Grains, whole flour, white flour, and some final goods: an elemental comparison

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Abstract

Concentrations of 27 elements (B, Na, Mg, Al, P, S, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Mo, Cd, Sb, Ba, Hg, Pb, and U) in grain, flour, different bread and pasta samples, all bought in Austria, were determined with an inductively coupled plasma mass spectrometer. In total, we have investigated 73 grain and flour samples, including rye (*Secale cereale*), spelt (*Triticum spelta*) and wheat (*Triticum aestivum*), and additional 54 bread samples and 21 pasta samples. All investigated samples were within legal limits and show no health risk with respect to metal contamination. Furthermore, the importance of whole flour over white flour in bread and pasta to meet the daily requirements with respect to major and trace elements is demonstrated.

Keywords Grain · Flour · Bread · Pasta · Elemental composition

Introduction

In 2015, the typical Austrian consumed ~89.3 kg of grains [1]. That is significantly more than the average 65.7 kg/year/capita in Europe [2] and demonstrates the importance of grains in the Austrian diet. Soft wheat is per capita the main grain (58.8 kg), followed by maize and rye (12.7 and 9.3 kg, respectively) [1]. The high wheat consumption is a result of the vast field of products containing wheat as main ingredient like bread, countless bakery products, pasta and its use for thickening gravy and sauces. Spelt is with less than 1 kg/capita/year mainly for bread production of interest.

From the large amount of consumed grains, only a small part can be attributed to whole grain products. However, it has to be stated that there is no consistent definition of whole grain products across Europe. The European Union agricultural legislation referred to whole grains as “grains from which only the part of the end has been removed, irrespective of characteristics produced at each stage of milling” [3]. Nevertheless, for Italy, mean whole grain intakes were published as 2.1 g/day in children and adolescents, whereas adults consume approximately 3.7 g/day, which means less

than 1.5 kg/year [4]. For the French population, similar results were published. For whole grain consumers, 9.1 g/day for children and 14.4 g/day for adults were reported, but more than half of the respondents to the questionnaire explained to never consume any whole grain [5]. These values demonstrate rather low whole grain intakes, although whole flour products are more suitable to meet the daily requirement of several elements than white flour products. The Swedish National Food Agency for instance recommends a whole grain consumption of 70 g/day for women and 90 g/day for men [6]. A study from 2007 revealed that consumers regard cereal products as good for their health, but the awareness of whole grain products being healthier than white flour products varied between consumer groups [7]. There are enough data published referring to element concentrations in grains and flours, as shown in [Results and discussion](#). Furthermore, gluten-free products, ancient crops as well as the physical and sensory characteristics of whole grain products have been investigated recently [8–13]. However, a comparison of the elemental composition of whole flour and white flour from spelt, rye, and wheat as well as their final goods at once has not been done.

To give meaning to determined element concentrations in different foodstuff, it is useful to compare them to published recommended dietary allowance (RDA) or adequate intake (AI) values [14, 15]. RDA values express the average daily dietary intake level that is sufficient enough to meet nutrient

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requirements of 97–98% of healthy people in particular life stages and gender groups. In case an RDA value cannot be determined, AI values are set. They are based on observed or experimentally determined approximations of nutrient intake by a group of healthy people, which are assumed adequate.

The aim of this work is to compare the elemental composition, including bulk elements, trace elements as well as heavy metals, of different grains as well as the final goods bread and pasta. To our knowledge, this is the first paper to compare and discuss the concentration of 27 elements in grains, whole flour, and flour from rye, spelt and wheat grown and processed in Austria. Furthermore, we want to highlight the importance of whole flour, in comparison with white flour, to meet RDA, or AI values for trace elements. This knowledge should further increase the awareness of the importance of whole flour products over white flour products. Therefore, concentrations of B, Na, Mg, Al, P, S, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Mo, Cd, Sb, Ba, Hg, Pb, and U were determined and discussed.

Materials and methods

Reagents and materials

Ultrapure water, supplied by a Milli-Q purification system (Merck Millipore, Darmstadt, Germany) and nitric acid Rotipuran p.a. $\geq 65\%$ (Carl Roth, Karlsruhe, Germany) further purified via a quartz sub boiling distillation (EMLS, Leutkirch, Germany) were used throughout. Calibration standards and internal standard solution were prepared via dilution from Single Element Standards for ICP (Carl Roth, Karlsruhe, Germany). The reference materials RM 8414 (NCR RM 8414—Bovine Muscle Powder, National Research Council Canada, Ottawa, Canada), RM 1568b (NIST SRM 1568b—Rice Flour, National Institute of Standards and Technology, Gaithersburg, USA) and SRM 1640a (NIST SRM 1640a—Trace Elements in Natural Water, National Institute of Standards and Technology, Gaithersburg, USA) as well as a round-robin wheat reference material ALVA-PA15/3 (ALVA-PA15/3, AGES, Linz, Austria) were used to ensure accuracy of the results.

Samples

All samples were bought in Austria. Their origin and the water content are listed in Table 1. “Wheat pasta coloured” puts all pasta samples together, where different powders have been added to change the colour of the noodles, e.g., spinach or basil for green and tomatoes or beetroot for different shades of red. For grain, flour samples and pasta samples between 0.5 and 1 kg depending on the packaging size were

bought. Bread samples including different rolls and buns varied between 60 and 0.5 kg.

Sample preparation

For each sample, a representative aliquot, for instance, for bread two slices approximately 1 cm thick, was taken. At least 50 g of each sample were freeze-dried for about 5 days (Christ Beta 2-16, LMC-1, Fa. CHRIST, Osterode, Germany). If required samples were further homogenized with a mortar and pestle and mineralized in a pressurized microwave-assisted acid digestion system (UltraCLAVE III, MLS GmbH, Leutkirch, Germany), flour samples were mineralized without prior drying. Therefore, each sample was weighed three times (about 0.5 g weighed to 0.1 mg) in a 12 cm³ Quartz tube and 5 cm³ of nitric acid were added. Thereafter, the tubes were placed in the autoclave pressurized with argon (40 bars) and heated according to the following program: ramp in 10 min to 75 °C/in 15 min up to 150 °C/in 20 min up to 250 °C/30 min at 250 °C. After finishing the microwave heating, the solutions were transferred into 50 ml Cellstar PP-Test tubes (Greiner Bio-One International AG, Kremsmünster, Austria) and filled to the mark with ultrapure water. In each run, at least three reference materials and three blanks were included. To obtain the flour samples water content, approximately 5 g flour were put into glass petri dishes and dried to constant weight in a drying oven (Heraeus T5050, Heraeus Holding GmbH, Hanau, Germany) at 80 °C.

Element analysis

Element analysis was carried out with an inductively coupled plasma mass spectrometer (ICPMS Agilent 8800, Agilent Technologies, Waldbronn, Germany). The following isotopes were used for determination: no-gas mode: ¹¹B, ²⁷Al, ³¹P, ³⁴S, ⁹⁸Mo, ¹¹¹Cd, ¹²¹Sb, ¹³⁷Ba, ²⁰¹Hg, ²⁰⁸Pb, and ²³⁸U; helium mode: ²³Na, ²⁴Mg, ³⁹K, ⁴³Ca, ⁵¹V, ⁵²Cr, ⁵⁵Mn, ⁵⁶Fe, ⁵⁹Co, ⁶⁰Ni, ⁶⁵Cu, ⁶⁶Zn, ⁸⁵Rb, and ⁸⁶Sr; hydrogen mode: ⁷⁸Se; and oxygen mode: ⁷⁵→⁹¹As.

Quality control

To control instrument stability, a solution of ⁹Be, ⁷⁴Ge, ¹¹⁵In, and ¹⁷⁵Lu was continuously added online as internal standard. Furthermore, every 10th sample a control standard was measured. Analysis of the reference material RM 8414 “Bovine muscle powder”, RM 1568b “Rice flour” and the round-robin wheat reference material ALVA-PA15/3 found all elements in good agreement with the given estimate and information values (see Table 2).

Table 1 Analyzed grain samples and when needed flour-type number in parentheses, scientific name, origin (AT Austria, DL Germany, IT Italy, FR France), number of samples, and water content [%]

English name	Scientific name	Origin	n	Water content [%] $\bar{x} \pm \sigma$
Rye grain	<i>Secale cereale</i>	AT	13	5.5 ± 1.5
Rye flour		AT	9	10 ± 1
Rye whole flour		AT	3	10 ± 1
Rye flour bread		AT	9	39 ± 3
Rye flour bread with seeds		AT	3	35 ± 7
Rye whole flour bread		AT	5	42 ± 3
Spelt grain	<i>Triticum spelta</i>	AT	8	6.4 ± 1.4
Spelt flour		AT	3	11 ± 1
Spelt whole flour		AT	3	10 ± 1
Spelt whole flour bread		AT	7	35 ± 6
Wheat grain	<i>Triticum aestivum</i>	AT	9	6.2 ± 1.1
Wheat flour (480 ^a)		AT	14	11 ± 1
Wheat flour (700 ^a)		AT	6	11 ± 1
Wheat whole flour		AT	5	10 ± 1
Wheat flour-pretzel stick		AT, DL	6	30 ± 1
Wheat flour bread		AT, DL, FR	11	31 ± 4
Wheat flour bread with seeds		AT	13	31 ± 4
Wheat pasta	<i>Triticum durum</i>	AT, DL, IT	7	7.0 ± 0.4
Wheat pasta coloured		AT, DL, IT	10	7.0 ± 0.4
Whole wheat pasta		IT	4	8.5 ± 0.4

^aGerman and Austrian flour-type numbers indicate the amount of ash (in mg) obtained from 100 g of the dry mass of the flour

Statistical evaluation

For the different grain and flour types as well as bread and pasta samples, mean values and standard deviations were calculated (see Tables 3, 4). Because of the limited sample number further, statistical analysis was refrained. The element concentrations in 100 g of different flour types, bread samples, and uncooked pasta samples were compared to published recommended dietary allowance (RDA) or adequate intake (AI) values [14, 15] to estimate the approximate contribution for the daily requirement (see Table 5). RDAs and AIs are sex and age dependent. For calculations, values for males/females from the group of 31–50 years were used. Quotients shown in Fig. 1 were calculated as element concentration whole flour wet mass divided by element concentration white flour wet mass. In the case of bread, products out of spelt whole flour and wheat flour were compared.

Limits of detection (LOD) were calculated as the signal response of the blank solutions plus three times the standard deviation of the blank signals divided by the slope of the calibration. These concentrations were multiplied with the sample dilution. Values below 1 µg/kg are all expressed as 1 µg/kg.

Unless otherwise stated all concentrations presented in Results and discussion as well as values presented in Tables 2, 3 and 4 are expressed on a dry mass basis.

Results and discussion

Na, Mg, P, S, K, and Ca

Sodium concentrations in all investigated grain and flour samples are quite low with five samples, even below the limit of detection of 1 mg/kg. The highest observed concentration 9.5 mg/kg is still rather low, which would make grains dietary favourable, when not considering the addition of salt during further processing. Higher sodium concentrations in grains from 8 mg/kg up to 37.1 mg/kg were reported in several papers [16–18]. Whereas also all pasta samples did not exceed 0.32 g/kg sodium, bread samples showed high sodium concentrations up to 11 g/kg, which is related to the addition of salt during production.

Magnesium concentrations range from 0.15 g/kg up to 1.7 g/kg in grain and flour samples. Very similar values are reported in the literature with concentrations between 0.35 and 1.4 g/kg in wheat and wheat flour [17, 19–21]. For rye flour 1.0 g/kg and for spelt 1.5 g/kg were reported by Podio et al. [17] and Suchowilska et al. [18], respectively. Bread samples have comparable concentrations between 0.19 and 1.7 g/kg as the main flour type used. To meet the daily magnesium requirements, adults should consume between 300 and 420 mg/day depending on age and sex [14], which can be met by the consumption of

Table 2 Best estimate and information (*) values in RM 8414 “Bovine muscle powder” and given values for the reference material RM1568b “rice flour” and the wheat flour round-robin reference material ALVA-PA15/3

	RM 8414		RM 1568b		ALVA-PA15/3		LOD
	Given	Analyzed (<i>n</i> = 39)	Given	Analyzed (<i>n</i> = 15)	Given	Analyzed (<i>n</i> = 15)	
B (mg/kg)	0.6 ± 0.4	0.13 ± 0.05	–	0.41 ± 0.09	–	0.55 ± 0.11	0.03
Na (mg/kg)	2100 ± 80	2084 ± 184	6.7 ± 0.2	8.9 ± 2.2	31.3 ± 29.68	9.1 ± 1.5	1
Mg (g/kg)	0.96 ± 0.095	0.98 ± 0.07	0.559 ± 0.01	0.53 ± 0.03	1.35 ± 0.107	1.3 ± 0.1	0.001
Al (mg/kg)	1.7 ± 1.4	1.2 ± 0.5	4.21 ± 0.34	3.4 ± 0.2	–	1.5 ± 0.1	0.15
P (g/kg)	8.36 ± 0.45	8.8 ± 1.0	1.53 ± 0.04	1.5 ± 0.2	3.65 ± 0.161	3.8 ± 0.3	0.025
S (g/kg)	7.95 ± 0.41	8.0 ± 0.8	1.2 ± 0.01	1.0 ± 0.1	1.69 ± 0.196	1.5 ± 0.2	0.1
K (g/kg)	15.2 ± 0.4	14 ± 1	1.282 ± 0.011	1.2 ± 0.1	4.22 ± 0.326	3.9 ± 0.3	0.1
Ca (g/kg)	0.145 ± 0.02	0.15 ± 0.02	0.12 ± 0.003	0.11 ± 0.02	0.43 ± 0.04	0.43 ± 0.04	0.001
V (mg/kg)	0.005*	0.004 ± 0.006	–	0.005 ± 0.001	0.05 ± 0.008	0.043 ± 0.003	0.002
Cr (mg/kg)	0.071 ± 0.038	0.078 ± 0.032	–	0.036 ± 0.010	0.154 ± 0.039	0.13 ± 0.01	0.009
Mn (mg/kg)	0.37 ± 0.09	0.33 ± 0.03	19.2 ± 1.8	19 ± 2	38.1 ± 3.26	37 ± 5	0.012
Fe (mg/kg)	71.2 ± 9.2	65 ± 6	7.4 ± 0.4	7.6 ± 0.6	38.2 ± 3.78	38 ± 2	0.1
Co (mg/kg)	0.007 ± 0.003	0.007 ± 0.001	0.018 ± 0.001	0.016 ± 0.002	0.048 ± 0.004	0.045 ± 0.002	0.001
Ni (mg/kg)	0.05 ± 0.04	0.047 ± 0.018	–	0.20 ± 0.06	0.212 ± 0.04	0.18 ± 0.01	0.015
Cu (mg/kg)	2.84 ± 0.45	2.7 ± 0.2	2.35 ± 0.16	2.3 ± 0.2	4.99 ± 0.847	4.8 ± 0.3	0.05
Zn (mg/kg)	142 ± 14	164 ± 19	19.42 ± 0.3	21 ± 2	27 ± 1.98	30 ± 2	0.15
As (mg/kg)	0.009 ± 0.003	0.011 ± 0.002	0.285 ± 0.014	0.32 ± 0.01	0.187 ± 0.026	0.20 ± 0.02	0.002
Se (mg/kg)	0.076 ± 0.01	0.083 ± 0.015	0.365 ± 0.029	0.33 ± 0.02	0.136 ± 0.05	0.14 ± 0.02	0.01
Rb (mg/kg)	28.7 ± 3.5	26 ± 2	6.198 ± 0.026	5.3 ± 0.5	–	1.1 ± 0.1	0.005
Sr (mg/kg)	0.052 ± 0.015	0.056 ± 0.016	–	0.12 ± 0.02	1.179 ± 0.091	1.1 ± 0.1	0.01
Mo (mg/kg)	0.08 ± 0.06	0.071 ± 0.014	1.451 ± 0.048	1.38 ± 0.09	0.317 ± 0.025	0.31 ± 0.01	0.006
Cd (mg/kg)	0.013 ± 0.011	0.013 ± 0.003	0.022 ± 0.001	0.022 ± 0.001	0.112 ± 0.007	0.11 ± 0.01	0.005
Sb (mg/kg)	0.01*	0.002 ± 0.001	–	< 0.001	0.037 ± 0.002	0.037 ± 0.003	0.001
Ba (mg/kg)	0.05*	0.026 ± 0.009	–	0.12 ± 0.01	1.719 ± 0.1	1.7 ± 0.1	0.007
Hg (mg/kg)	0.005 ± 0.003	0.007 ± 0.002	0.006 ± 4E–04	0.004 ± 0.002	0.035 ± 0.005	0.030 ± 0.012	0.002
Pb (mg/kg)	0.38 ± 0.24	0.327 ± 0.067	0.01 ± 0.003	0.006 ± 0.001	0.183 ± 0.034	0.16 ± 0.02	0.004
U (mg/kg)	–	< 0.001	–	< 0.001	–	< 0.001	0.001

approximately 0.38–0.53 kg of whole spelt bread compared to 1.3–1.8 kg of pretzel sticks.

Phosphorus ranges from 2.9 to 5.4 g/kg and is far more present in grain and whole flour samples, with spelt showing the highest concentrations, than in white flour samples with a range between 0.76 and 2.8 g/kg. In the literature, concentrations between 1.4 and 5.4 g/kg in various grain samples were reported [18, 20, 21]. In pasta samples, phosphorus concentration is twice as high for whole flour pasta than for white flour pasta. The difference between whole flour and white flour is not so evident, although still present when examining bread products. In the case of bread products the consumption of 230–600 g of whole spelt flour bread is enough to meet the daily requirement, whereas 410–1000 g of wheat flour bread would be needed.

Sulfur shows, in contrast to phosphorus, less difference between flour and whole flour concentrations. Overall, the concentrations range between 0.78 and 2.1 g/kg for grain and flour samples and between 0.88 and 1.9 g/kg for bread

and pasta samples. Concentrations for total sulfur are scarce in literature. Suchowilska et al. reported a sulfur value of 1.8 g/kg for spelt, which is in good agreement with results obtained in this study [18].

Potassium, similar to phosphorus, shows with 3.0–4.8 g/kg for whole flour much higher concentrations than flour samples with 0.76–3.2 g/kg. All flour mean concentrations show a pronounced difference to the corresponding whole flour mean concentrations. Literature data for wheat flour were reported from 1.7 g/kg up to 4.9 g/kg [17, 18, 20–22]. In addition, bread and pasta with overall range from 1.0 to 5.5 and 1.8 to 4.4 g/kg, respectively, and show clear differences between whole flour and flour products.

Calcium is together with magnesium, phosphorus, sulfur, and potassium one of the main elements present in grains. The overall range from 0.13 to 0.46 g/kg for grains and flours is in good agreement with published results ranging between 0.14 and 0.65 g/kg [17–22]. 100 g of bread can provide between 2 and 4% of the daily requirement depending

Table 3 Elemental composition of different grain and flour types expressed on a dry mass basis

	Rye flour (<i>n</i> =9)	Rye whole flour (<i>n</i> =3)	Rye grain (<i>n</i> =13)	Spelt flour (<i>n</i> =3)	Spelt whole flour (<i>n</i> =3)	Spelt grain (<i>n</i> =8)	Wheat flour (480) (<i>n</i> =14)	Wheat flour (700) (<i>n</i> =6)	Wheat whole flour (<i>n</i> =5)	Wheat grain (<i>n</i> =9)
	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$
B (mg/kg)	0.82±0.15	0.91±0.12	0.94±0.08	0.18±0.02	0.45±0.03	0.45±0.07	0.15±0.04	0.16±0.04	0.51±0.05	0.40±0.07
Na (mg/kg)	3.4±1.1	1.9±0.2	2.5±1.5	4.7±1.1	7.3±1.8	4.3±1.5	3.4±0.8	5.5±1.3	6.6±1.4	6.7±2.5
Mg (g/kg)	0.59±0.09	1.0±0.1	1.02±0.05	0.40±0.03	1.3±0.1	1.3±0.1	0.21±0.07	0.40±0.11	1.4±0.2	1.4±0.1
Al (mg/kg)	5.7±2.9	1.4±0.7	1.5±0.9	2.4±1.8	4.0±3.7	0.86±0.39	2.0±1.3	1.7±1.4	1.7±0.5	1.3±1.0
P (g/kg)	2.1±0.4	3.3±0.2	3.2±0.2	1.8±0.1	4.4±0.4	4.4±0.5	1.01±0.20	1.3±0.2	3.9±0.5	3.8±0.3
S (g/kg)	0.92±0.11	1.4±0.1	0.99±0.13	1.3±0.1	1.4±0.1	1.4±0.3	1.1±0.1	1.1±0.1	1.3±0.1	1.2±0.1
K (g/kg)	2.8±0.2	4.4±0.2	4.4±0.3	1.5±0.1	3.6±0.3	3.7±0.3	1.01±0.14	1.3±0.1	3.6±0.6	3.4±0.3
Ca (g/kg)	0.29±0.03	0.37±0.04	0.36±0.04	0.26±0.01	0.40±0.01	0.32±0.03	0.19±0.03	0.19±0.05	0.39±0.08	0.33±0.07
V (mg/kg)	0.011±0.007	0.003±0.001	0.003±0.002	0.003±0.001	0.006±0.004	0.002±0.001	0.003±0.002	0.003±0.003	0.003±0.001	0.003±0.002
Cr (mg/kg)	0.024±0.011	0.031±0.012	0.009±0.004	0.015±0.003	0.019±0.008	0.009±0.004	0.011±0.006	0.014±0.008	0.020±0.010	0.014±0.014
Mn (mg/kg)	22±4	24±2	24±6	10±2	32±4	32±4	4.3±1.6	7.3±1.6	37±4	32±5
Fe (mg/kg)	20±1	23±1	22±3	14±2	39±2	35±4	7.1±3.2	11±2	34±3	27±2
Co (mg/kg)	0.007±0.002	0.010±0.001	0.007±0.004	0.007±0.005	0.012±0.005	0.010±0.008	0.001±0.001	0.002±0.001	0.010±0.006	0.008±0.006
Ni (mg/kg)	0.13±0.02	0.24±0.09	0.12±0.04	0.15±0.06	0.44±0.09	0.34±0.13	0.033±0.016	0.046±0.018	0.25±0.14	0.15±0.06
Cu (mg/kg)	2.7±0.4	3.5±0.2	3.4±0.3	2.2±0.1	4.9±0.2	5.2±0.7	1.4±0.2	1.8±0.2	4.4±0.6	3.7±0.5
Zn (mg/kg)	21±3	30±2	25±2	17±1	43±3	40±7	7.4±2.5	13±3	39±6	34±4
As (mg/kg)	0.006±0.003	0.002±0.001	0.004±0.002	<0.002	0.009±0.004	0.004±0.002	0.007±0.001	0.007±0.001	0.009±0.003	0.005±0.003
Se (mg/kg)	0.014±0.008	0.056±0.021	0.023±0.020	0.058±0.030	0.050±0.025	0.060±0.050	0.080±0.027	0.058±0.042	0.087±0.069	0.066±0.079
Rb (mg/kg)	3.6±5.6	2.2±0.2	2.4±1.1	10±2	23±5	7.9±6.9	4.2±2.1	3.5±3.5	15±11	4.0±3.8
Sr (mg/kg)	0.96±0.18	0.88±0.19	0.95±0.20	0.54±0.08	1.0±0.1	0.63±0.16	0.45±0.07	0.44±0.20	1.3±0.6	0.85±0.32
Mo (mg/kg)	0.34±0.15	0.60±0.06	0.62±0.37	0.32±0.05	0.47±0.05	0.55±0.31	0.38±0.13	0.61±0.14	0.81±0.49	0.99±0.50
Cd (mg/kg)	0.011±0.006	0.009±0.002	0.008±0.003	0.024±0.008	0.039±0.003	0.025±0.008	0.020±0.004	0.015±0.006	0.034±0.015	0.019±0.008
Sb (mg/kg)	0.001±0.001	0.002±0.001	<0.001	<0.001	0.002±0.002	<0.001	0.001±0.001	<0.001	0.002±0.001	<0.001
Ba (mg/kg)	2.8±1.4	1.9±0.2	2.3±1.5	1.2±0.3	3.1±0.1	1.9±0.5	0.78±0.28	0.66±0.36	4.1±1.3	2.6±1.5
Hg (mg/kg)	0.004±0.003	<0.002	0.003±0.003	<0.002	<0.002	0.002±0.001	<0.002	0.002±0.001	<0.002	0.003±0.003
Pb (mg/kg)	0.008±0.003	<0.004	<0.004	<0.004	<0.004	0.004±0.001	<0.004	<0.004	<0.004	<0.004
U (mg/kg)	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001

Table 4 Elemental composition of different bread and pasta products expressed on a dry mass basis

	Rye flour bread (n=9)	Rye flour bread with seeds (n=3)	Rye whole flour bread (n=5)	Spelt whole flour bread (n=7)	Wheat flour- pretzel stick (n=6)	Wheat flour bread (n=11)	Wheat flour bread with seeds (n=13)	Wheat pasta (n=7)	Wheat pasta col. (n=10)	Whole wheat pasta (n=4)
	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$	$\bar{x} \pm \sigma$
B (mg/kg)	0.69±0.10	1.4±0.4	0.95±0.10	1.4±0.7	0.30±0.19	0.21±0.03	1.0±0.5	0.43±0.61	0.59±0.37	0.40±0.05
Na (g/kg)	8.5±0.9	7.7±1.1	8.5±1.4	8.9±1.9	7.4±1.8	8.1±0.8	8.0±1.2	0.12±0.10	0.096±0.049	0.051±0.033
Mg (g/kg)	0.70±0.09	0.80±0.06	1.2±0.2	1.2±0.4	0.23±0.04	0.36±0.08	0.73±0.13	0.35±0.09	0.40±0.08	0.86±0.08
Al (mg/kg)	3.4±1.1	3.1±0.2	4.0±2.0	12±10	1.5±0.9	4.6±1.4	3.6±1.1	1.2±0.9	13±21	7.7±9.8
P (g/kg)	2.6±0.3	2.8±0.1	3.9±0.5	4.7±0.7	1.1±0.1	1.7±0.3	2.6±0.4	1.7±0.2	1.7±0.1	3.3±0.2
S (g/kg)	1.2±0.1	1.4±0.1	1.3±0.1	1.8±0.2	0.93±0.06	1.4±0.1	1.6±0.1	1.5±0.1	1.5±0.1	1.5±0.2
K (g/kg)	3.5±0.5	3.8±0.2	5.0±0.3	4.2±1.0	1.1±0.1	1.8±0.2	2.6±0.4	2.2±0.3	3.0±0.7	3.9±0.3
Ca (g/kg)	0.47±0.07	0.44±0.09	0.54±0.07	0.57±0.03	0.25±0.18	0.57±0.25	0.55±0.21	0.23±0.05	0.44±0.27	0.37±0.03
V (mg/kg)	0.005±0.001	0.006±0.001	0.011±0.008	0.041±0.037	0.004±0.002	0.009±0.004	0.006±0.002	0.003±0.003	0.023±0.024	0.012±0.015
Cr (mg/kg)	0.028±0.019	0.021±0.018	0.026±0.011	0.060±0.048	0.014±0.006	0.036±0.016	0.030±0.014	0.013±0.006	0.058±0.069	0.024±0.021
Mn (mg/kg)	24±2	18±2	35±6	29±9	6.2±0.8	11±3	17±4	7.2±2.8	8.1±1.6	22±3
Fe (mg/kg)	22±1	22±0	34±1	44±20	8.7±1.1	16±2	21±4	11±4	19±13	28±5
Co (mg/kg)	0.009±0.002	0.037±0.011	0.013±0.001	0.031±0.006	0.006±0.004	0.007±0.001	0.019±0.013	0.003±0.001	0.009±0.006	0.006±0.003
Ni (mg/kg)	0.14±0.03	0.34±0.13	0.25±0.07	1.5±1.1	0.018±0.006	0.060±0.036	0.40±0.27	0.092±0.032	0.12±0.04	0.17±0.04
Cu (mg/kg)	3.2±0.2	4.0±0.4	4.4±0.6	6.0±2.1	1.1±0.1	2.0±0.4	3.9±0.7	2.9±0.4	2.9±0.5	4.8±1.0
Zn (mg/kg)	26±1	26±3	34±3	37±10	8.1±0.9	13±3	22±5	14±5	11±3	31±6
As (mg/kg)	0.005±0.001	0.005±0.001	0.006±0.002	0.013±0.005	0.006±0.001	0.005±0.001	0.006±0.001	0.005±0.001	0.009±0.008	0.009±0.007
Se (mg/kg)	0.034±0.014	0.055±0.013	0.037±0.033	0.040±0.014	0.029±0.017	0.042±0.023	0.060±0.018	0.13±0.04	0.18±0.05	0.064±0.014
Rb (mg/kg)	2.2±0.4	2.0±0.4	3.6±1.0	2.9±1.1	0.63±0.05	1.1±0.3	1.8±0.9	1.1±0.3	1.2±0.2	3.2±2.4
Sr (mg/kg)	1.4±0.5	1.3±0.2	2.1±0.6	1.7±0.5	0.52±0.19	1.1±0.3	1.9±0.9	0.69±0.25	1.8±1.6	1.38±0.47
Mo (mg/kg)	0.52±0.13	0.62±0.09	0.51±0.04	0.42±0.02	0.21±0.05	0.30±0.11	0.43±0.05	0.54±0.26	0.60±0.25	0.60±0.11
Cd (mg/kg)	0.017±0.004	0.019±0.020	0.018±0.004	0.070±0.038	0.014±0.003	0.022±0.004	0.034±0.016	0.030±0.006	0.036±0.009	0.028±0.011
Sb (mg/kg)	0.001±0.001	0.001±0.001	0.001±0.001	0.002±0.001	<0.001	0.001±0.001	0.002±0.001	0.001±0.001	0.001±0.001	0.001±0.001
Ba (mg/kg)	1.8±0.4	1.3±0.1	2.5±0.4	2.2±1.1	0.59±0.13	0.98±0.26	1.4±0.3	0.91±0.12	1.3±0.3	1.9±0.4
Hg (mg/kg)	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002
Pb (mg/kg)	0.005±0.001	0.007±0.004	0.007±0.002	0.013±0.009	<0.004	0.005±0.001	0.005±0.001	0.009±0.006	0.012±0.014	0.009±0.005
U (mg/kg)	0.001±0.001	<0.001	0.001±0.001	0.002±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001

Table 5 Percentage of the daily requirement (RDA or AI for male/female between 31 and 50 years) that can be reached through the consumption of 100 g of different flour types, bread types and of raw pasta products

	Mg (%)	P (%)	Ca (%)	Cr (%)	Mn (%)	Fe (%)	Cu (%)	Zn (%)	Se (%)	Mo (%)
Rye flour	13/17	27	3	6/9	88/113	23/10	27	17/24	2/2	68
Rye whole flour	24/31	47	4	9/12	104/133	29/13	39	27/38	10/10	134
Spelt flour	9/11	23	2	5/5	40/51	16/7	22	14/19	9/9	64
Spelt whole flour	29/38	58	4	5/7	126/161	44/20	50	35/48	8/8	94
Wheat flour (480)	5/6	13	2	3/4	17/22	8/4	14	6/8	49/49	78
Wheat flour (700)	9/11	17	2	4/5	29/37	13/6	18	11/15	9/9	121
Wheat whole flour	31/41	51	4	5/7	147/187	38/17	44	32/44	14/14	162
Rye bread	10/13	23/23	3/3	5/7	63/80	17/8	22/22	14/20	4/4	71/71
Rye bread with seeds	12/16	26/26	3/3	4/6	52/66	18/8	29/29	16/21	7/7	90/90
Rye whole flour bread	16/21	33/33	3/3	4/6	88/113	25/11	29/29	18/25	4/4	66/66
Spelt whole flour bread	19/25	44/44	4/4	11/16	81/104	36/16	43/43	22/30	5/5	60/60
Wheat flour-pretzel sticks	6/7	16/16	2/2	4/6	27/34	11/5	12/12	7/10	5/5	46/46
Wheat flour bread	6/8	17/17	4/4	7/10	32/41	14/6	16/16	8/11	5/5	45/45
Wheat flour bread with seeds	12/16	26/26	4/4	6/8	49/63	18/8	30/30	14/19	8/8	65/65
Wheat pasta	8/10	23/23	2/2	4/5	29/38	13/6	30/30	12/17	23/23	113/113
Wheat pasta col.	9/12	22/22	4/4	16/22	33/42	22/10	30/30	10/13	31/31	125/125
Whole wheat pasta	19/25	44/44	3/3	6/9	88/113	32/14	49/49	26/36	11/11	123/123

Fig. 1 Quotient whole flour/flour for rye, spelt, wheat, bread, and pasta

on the flour used for production, although milk and milk products would be a more efficient calcium source.

Mn, Fe, and Zn

Manganese concentrations show high variations between whole flour and flour samples, except for rye. Rye flour samples are also rich in manganese and, therefore, very similar to whole flour samples. Overall concentrations for grains and flours range between 2.1 and 42 mg/kg. Several research groups have reported manganese concentrations mainly in wheat samples with concentrations between 8 and 87 mg/kg [16–24]. For bread and pasta, concentrations between 5.5 and 42 mg/kg could be observed with

rye whole flour bread showing the highest manganese concentrations. Both white flour and whole flour products are suitable to meet the daily requirement. Between 86 and 310 g of bread are enough to meet the recommended uptake.

Mean iron values are very similar to mean manganese values with an overall range between 3.1 and 42 mg/kg for grains and flours and between 7.6 and 65 mg/kg for bread and pasta samples. The literature is mainly available for wheat samples and reports concentrations between 13 and 103 mg/kg [16–22, 24, 25]. To meet the daily requirement, meat is known to be the most efficient iron source which means that especially vegetarians are in need of other iron suppliers. 100 g of spelt whole flour bread, for example, can provide up to 36% of the daily requirement for men and up to 16% for women in the age group 31–50 years providing all iron is bioavailable.

The overall range for zinc ranges from 3.8 to 50 mg/kg in grains and flours, which is similar to 7.2–48 mg/kg found in bread and pasta products. Like for several other elements, zinc concentrations are higher in whole flour than in white flour. Published zinc concentrations in grains range from 9.4 mg/kg up to 133 mg/kg, with most values below 50 mg/kg [16–27]. Zinc is one of the most important trace elements, which makes an adequate supply mandatory. Main zinc suppliers are on the one hand the animal products meat and cheese and on the other hand grains and their products. Approximately 330–450 g of spelt whole flour bread would be enough to meet the daily requirement.

B, Co, Ni, Cu, Se, and Mo

Boron concentrations range from 0.090 to 1.1 mg/kg in all investigated grain and flour samples. The obtained results are in good agreement with 0.43–1.0 mg/kg obtained for wheat samples in different Argentinean regions [17] and 0.7 mg/kg reported for spelt [18]. For bread and pasta samples, values between 0.16 and 2.2 mg/kg could be observed. Higher boron concentrations occur in various bread samples with sesame, sunflowers, or walnuts added. All 15 bread samples with sesame, sunflower, or walnuts added show a mean value of 1.5 ± 0.4 mg/kg, whereas all other bread samples ($n = 39$) show a mean value of 0.52 ± 0.28 mg/kg. This is not surprising as sunflowers and walnuts have a mean concentration of 11 ± 2 mg/kg boron as internal studies have shown.

Most grain and flour samples as well as bread and pasta samples show cobalt concentrations above the limit of detection but do not exceed 0.047 mg/kg and are, therefore, in good agreement with most cereal literature values with a range between 0.004 and 0.14 mg/kg [16, 17, 19, 21–23].

Obtained nickel concentrations range from 0.018 to 0.91 mg/kg with many concentrations below 0.1 mg/kg. Only four spelt whole flour bread samples show concentrations between 2.0 and 2.6 mg/kg. In the literature, flour and grain values up to 6.2 mg/kg are reported [27], but most concentrations are less than 1 mg/kg [16, 17, 21, 22, 24, 26].

Compared to other elements, copper concentrations are within a narrow range from 1.0 to 8.1 mg/kg. Our findings are again in good agreement with reported cereal literature values between 1.8 and 11 mg/kg [16–24, 26, 27]. Despite several vegetables and chocolate, grains are helpful to meet the daily copper requirement. 100 g of uncooked whole wheat pasta can supply up to 50% of the daily need.

Overall selenium values range for grain and flour samples between the limit of detection and 0.26 mg/kg and for bread and pasta samples between 0.011 and 0.24 mg/kg. Literature reports values from 0.0096 mg/kg up to 0.5 mg/kg for grains and flours [19, 22, 23, 28, 29]. Selenium shows great variations within the investigated groups, but does not show a tendency towards higher concentrations in whole flour. 100 g of uncooked pasta provide up to 30% and 100 g of bread up to 8% of the daily requirement.

Molybdenum concentrations are in good agreement with published literature. Determined concentrations range from 0.17 to 1.6 mg/kg for grains and flours, whereas literature reports concentrations of 0.42, 0.7, and 0.23–0.58 mg/kg [17, 18, 22]. Depending on the bread composition between 150 and 250 g are necessary to meet the daily requirement, whereas less than 100 g of uncooked pasta are also suitable to meet the daily need.

Al, V, Cr, As, Rb, Sr, Cd, Sb, Ba, Hg, Pb, and U

Aluminium concentrations for grains and flours are between 0.45 and 9.7 mg/kg and in good agreement with literature values such as 1.33–4.74, 4, and 5 mg/kg reported by Vrčec and Vinković Vrčec [22]; Podio et al. [17] and Ekholm et al. [21], respectively. For bread and pasta samples, most concentrations are below 5 mg/kg, but also a value of 70 mg/kg for coloured pasta was observed. This high value is most likely related to the added spinach powder. In general, aluminium concentrations do not show great differences between the investigated grain types and are, furthermore, typical for foodstuff. Seen alone, they are not likely to cause health problems, but contribute to the general aluminium uptake.

Vanadium concentrations are very low and range from the limit of detection up to 0.021 mg/kg for grain and flour samples and up to 0.093 mg/kg for bread and pasta samples. Vrčec and Vinković Vrčec [22] reported with 0.002–0.012 mg/kg in wheat flour very similar concentrations.

Chromium does not exceed 0.047 mg/kg for grains and flour and 0.20 mg/kg for bread and pasta samples and is, therefore, compared to grain literature concentrations, which range from 0.021 and 0.799 mg/kg, rather low [16, 22, 24, 26, 27]. The highest concentrations with 0.20 and 0.17 mg/kg were found in green and orange pasta samples.

Most arsenic concentrations are below or around the limit of detection with 0.006 mg/kg. Only four spelt whole flour samples and with 0.032 mg/kg one coloured pasta sample show higher concentrations. Reported literature concentrations are between 0.02 and 0.086 mg/kg and higher than the concentrations found in this study [19, 23, 26]. Only Vrčec and Vinković Vrčec [22] reported similar low values with a concentration range for wheat flour from 0.0002 to 0.0028 mg/kg. Arsenic concentrations found in wheat, spelt, and rye are low, especially when compared to rice, and with that not likely to be of any health concerns. Cubadda et al. [30] reported ten times higher concentrations for inorganic arsenic in rice than in wheat flour.

Rubidium shows large variations from one specimen to another, with an overall range between 0.54 and 30 mg/kg for grain and flour samples and 0.79 and 6.0 mg/kg for bread and pasta samples. Only few literature is available. Reported concentrations for grains and flours are 0.2, 1.1, and 1.7 mg/kg by Podio et al. [17]; Suchowilska et al. [18] and Bermudez et al. [16], respectively.

Obtained strontium concentrations for grains and flours with a range from 0.30 mg/kg up to 1.9 mg/kg are rather low compared to 2.8 and 3.6 mg/kg, reported by Podio et al. [17] and Suchowilska et al. [18]. Only Vrčec and Vinković Vrčec [22] reported lower values for wheat flour with a range between 0.0021 and 0.0034 mg/kg. For bread

and pasta samples, higher concentrations with an overall range from 0.50 to 4.8 mg/kg could be observed.

The European Commission established a maximum value for cadmium concentrations in grains. The concentration is set at 0.1 mg/kg wet mass for grains in general and 0.2 mg/kg wet mass for wheat in particular [31]. Even the highest observed concentration in grains and flour samples is with 0.046 mg/kg wet mass far below the given maximum. Although not regulated by the European Commission, most bread and pasta samples show concentrations below 0.050 mg/kg wet mass with the exception of again four whole spelt flour bread with concentrations up to 0.067 mg/kg wet mass. Reported literature concentrations range from 0.006 mg/kg up to 0.179 mg/kg [16, 17, 21, 22, 24, 32, 33]. The highest concentrations from 0.428 to 0.897 mg/kg were found for wheat grown on soil amended with domestic sewage sludge [27].

All investigated antimony concentrations are below or only slightly above the limit of detection, but do not exceed 0.004 mg/kg. The literature for antimony in grains is scarce, and only Bermudez et al. [16] reported 0.008 mg/kg for wheat.

With barium concentrations ranging from 0.34 mg/kg up to 6.3 mg/kg, obtained values are rather high compared to other foodstuff, but in good agreement with literature values ranging between 3.5 and 9 mg/kg [16–18].

Most mercury concentrations are below 0.002 mg/kg. Only some grain and flour samples show higher concentrations, but do not exceed 0.010 mg/kg. Huang et al. [26] reported values between 0.003 and 0.006 mg/kg.

In the case of lead, the European Commission also established a maximum allowed concentration of 0.02 mg/kg wet mass in grains [34]. Regarding the fact that all investigated grain and flour samples are below 0.012 mg/kg wet mass, no harm due to lead concentrations in Austrian grains is likely. This is also true for bread and pasta samples with lead concentrations below 0.011 mg/kg wet mass. Only one coloured pasta sample shows with 0.050 mg/kg wet mass a higher concentration. This could be caused by the use of spinach as colouring agent. In contrast to these results, some concentrations reported in the literature are well above the limit given by the European Commission. Literature data range between 0.002 and 1.6 mg/kg [16, 17, 21, 22, 24, 26, 32, 33]. The highest concentrations with 19.9–22.6 mg/kg were reported for wheat growing on soil amended with domestic sewage sludge [27].

All grain and flour uranium concentrations do not exceed the limit of detection of 0.001 mg/kg. For bread and pasta samples, some concentrations are above this limit but with a highest observed concentration of 0.003 mg/kg also quite low.

Elemental comparison whole flour–white flour

As shown in Fig. 1, whole flour is compared to the corresponding white flour richer in the essential elements magnesium, phosphorus, potassium, calcium, manganese, iron, cobalt, copper, zinc, and molybdenum. This not surprising, as with every sieving step, bran, which is very rich in essential elements, is removed from the flour.

The quotient between rye whole flour and rye flour for essential elements is, with values below two, the smallest. That can be explained by the higher flour-type number (more bran left) and, therefore, the smaller difference between element concentrations in the whole flour and flour. For rye flour, flour-type numbers around 1000 are typically on the Austrian market. Boron, sulfur, chromium, nickel, and selenium show with quotients of approximately 1.2, 1.6, 1.4, 2, and 4, respectively also higher concentrations in rye whole flour than in rye flour. Aluminium concentrations are with a quotient of 0.3 considerably lower in rye whole flour than in rye flour. For all other elements, no significant difference between rye whole flour and rye flour could be observed.

For the essential elements in spelt flour, quotients between 1.5 (sodium, calcium, and molybdenum) and 3 (manganese) were found (see Fig. 1). Higher concentrations in spelt whole flour than in spelt flour are also true for the elements boron (3), aluminium (2), vanadium (2), chromium (1.3), nickel (3), rubidium (2), strontium (1.9), cadmium (1.6), antimony (2), and barium (3). Sulfur, selenium, mercury, lead, and uranium are not significantly influenced by the flour type.

Wheat shows the highest quotients between element concentrations in whole flour and flour, with values up to 8.6 for manganese (see Fig. 1) in agreement with the flour-type number (in this case 480). That is why, the replacement of 125 g in 1 kg wheat flour with wheat whole flour can double the manganese concentration. For wheat flour, all elements show higher concentrations in whole flour samples than in white flour samples with the exception of aluminium, vanadium, selenium, mercury, lead, and uranium which do not show a significant difference. Additional to manganese also boron (4), sodium (2), magnesium (7), phosphorus (4), potassium (4), calcium (2), iron (5), cobalt (7), copper (3), zinc (5), rubidium (4), molybdenum (2), and barium (5) show quotients above 2. However, the heavy metals nickel and strontium are, by a factor of 8 and 3, respectively, also higher in wheat whole flour.

In general, for the essential elements, pasta samples, with a quotient up to 3 (manganese), show a larger difference between whole flour and flour products than bread samples. This can be explained by the lower water content in uncooked pasta than in bread (see Table 1), and therefore, the difference is more apparent. Nevertheless, whole flour bread is richer in elements than white flour bread as the following quotients demonstrate: boron (1.4), magnesium

(1.7), phosphorus (1.5), potassium (1.4), vanadium (2), manganese (1.5), iron (1.5), cobalt (1.4), copper (1.4), zinc (1.3), and rubidium (1.7). However, also heavy metals like nickel, strontium, antimony, and lead, with quotients between 1 and 2, tend to be higher in whole flour bread compared to white flour bread.

For pasta elements like magnesium (2), aluminium (7), phosphorus (2), vanadium (4.0), manganese (3), iron (3), zinc (2), arsenic (2), rubidium (3), strontium (2), and barium (2) show at least twice as high concentrations in whole flour pasta than in flour pasta.

In addition, element concentrations can be affected through contamination during milling/processing. Mainly, the concentrations of aluminium and chromium are affected, which are on average approximately twice as high in flours than in grains. Nickel, strontium, cadmium, and lead concentrations can also be affected.

Conclusion

Coloured pasta samples show higher standard deviations than their uncoloured counterparts because of the addition of different powders to change the noodle colour. In addition, on average, they contain higher amounts of aluminium, chromium, and vanadium. For grains and whole flours with their corresponding end products, it is demonstrated that they are in most cases far more suitable to meet the daily requirement of various elements than the so-called white flours with their products. In addition, bread types with seeds can further improve the daily elemental uptake compared to similar bread varieties without seeds. The amount of improvement is strongly dependent on the seed, seed amount, and flour type used, but can reach a factor of 3. Furthermore, all samples, independent from the type, showed no contamination due to heavy metals. To sum it up, the consumption of whole flour products, like whole flour bread or pasta, can increase the element uptake significantly and is, therefore, highly recommended.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Compliance with ethics requirements This article does not contain any studies with human or animal subjects performed by any of the authors.

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