically and conventionally produced MEAL foods, the variability and availability have to be taken into account. In total, all pooled samples of organically and conventionally produced foods consist of at least 15 subsamples, each. As all subsamples are sampled representatively for the population in Germany, the composition of subsamples depends on the market data and market availability. For example, the organically produced pools sample cereal cracker (puffed) consists of 15 subsamples, with seven individual subsamples whereas the conventionally produced pooled sample consists of two individual subsamples. In contrast, the conventionally produced pooled sample beer consists of 14 individual subsamples whereas the organically produced pooled sample consists of six individual ones. The individual subsamples were conclusively sampled several times that each pooled sample consists of at least 15 subsamples in total. In both examples, market data define the subsample variety, as foods are sampled mimicking the German shopping behaviour. Based on the same consumption data, the individual subsamples of organically and conventionally produced pooled samples can differ due to availability and will represent the consumption habits and preferences of the consumers. For example, the pooled sample potatoes (pan-fried) should consist of ready-to-eat potatoes (pan-fried) from restaurants, as well as from self-made potatoes (pan-fried) prepared in the kitchen. The availability of potatoes (pan-fried) in restaurants from organically produced potatoes is low (as well as not controllable), these subsamples were replaced by recipes to ensure the usage of organically produced potatoes.

### *Levels of potassium in different potato products*

Within the BfR MEAL Study, potatoes (pan-fried) and fries/chips as well as boiled potatoes (unpeeled) and boiled potatoes (peeled) were prepared and analysed (Fig. S1). High levels of K were measured in potatoes (pan-fried) (5024 mg kg$^{-1}$) and fries/chips (4615 mg kg$^{-1}$). Lower K levels were found in boiled potatoes, unpeeled and peeled, with 4173 and 2955 mg kg$^{-1}$, respectively.

Cooking methods are important factors affecting the chemical composition and physical structure of potatoes (Tian, Chen, Ye, & Chen, 2016). Results of the present study were in line with others, reporting that levels of K decreased by 26–53% in boiled potatoes compared to fried potatoes and fries/chips (Tian et al., 2016). This effect can be attributed to the variety of the used potatoes, different preparation techniques like cutting into cubes, slices or strips as well as soaking or boiling of raw potatoes before frying. The loss of K during frying is almost nil due to its hydrophilic character and the dehydration of the surface retaining K inside the potato pieces (Fillion & Henry, 1998; Rojas-Gonzalez, Avallone, Brat, Trystram, & Bohuon, 2006). Additionally, Öhrvik and colleagues (2010) reported significantly lower K levels in boiled compared to raw potatoes as minerals pass into the boiling water (Öhrvik, Mattisson, Wretling, & Åstrand, 2010). In the present study, levels of K in raw potatoes were not determined because they are usually not consumed as such.

In the present study, levels of substances cannot be assigned to a specific cooking method as within one pooled sample, different preparation techniques were combined and taken into account proportionally according to consumption data. For example, the pooled sample potatoes (pan–fried) combined four different techniques: (i) Unpeeled or (ii) peeled potatoes were boiled before cutting and frying. Before frying, raw potatoes were cut into (iii) small cubes or (iv) slices. The combination of different preparation techniques is proportional to the use of these practices in private households and is accepted as a TDS collects representative data for the whole population.

The results presented in this study are relevant for both the public as well as dietary risk assessment. Moreover, they are of special interest for specific vulnerable population groups such as patients with comprised kidney function. These patients have to minimize their K intake and may benefit from the results especially those comparing domestic cooking techniques (Picard et al., 2021).

### *Levels of phosphorus in MEAL foods sampled according to regionality and seasonality*

Levels of P were determined in regionally and seasonally sampled MEAL foods. The 70 regionally sampled MEAL foods were purchased in four different regions of Germany (east, west, north, south). These 70 MEAL foods are assigned to nine main food groups of the BfR MEAL Study. Mean levels of P for each of the four regions for the nine main foods groups are shown in figure S3. No significant differences of P levels were found between MEAL foods sampled in the four regions of Germany ($p \geq 0.05$). For the main food group “water and water based beverages” including drinking water (tap water) and carbonated mineral water, mean levels of P were 6-fold higher in samples taken in region east, south and west (0.364 mg kg$^{-1}$) compared to samples taken in region north (0.064 mg kg$^{-1}$) (Fig. S3). Drinking water (tap water) taken in region west, high mean levels were determined compared to the other regions (0.480 to 0.032 mg kg$^{-1}$, respectively) (Table S5). Results of P levels in drinking water (tap water) samples by ICP-MS analysis were confirmed by photometric analysis (data not shown). For carbonated mineral water, high mean levels were found in samples taken in region east and south compared to region west and north (0.808 and 0.060 mg kg$^{-1}$, respectively). No differences were found between regionally sampled foods in the remaining main food groups.

Levels of P were mainly similar between regionally sampled foods. Foods were purchased in the four different regions of Germany to represent consumer behaviour without considering food origin. Therefore, imported foods were considered and purchased as well. Wholesale trades might import foods from the same international providers and therefore, similar results were found (Table S5). For regionally sampled drinking water (tap water), higher mean levels of P were determined in region west compared to region east, south and north (Table S5). In region west, six samples were taken, analysed and averaged to one sample. High levels of P in drinking water (tap water) sampled in region west can be mainly attributed to one sample with high levels of P in comparison to the other samples taken in region west (2.04 to 0.278 mg kg$^{-1}$). Regionally differences in drinking water (tap water) are inconclusive as those differences can be attributed to only one sample. Higher P levels in the drinking water (tap water) sample might be attributed to the following reasons: (i) usage of phosphate fertilization on agricultural areas (ii) P–containing pesticide residues of plant protection products (iii) natural variabilities.

Additionally, 58 MEAL foods were purchased in Germany at two different times of the year (season 1 and season 2) including vegetables, fruits, composite dishes and animal products (Table S5). For all 58 MEAL foods, similar levels of P were found comparing foods of season 1 and season 2.

Similar levels of P in seasonally sampled foods might be attributed to global food supply and storage of vegetables and fruits after harvesting. For example, the potatoes of season 1 were harvested and purchased in October whereas the potatoes of season 2 were purchased in May but might have been harvested in October as well.

### *Limitations and uncertainties of the study*

TDS are a recommended and powerful tool to determine average levels of chemical substances in foods as consumed to provide data for chronic dietary risk assessment to chemical substances (EFSA/FAO/WHO, 2011). Nevertheless, the specific design of TDS has to be taken into account to correctly interpret the results obtained.

Within a TDS, similar foods are pooled before analysis. Therefore, no evaluation about individual levels in single foods can be concluded. Pooling of foods is accepted as TDS collects representative data for the whole population.

For food preparation, drinking water (tap water) was used from the BfR MEAL study kitchen. Due to regional differences in levels of Ca, K and P in German drinking water (tap water), levels of those elements in drinking-water based foods might differ. Regional differences in foods prepared with drinking water (tap water) were not reflected within the BfR MEAL Study. Nevertheless, drinking water (tap water) was sampled 33 times in Germany.

The food consumption data used to establish the MEAL food list are based on 24 h protocols of two days (NVS II) and six days (VELS). Therefore, rarely consumed foods might be underrepresented (Sarvan et al., 2017).

## Conclusion

The determination of Ca, K and P in foods of the BfR MEAL Study provides a detailed data basis to estimate dietary exposure for the population in Germany. Foods, which were consumed by the population in Germany, were considered. Levels of Ca, K and P were determined in almost all 356 MEAL foods with high mean levels in the main food group “milk and dairy products” as well as “legumes, nuts, oilseeds and spices”. Moreover, chewing gum and cocoa powder contain high levels of Ca and K as well, respectively. The present finding tend to confirm the trend evidenced by studies carried out in other European and international countries. Levels of Ca, K and P determined in the MEAL foods can be attributed to natural origin and to food supplementation during processing. Within Europe, the BfR MEAL Study is one of the first TDS determining levels of Ca, K and P in organically and conventionally produced foods. Different levels were found in MEAL foods such as olives, tofu as well as cereal cracker (puffed). Similar levels of P were found in most MEAL foods regionally and seasonally sampled. Levels of Ca, K and P in MEAL foods sampled regionally, seasonally and according to type of production substantially improve data basis. Moreover, different domestic cooking techniques for potatoes were considered showing lower K levels in boiled compared to fried potatoes.

The data presented in this study combined with national consumption data provide a profound evidence base to estimate dietary exposure for the population in Germany and thus contribute substantially to consumer safety.

## CRediT authorship contribution statement

**Kristin Schwerbel:** Formal analysis, Investigation, Validation, Visualization, Writing – original draft. **Madlen Tüngerthal:** Investigation, Validation, Visualization, Writing – review & editing. **Britta Nagl:** Writing – review & editing. **Birgit Niemann:** Validation, Writing – review & editing. **Carina Drößer:** Methodology, Investigation, Writing – review & editing. **Sophia Bergelt:** Validation, Writing – review & editing. **Katrin Uhlig:** Validation, Writing – review & editing. **Tobias Höpfner:** Validation, Writing – review & editing. **Matthias Greiner:** Conceptualization, Writing – review & editing. **Oliver Lindtner:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing. **Irmela Sarvan:** Conceptualization, Project

administration, Methodology, Writing – review & editing.

## Declaration of Competing Interest

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

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2022.100221.

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