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The iron stable isotope fingerprint of the human diet

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ABSTRACT The stable isotopes of iron disclose the metabolic pathways of iron within the human food chain. We have measured with precise multicollector ICP-MS the iron concentrations and stable isotope composition of 60 food products that are representative of the average German diet. We find that vegetable falls within the range typical of strategy I plants (-0.1 ‰ to -1.4 ‰ in δ^{56}Fe), crop products and processed crop food into the range typical of strategy II plants (-0.6 ‰ to +0.4 ‰), and animal products into the ^{54}Fe-enriched range known for animal tissue and blood (-1.1 ‰ to -2.7 ‰). Weighting these isotope compositions by the average iron dietary sources, we find a representative composition of European vegetarian diet of -0.45 ‰, while that of omnivores is -0.82 ‰. For human blood, known to be enriched in light iron isotopes, we find a fractionation factor for iron absorption of -2.0 ‰ to -2.3 ‰ for vegetarians (female and male, respectively), and -1.3 ‰ to -1.5 ‰ for omnivores (female and male, respectively). Knowing these fractionation factors is a prerequisite for using stable iron isotope ratios in blood as monitors of intestinal iron uptake regulation.

Introduction

Iron stable isotopes now begin to serve as tracers of nutrient uptake paths, processes, and efficiencies in humans [1]. All humans contain a specific iron stable isotope signature in their blood. This novel isotope biomarker was disclosed with the introduction of precise multicollector ICP mass spectrometry [2]. Human blood and muscle tissue are enriched in the light iron isotope, ^{54}Fe, by one to two per mil [‰] over the heavy iron isotope, ^{56}Fe, when compared to initial surveys of the human diet

[1-4]. As a model organism, the whole body of a minipig was found to be enriched by exactly 1.5 ‰ in ^{54}Fe over ^{56}Fe relative to its feed [5]. Yet the full exploitation of the potentially rich information contained in this biomarker is still impaired by our lack of an accurate description of the composition of the human diet. Knowing this composition is a prerequisite to a) estimate the extent to which an individuals unique blood composition is affected by that individuals diet; b) identify whether an individual bloods' composition is solely due to fractionation during absorption or due to internal distribution between organs [6, 7]; c) identify whether the blood of patients affected by hemochromatosis approaches that of their diet as expected if the degree of uptake is high [8]; d) single out whether ethnic differences in iron isotope composition, such as the reported difference between Swiss and Thai subjects [9] is due to metabolic processes or due to differences in the diet; e) attribute differences in the bloods iron isotope composition to uptake efficiency rather than to differences in the individuals diet [9]; and finally, explore whether a stable metal isotope fingerprint could serve as a tool for food authentication [10].

Mapping-out the iron stable isotope composition of the human diet is so difficult as all forms of food differ widely in their composition: plant food of "strategy I" plant origin (most non-graminaceous plants) encompasses a range of 0 ‰ to -3 ‰ in ^{56}Fe/^{54}Fe, given that these plants use a reductive uptake strategy [11-14]. These plants also differ in the composition of their different parts, with fruits and highest leaves typically contain the strongest enrichment of the light iron isotope, ^{54}Fe [11, 13, 14]. In contrast, "strategy II" plants (graminaceous plants) feature a narrower range (-0.5 ‰ to +0.2 ‰) in ^{56}Fe/^{54}Fe, and do not differ between their parts [11, 13, 14]. Initial surveys of animal food show that most meat sources, except liver, are enriched in the light iron isotope to a degree similar to human blood (-2 ‰ to -2.7 ‰ in ^{56}Fe/^{54}Fe) [2, 5, 7]. This similarity to blood is explained by the heme molecule containing most iron in both blood and muscle tissues.

In this study, we have measured the iron concentrations and stable iron isotope compositions of 60 food products, which we purchased in German supermarkets. These products are representative of the average German diet. To calculate the representative diet's composition, we have weighted these results by the Fe dietary sources described for the typical German "food basket" [15]. These estimates of the dietary iron intake are similar to those of the USA [16]. Finally, we have calculated typical fractionation factors for intestinal iron absorption in humans.

Materials and methods

Food sources. As our aim is to characterise the middle European diet, we have guided our food survey by the health survey of the German Robert Koch Institute using the survey tool DISHES 98 (Dietary Interview Software for Health Examination Studies) [15]. This report lists the 8 most important Fe sources that account for 75 % of the Fe intake (Figure 1). The remaining 25 % is distributed over a further 21 food sources. These are negligible as they either contain little iron or are consumed only rarely. The 8 most important groups differ in their relative proportions between men and women (Figure 1). 42 food products were obtained in 2007 in a conventional German supermarket (Table 1). In addition, 18 products grown under certified organic "biologisch" agricultural conditions were obtained to ensure that our sampling was representative over all consumed food sources. For four plant foods multiple

producers were measured, to provide a first estimate of source-specific variability. It must be noted that iron stable isotope measurements were developed to explore the composition of geological materials [17], mass-spectrometric machine time is still precious, and that the analysis of foodstuffs represents a formidable non-routine analytical effort and cost that still prohibits the analysis of a larger population of samples. This study can be hence considered as an upper limit for such survey at the current state of the methods capacity.

Sample preparation. All analytical work was undertaken at the Institute of Mineralogy at the Leibniz University of Hannover. All samples were dried down to accurately calculate the concentrations in the sample based on their dry weight. Plant samples were dried at 80 °C for two days in an oven, ground to mince if necessary and homogenized. As we did not check for the attainment of full dryness the iron concentrations reported in Table 1 should be regarded as good approximates. As the samples iron concentrations are not an aim of this survey, and isotope ratios do not depend on initial sample weights, this potential inaccuracy is immaterial to this study. Samples of animal origin were freeze-dried to -18 °C and the pressure was reduced to 0.3 mbar to sublimate the water.

Samples were prepared for iron isotopic analysis following the method of Schoenberg and von Blanckenburg [17]. About 300 mg of each sample was decomposed by microwave agitation at 200 °C in 8 mL of concentrated nitric acid (15 M) and 1 mL of 30 % hydrogen peroxide, evaporated on a hotplate in Teflon beakers and treated with 600 µL of 30 % H_2O_2 to oxidize any remaining organic compounds and ferrous iron to ferric iron. Solutions were clear after these steps and devoid of any solid residue. Due to the small amount of iron in some samples (less than 5 µg/g) 300 mg weighted sample were not sufficient for precise measurements of iron isotopes by MC-ICP-MS. These samples were decomposed in multiple batches that were recombined for measurements. The concentrations of iron and the inorganic matrix were determined using inductively coupled plasma optical emission spectrometry (ICP-OES). Following elemental analysis the samples were re-dissolved in 6 M HCl to separate the iron from the inorganic matrix by anion-exchange chromatography (resin *DOWEX® AG* 1X8, 100-200 mesh) with quantitative recovery, evaporated, and dissolved in 1 mL 0.3 M HNO_3 [17]. Previous studies revealed that Fe separates of samples with high transition metal contents or organic matrices may not be entirely matrix-free after anion-exchange chromatography and require further purification [17]. To ensure an interference-free measurement from the mass spectrometer an additional precipitation step was applied to ensure for a quantitative precipitation of all $Fe^{(III)}$ as $Fe^{(III)}OOH$ while Cu, Zn, Co, Cd, Mn, and V as well as organic compounds remain in solution. The samples were precipitated at pH ≈ 10 with 25 % NH_3 and solutions were equilibrated for 1 h before centrifugation. The supernate solutions were discarded; the precipitates were washed twice with *Milli-Q* H_2O and then re-dissolved in HNO_3. The Fe concentration was measured before and after the precipitation step with ICP-OES, to ensure a near-quantitative (±10 %) recovery during precipitation, which is essential to avoid Fe isotope fractionation. The samples were diluted to 3-6 µg/g Fe in 0.3 M HNO_3 for isotopic analysis.

Concentration measurements. Quantitative recovery and removal of matrix elements during iron separation and precipitation was controlled on small aliquots of

the dissolved samples before and after each step by ICP-optical emission spectrometry (ICP-OES: Varian Vista PRO CCD Simultaneous). After sample decomposition, the iron concentrations of all samples were obtained by ICP-OES on a dry-weight basis (Table 1). We estimate the total analytical uncertainty of the concentration measurements, comprising uncertainty of sample weight and the uncertainty of the ICP-OES measurement to be around 5 %. Total procedural blanks were also measured and were found to be between 12 ng and 27 ng – a negligible amount compared to the samples' Fe.

Iron isotope measurements. The determination of the ratios of the stable iron isotopes was carried out by a multiple collector inductively coupled plasma mass spectrometer (MC-ICP-MS; Neptune, Thermo Finnigan located at the Institute of Mineralogy at the Leibniz University of Hannover) according to the analytical protocol described by Schoenberg and von Blanckenburg [17]. The sample-standard bracketing technique was used to correct for instrumental mass bias using the Fe standard material IRMM-014, which was obtained from the Institute for Reference Materials and Measurements, Geel, Belgium, and has a certified $^{56}Fe/^{54}Fe$ isotope ratio of 15.69859. Iron isotope data are reported as $\delta^{56}Fe$ relative to the IRMM-014 standard of which the isotopic composition is close to that of rocks at the Earth's surface [18, 19] (defined as $\delta^{56}Fe = 0$), as defined in equation 1 below.

$$\frac{\delta^{56}Fe_{sample}}{‰} = \left(\frac{\left(^{56}Fe/^{54}Fe\right)_{sample}}{\left(^{56}Fe/^{54}Fe\right)_{IRMM-014}} - 1 \right) \cdot 1000 \qquad (1)$$

To check for molecular or elemental interferences, all $\delta^{56}Fe$ and $\delta^{57}Fe$ of the samples were plotted against each other and were found to follow a mass-dependent fractionation law.

As an additional quality check of isotope ratio measurements, a commercially available pure Fe wire (99.998 % purity, lot NM36883) from Johnson & Matthey (further on referred to as JM) has been measured throughout all mass-spectrometric sessions. 66 measurements of the internal "JM" standard gave the following results: $\delta^{56}Fe$ = 0.424 ± 0.065 ‰ (2σ); $\delta^{57}Fe$ = -0.610 ± 0.099 ‰ (2σ); $\delta^{58}Fe$ = 0.871 ± 0.434 ‰ (2σ). These values are identical to those reported by Schoenberg & von Blanckenburg [17]. 16 food samples were decomposed twice, to check for reproducibility and sample homogeneity. We achieved an external reproducibility of 0.059 ‰ for $\delta^{56}Fe$ and 0.107 ‰ for $\delta^{57}Fe$. In Table 1, we report the uncertainty of iron stable isotope ratios as the reproducibility of repeat measurements, or, the external reproducibility of the 16 repeat decompositions, whichever was larger.

Results and Discussion

Iron concentrations and iron isotope ratios. The analysed food products yielded a spread in iron concentration of 0.8 µg/g to 624 µg/g and in $\delta^{56}Fe$ from -2.7 ‰ to 0.39 ‰ (Table 1). The analysed strategy II plants yield the highest $\delta^{56}Fe$, between -0.56 ‰ and 0.39 ‰, and lower concentrations ranging from 5 µg/g to 56 µg/g. These ranges are similar to those found in growth experiments [11, 13]. The analysed strategy I plants contain mostly a lighter iron isotope composition than strategy II plants (-1.4 ‰ to -0.1 ‰) and iron concentrations mostly between 38 µg/g and 200 µg/g (with two exceptions that are discussed below). Again, these values are within the

range of growth experiments. The enrichment of ^{54}Fe in strategy I plants is due to the reduction during uptake [20, 21], which favours the light isotope (^{54}Fe) in the reduced compartment [22, 23].

Exceptions to these general rules are presented by rice and by tea. Both white and brown rice, even though being a strategy II plant, contains a light Fe isotope composition, with $\delta^{56}Fe$ of -0.9 ‰ and -1.2 ‰, respectively. This can be explained by the fact that rice, growing under anoxic conditions, does not suffer from Fe deficiency in waterlogged soils and is not required to invoke the $Fe^{(III)}$ complexing to combat deficiency otherwise characterising well-aerated soils [21, 24]. The case for tea, being a strategy I plant, yet featuring $\delta^{56}Fe$ of 0.1 ‰ to +0.4 ‰ and also unusually high concentrations of 120 µg/g to 350 µg/g, is more difficult to explain. One possibility is that tea leaves are contaminated during growth by atmospheric dust, containing iron with $\delta^{56}Fe$ of 0.1 ‰ to 0.2 ‰ [25]. Another possibility is that all the processing of tea (fermenting, drying, sieving) provides frequent exposure to steel surfaces in the absence of washing during processing.

Processed food (noodles, bread, rolls) fall into the strategy II range, which is expected as their main Fe source is flour from crop plants. However, these products are on average slightly (~0.1 ‰) lighter in their iron isotope composition than strategy II plants.

Animal products yield the largest range in both $\delta^{56}Fe$, ranging from -2.7 to -1.1 %, and their iron concentration, ranging from 10 µg/g to 620 µg/g. However, egg white and soft cheese contain even less Fe, with 1 µg/g to 2 µg/g.

In general, meat products are enriched in the light iron isotope (^{54}Fe), as expected for muscle tissue that contains mostly heme-bound iron and hence a light Fe isotope composition [1]. With $\delta^{56}Fe$ of -1.1 ‰, pork liver contains the heaviest iron isotope composition and with 624 µg/g also the highest iron concentration of all analysed animal products. Again, these values are expected as the liver is a store of ferritin-bound Fe that contains more ^{56}Fe than ^{54}Fe in blood [5].

The two fish samples yield contrasting results: while the herring contains Fe with $\delta^{56}Fe$ of -2.5 ‰ which is as isotopically light as iron in pork, beef or chicken muscle, the tuna contains Fe with $\delta^{56}Fe$ as heavy as -0.72 ‰. This comparatively heavy animal Fe isotope composition is only found in seafood, and is compatible with the heavy Fe isotope composition found in tuna and shrimp muscle in the initial survey of Walczyk & von Blanckenburg [2]. This phenomenon might be related to the high myoglobin content of tuna muscle. This feature sets tuna apart from most other fish.

The analysed chicken egg is interesting as Fe is fractionated between egg yolk ($\delta^{56}Fe$ of -2.4 ‰) and egg white ($\delta^{56}Fe$ of -1.5 ‰). The Fe concentrations are also vastly different, with only 1 µg/g for the egg white and 150 µg/g for the egg yolk.

In general, organically grown food features similar isotope ratios and concentrations than the counterparts sourced in conventional agriculture. Addition of extraneous iron to soil might potentially lead to deviations of $\delta^{56}Fe$ from the common range in these plants, as such additions potentially differ in their isotope composition to that of

"plant-available" iron in soil [10]. Also iron from average strategy II plants and processed food derived from strategy II plants cannot be explained by Fe fortification, as both the concentrations and isotope compositions are similar. These initial results might hint at stable iron isotopes possibly serving as tools for food authentication, but more food surveying is required to map out possible applications of this potential tool in detail.

The average middle European diet. When we average the found iron isotope ratios into the groups designated by the German "food basket" [15], we find the average values shown in Table 2 and Figure 3. These do not differ fundamentally from the average USA diet [16]. We mainly see two groups: animal products at δ^{56}Fe of around -2 ‰, and plant products around -0.7 ‰ to 0 ‰. Hence, it is necessary to calculate the average diet separately for omnivorous and vegetarian individuals. Weighting the isotope compositions of Table 2 with the relative dietary Fe intake we obtain a δ^{56}Fe of -0.77 ‰ for female omnivores and -0.46 ‰ for female vegetarians for their average diet. We obtain a δ^{56}Fe of -0.87 ‰ for male omnivores and -0.44 ‰ for male vegetarians as the composition of their average diet.

Isotope fractionation during intestinal absorption. To calculate the average fractionation factor for intestinal absorption, we need to take into account that heme-bound iron is absorbed with an efficiency of 15 % to 35 %, and non-heme iron absorption with 2 % to 20 % [16]. Non-heme iron absorption depends systematically on ferritin concentration and varies from 14 % at a ferritin concentration of 15 μgL^{-1} to 4 % at a ferritin concentration of 60 μgL^{-1} [26]. For our calculation we use an efficiency of 30 % for heme-bound iron, and of ca. 10 % for non-heme iron [1]. To take this effect into account, we weigh the mean diets isotope composition by these efficiencies and the iron consumption (Table 2). We used the average δ^{56}Fe of blood in young European adults that is based on three studies: Walczyk and von Blanckenburg [1, 2] yielded an average δ^{56}Fe of -2.78 ± 0.19 ‰ for 21 males and -2.45 ± 0.19 ‰ for 31 females, and Albaréde et al [4] an average δ^{56}Fe of -2.74 ± 0.16 ‰ for 22 males and -2.57 ± 0.19 ‰ for 25 females. The average of all samples from these three studies is -2.76 ± 0.18 ‰ for males and -2.50 ± 0.20 ‰ for females from which we obtained a fractionation factor for intestinal absorption of -1.3 ‰ for female omnivores and -2.0 ‰ for female vegetarians. We obtain a fractionation factor of -1.5 ‰ for male omnivores and -2.3 ‰ for male vegetarians, respectively.

That vegetarians and omnivores still do not differ in their bloods' composition can be explained by the fact that all heme-bound Fe passes through the intestinal mucosa without ligand exchange, thereby maintaining its light isotope composition, while all plant Fe, that is mostly present in the ferric form, is reduced in the intestine and is fractionated in the process to the heme Fe composition found in human blood. Hence, ultimately, all iron obtains a heme-like isotope composition.

The human food chain. It was shown by Walczyk & von Blanckenburg [1] that Fe is enriched as Fe is passed along the human food chain. This pattern was confirmed recently in a study of bones from herbivore and carnivore mammals [27]. Using our much more representative dietary dataset in conjunction with soil data that explicitly resolved the plant-available soil fraction [12, 28-30], we can confirm this picture (Figure 4). We see that soils contain a mobile fraction of -0.05 ± 0.2 ‰, which is reflected in the average of all strategy II plants (0.07 ± 0.25 ‰), taking a minor fractionation

towards ^{56}Fe by ligand exchange in the rhizosphere into account [12]. Adding strategy I plants yields an average vegetarian diet of -0.4 ‰ (see above). Animal food displays on average $\delta^{56}Fe$ of -1.9 ‰, while human blood and muscle tissue contains a $\delta^{56}Fe$ of roughly -2.6 ‰. Adding the liver as containing roughly 25 % of human iron, we get an approximate composition for the human body of -2.5 ‰. The human body is therefore the lightest member of the human food chain.

Finally, we note that stable isotope ratio measurements of metal and metalloid elements that is now routine with multicollector ICP-MS opens the possibility to explore the metabolism along the food chain in a variety of elements. This high potential with the possibility to serve in certain aspects of food authentication is demonstrated by promising biomarker experiments on plants that have been made for metals, for example for Mg [31, 32], Ca [33], Cu [7, 34], Zn [7, 34, 35], Ni [36], and for metalloids, for example B [37] and Si [38]. First steps have been made in mapping out these metal biomarkers, and these new avenues now need to be explored in detail.

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The full dataset summarised in Table 1 is available in the supplementary material. This information is available free of charge via the Internet at http: //pubs.acs.org.

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Author Contributions

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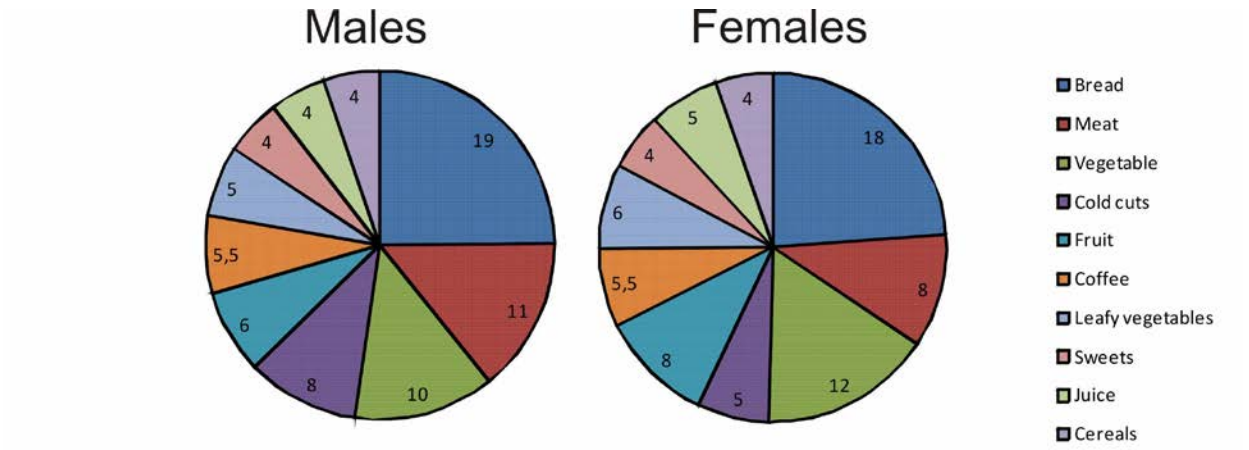

FIGURES

Figure 1. Components of the average German "food basket" [15], comprising 76.5 % (males) and 75.5 % (females) of the most important iron sources that are shown in percent.

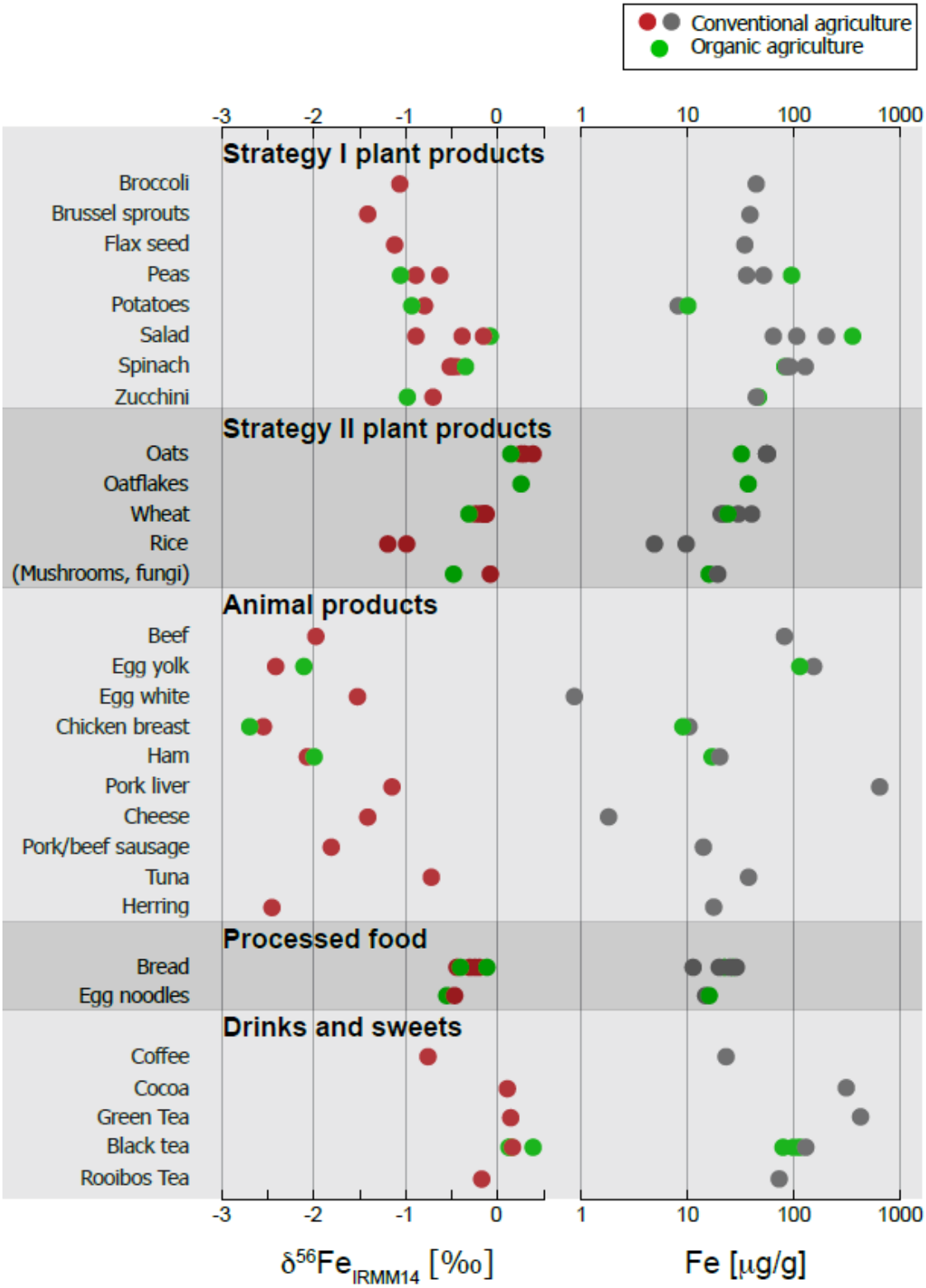

Figure 2. Iron isotope composition and iron concentrations (on a dry weight basis) of measured food products.

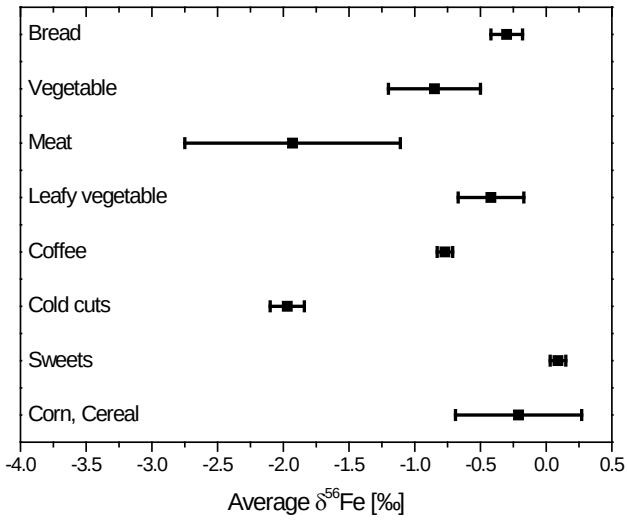

Figure 3. Average composition and standard deviation of the groups of diet using the relative amounts taken up and the isotope compositions from Table 2.

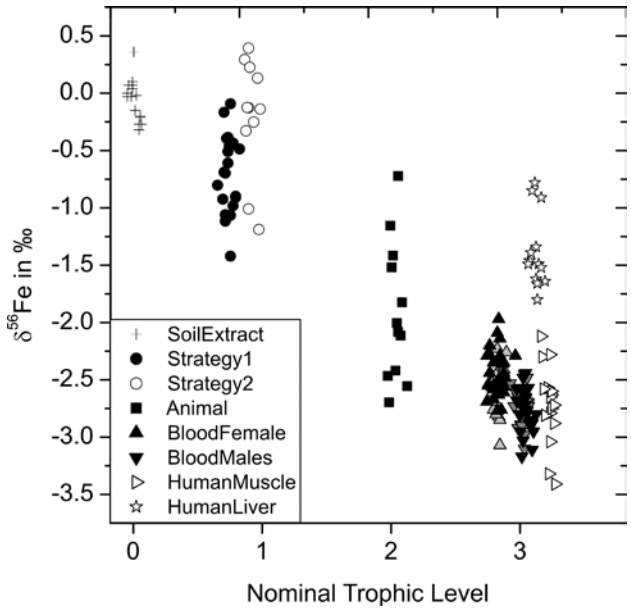

Figure 4. Iron isotope fractionation along the human food chain. Soil data represents the weakly-bound (HCl-leachable) iron fraction[12, 28-30]. All plant and animal data is from this study. Blood, muscle tissue, and liver data is from Walczyk & von Blanckenburg [1, 2] and blood data shown in grey symbols is from Albaréde et al. [4].

TABLES

Table 1. Sources, Fe concentrations, and Fe isotope composition of measured foodstuff

	Conc Fe[a]	$\delta^{56}Fe$[b]	2σ[c]
	$\mu g\ g^{-1}$	‰	‰
Strategy I plant products			
Arugula salad	201.0	-0.166	0.108
Broccoli	44.8	-1.059	0.059
Brussels Sprouts	38.0	-1.421	0.076
Field salad	107	-0.396	0.004
Field salad (organic)	345.7	-0.087	0.015
Flax Seed (organic)	35.4	-1.116	0.059
Iceberg lettuce	65.3	-0.908	0.059
Peas 1	35.7	-0.609	0.059
Peas 2	52.2	-0.859	0.113
Peas (organic)	92.1	-1.137	0.160
Potato	8.1	-0.802	0.082
Potato (organic)	10.2	-0.924	0.059
Spinach 1	127	-0.434	0.059
Spinach 2	85.6	-0.509	0.151
Spinach	92.8	-0.499	0.062
Spinach (organic)	83	-0.355	0.085
Zuccini	45.4	-0.697	0.059
Zuccini (organic)	47.1	-0.982	0.082
Strategy II plant products			
Oats (organic)	31.7	0.133	0.059
Oatmeal 1	55.9	0.294	0.059
Oatmeal 2	55	0.253	0.075
Oatmeal 3	56.4	0.393	0.059
Oat flakes (organic)	36.8	0.257	0.059
Wheat flour type 1050 1	21.3	-0.126	0.059
Wheat flour type 1050 2	21.9	-0.251	0.059
Wheatwhole flour	40.2	-0.138	0.102
Wheatwhole flour	30.1	-0.124	0.092
Wheatwhole flour (organic)	23.5	-0.328	0.059
White Rice	5.1	-1.189	0.059
Brown Rice	10.1	-0.942	0.190
Mushrooms			
Brown Mushrooms	19.6	-0.076	0.112
White Mushrooms (organic)	15.5	-0.431	0.195
Animal products			

Beef	80.4	-1.975	0.059
Egg Yolk	147.8	-2.419	0.059
Egg Yolk (organic)	113.5	-2.114	0.096
Egg White	0.8	-1.519	0.059
Chicken breast	10.2	-2.555	0.059
Chicken breast (organic)	8.6	-2.696	0.097
Boiled ham	21.5	-2.083	0.059
Boiled ham (organic)	17.4	-2.005	0.059
Pork Liver	623.5	-1.155	0.059
Soft Chease	1.8	-1.416	0.059
Minced meat sausage	13.8	-1.824	0.059
Tuna	37.7	-0.722	0.140
Salted Herring	17.5	-2.466	0.059
Processed Food			
Multi-grain bread	25.1	-0.294	0.059
Wheat bread (organic)	27.4	-0.134	0.059
Rye bread (organic)	21.7	-0.410	0.059
Wheat roll	10.7	-0.246	0.059
Crispbread	28.8	-0.433	0.059
Egg noodle	14.7	-0.481	0.059
Egg noodle (organic)	15.7	-0.561	0.059
Drinks and sweets			
Coffee	23.1	-0.761	0.059
Cocoa	316.6	0.091	0.100
Green tea	417.4	0.127	0.059
Assam Tea (organic)	100.1	0.123	0.071
English Breakfast Tea (organic)	123.6	0.133	0.074
Black Tea (organic)	78.4	0.130	0.073
Darjeeling Tea (organic)	111.8	0.351	0.162
Roibos Tea	73.3	-0.165	0.059

a Concentrations on a dry weight basis

b Average of replicate dissolutions were available

c 2σ error of mass-spectrometric measurement, of individual replicate dissolutions, or that calculated from 64 pooled replicate dissolutions (2σ δ^{56}Fe = 0.059 ‰), whichever is larger

The full dataset is available in supplementary material

Table 2. Average Fe isotope composition and their standard deviation of the main dietary Fe sources

	δ^{56}Fe	σ	Diet females[a]	Diet males[a]
	‰	‰	% Fe	% Fe
Bread	-0.30	0.12	18	19
Vegetable	-0.85	0.35	12	11
Meat	-1.93	0.82	8	10
Leafy vegetable	-0.42	0.25	6	6
Coffee	-0.77	--	5.5	5.5
Cold cuts	-1.97	0.13	5	4
Sweets	0.09	--	4	4
Corn, Cereal	-0.21	0.48	4	4

a According to Reference [15].

Online Supplement to "The iron stable isotope fingerprint of the human diet" by von Blanckenburg, Noordmann, Guelke-Stelling
Food sources, Fe concentrations and measured Fe isotope ratios of replicate samples

Foodstuffs	Supermarket	Producer	Date of Purchase	Conc Fe[a] [µg g^{-1}]	Std Dev[b] [µg g^{-1}]	No of [c] measur.	$\delta^{56}Fe$[d] [‰]	2s[e] [‰]	$\delta^{57}Fe$ [‰]	2s[e] [‰]	No of [f] measur.
Strategy I plant products											
Arugula salad	Kaufland	Frischeabteilung	01.09.2007	201.0		1	-0.166	0.108	-0.223	0.221	2
Broccoli	Kaufland	Holland (Klasse 1)	23.08.2007	44.8	0.6	2	-1.065	0.059	-1.600	0.107	3
dto.							-1.052	0.059	-1.547	0.107	3
Brussels Sprouts	Kaufland	Elbtal Tiefkühlkost	23.08.2007	38.0		1	-1.421	0.076	-2.110	0.107	3
Field salad	Kaufland	Frischeabteilung	20.07.2007	107	12	3	-0.394	0.059	-0.562	0.107	2
dto.							-0.398	0.068	-0.587	0.107	2
dto.							-0.395	0.059	-0.602	0.107	2
Field salad (organic)	denn's bio	Ökologischer Anbau	26.09.2007	345.7	1.2	2	-0.092	0.059	-0.114	0.107	2
dto.							-0.090	0.059	-0.140	0.107	2
dto.							-0.078	0.060	-0.127	0.107	2
Flax Seed (organic)	denn's bio	rapunzel	20.07.2007	35.4		1	-1.116	0.059	-1.671	0.107	1
Iceberg lettuce	Kaufland	Frischeabteilung	20.07.2007	65.3		1	-0.908	0.059	-1.330	0.107	3
Peas 1	Kaufland	Frenzel Tiefkühlkost	23.08.2007	35.7	6.4	2	-0.609	0.059	-0.910	0.107	2
dto.							-0.689	0.061	-1.022	0.107	2
Peas 2	Kaufland	Iglo (Unilever Deutschland)	23.10.2007	52.2	3.0	2	-0.899	0.093	-1.323	0.158	2
dto.							-0.819	0.059	-1.222	0.107	2
Peas (organic)	denn's bio	Denree	26.09.2007	92.1	6.4	3	-1.060	0.059	-1.566	0.107	3
dto.							-1.130	0.076	-1.664	0.107	2
dto.							-1.220	0.059	-1.814	0.107	2

Potato	Kaufland	Frischeabteilung	11.08.2007	8.1		1	-0.802	0.082	-1.186	0.124	3
Potato (organic)	Froh Naturkost	Ökologischer Anbau	26.09.2007	10.2		1	-0.924	0.059	-1.378	0.107	3
Spinach 1	Kaufland	Ardo	23.08.2007	127	24	2	-0.434	0.059	-0.648	0.107	3
dto.							-0.463	0.059	-0.682	0.107	2
Spinach 2	Kaufland	Tiefkühlkost	23.10.2007	85.6		1	-0.509	0.151	-0.728	0.107	2
Spinach	Kaufland	Iglo (Unilever Deutschland)	23.10.2007	92.8	8.2	3	-0.486	0.059	-0.740	0.144	2
dto.							-0.477	0.064	-0.715	0.130	2
dto.							-0.535	0.091	-0.812	0.107	2
Spinach (organic)	denn's bio	Denree	26.09.2007	83	19	2	-0.385	0.070	-0.586	0.107	2
dto.							-0.325	0.059	-0.494	0.107	2
Zuccini	Kaufland	Frischeabteilung	01.09.2007	45.4	4.4	3	-0.697	0.059	-0.984	0.146	2
Zuccini (organic)	denn's bio	Ökologischer Landbau	26.09.2007	47.1		1	-0.982	0.082	-1.418	0.124	2

Strategy II plant products

Oats (organic)	denn's bio	Bioland	26.09.2007	31.7		1	0.133	0.059	0.215	0.107	1
Oatmeal 1	Kaufland	Brüggen	23.10.2007	55.9		1	0.294	0.059	0.428	0.107	2
Oatmeal 2	Kaufland	Kclassic	11.08.2007	55	14	2	0.226	0.087	0.325	0.160	2
dto.							0.279	0.059	0.417	0.107	2
Oatmeal 3	Kaufland	Kölln's	23.10.2007	56.4		1	0.393	0.059	0.554	0.107	2
Oat flakes (organic)	denn's bio	Spielberger demeter	26.09.2007	36.8	5.2	3	0.255	0.059	0.388	0.107	1
dto.							0.259	0.059	0.428	0.107	1
Wheat flour type 1050 1	Kaufland	Weltgold	11.08.2007	21.3		1	-0.126	0.059	-0.177	0.107	2
Wheat flour type 1050 2	Kaufland	Aurora	23.10.2007	21.9		1	-0.251	0.059	-0.329	0.107	2
Wheatwhole flour	Kaufland	Küchenmeister	23.10.2007	40.2		1	-0.138	0.102	-0.207	0.107	2
Wheatwhole flour	Kaufland	Aurora	23.10.2007	30.1		1	-0.124	0.092	-0.167	0.207	2
Wheatwhole flour (organic)	denn's bio	Bohlsener Mühle	26.09.2007	23.5		1	-0.328	0.059	-0.486	0.107	3

White Rice	Kaufland	Kclassic	11.08.2007	5.1		1	-1.189	0.059	-1.730	0.107	1
Brown Rice	Kaufland	Curtiriso	11.08.2007	10.1	2.0	2	-1.009	0.059	-1.460	0.107	1
dto.							-0.875	0.059	-1.284	0.107	1

Mushrooms

Brown Mushrooms	Kaufland	Frischeabteilung	23.08.2007	19.6		1	-0.076	0.112	-0.092	0.264	2
White Mushrooms (organic)	denn's bio	Ökologischer Anbau	26.09.2007	15.5	4.2	2	-0.500	0.059	-0.706	0.107	2
dto.							-0.362	0.082	-0.528	0.107	2

Animal products

Beef	Kaufland	Purland	11.08.2007	80.4		1	-1.975	0.059	-2.893	0.107	3
Egg Yolk	Kaufland	KG Nordmark-Landei GmbH & Co	11.08.2007	147.8		1	-2.419	0.059	-3.558	0.107	2
Egg Yolk (organic)	Froh Naturkost	Bioland	26.09.2007	113.5		1	-2.114	0.096	-3.122	0.131	2
Egg White	Kaufland	KG Nordmark-Landei GmbH & Co	11.08.2007	0.8	0.6	3	-1.519	0.059	-2.261	0.107	1
Chicken breast	Kaufland	Original Wiesenhof	11.08.2007	10.2		1	-2.555	0.059	-3.778	0.107	1
Chicken breast (organic)	denn's bio	Freiland Puten Fahrenzhausen GmbH	26.09.2007	8.6		1	-2.696	0.097	-3.963	0.135	2
Boiled ham	Kaufland	Frischwaren: Wursttheke	11.08.2007	21.5		1	-2.083	0.059	-3.082	0.107	1
Boiled ham (organic)	denn's bio	Ökoland	26.09.2007	17.4		1	-2.005	0.059	-2.956	0.153	2
Pork Liver	Kaufland	Purland	11.08.2007	623.5		1	-1.155	0.059	-1.743	0.107	2
Soft Chease	Kaufland	NORDGut	23.08.2007	1.8	0.0	2	-1.416	0.059	-2.176	0.107	1
Minced meat sausage	Kaufland	Frischwaren: Wursttheke	11.08.2007	13.8		1	-1.824	0.059	-2.707	0.107	2
Tuna	Kaufland	Kclassic	11.08.2007	37.7		1	-0.722	0.140	-1.041	0.176	2
Salted Herring	Kaufland	Rügen Fisch	11.08.2007	17.5	2.8	2	-2.466	0.059	-3.649	0.107	1

Processed Food

Multi-grain bread	Kaufland	Brotland GmbH	11.08.2007	25.1		1	-0.294	0.059	-0.490	0.107	2
Wheat bread (organic)	denn's bio	Bioland	26.09.2007	27.4		1	-0.134	0.059	-0.217	0.107	3
Rye bread (organic)	Froh Naturkost	Ökologischer Anbau	26.09.2007	21.7		1	-0.410	0.059	-0.650	0.107	1
Wheat roll	Kaufland	Bäcker im Kaufland	02.10.2007	10.7		1	-0.246	0.059	-0.375	0.107	2
Crispbread	Kaufland	Barilla Wasa Deutschland GmbH	11.08.2007	28.8		1	-0.433	0.059	-0.672	0.107	2
Egg noodle	Kaufland	Kclassic	01.09.2007	14.7		1	-0.481	0.059	-0.710	0.107	1
Egg noodle (organic)	denn's bio	Spielberger demeter	26.09.2007	15.7		1	-0.561	0.059	-0.815	0.107	1

Drinks and sweets

Coffee	Kaufland	Jacobs Krönung (Grüne Packung)	11.08.2007	23.1	0.4	2	-0.773	0.059	-1.150	0.107	1
dto.							-0.749	0.059	-1.154	0.107	1
Cocoa	Kaufland	cebe	23.08.2007	316.6		1	0.091	0.100	0.151	0.118	2
Green tea	Kaufland	Euro-tea	20.07.2007	417.4		1	0.127	0.059	0.146	0.107	3
Assam Tea (organic)	denn's bio	Ökologischer Landbau	02.10.2007	100.1		1	0.123	0.071	0.182	0.107	2
English Breakfast Tea (organic)	denn's bio	Ökologischer Landbau	02.10.2007	123.6		1	0.133	0.074	0.230	0.107	3
Black Tea (organic)	denn's bio	Ökologischer Landbau	02.10.2007	78.4		1	0.130	0.073	0.182	0.153	2
Darjeeling Tea (organic)	denn's bio	Ökologischer Landbau (Demeter)	02.10.2007	111.8	3.6	2	0.380	0.059	0.577	0.107	2
dto.							0.414	0.059	0.600	0.107	2
dto.							0.260	0.059	0.403	0.107	2
Roibos Tea	Kaufland	Kclassic	20.07.2007	73.3		1	-0.165	0.059	-0.232	0.107	2

[a] Concentrations on a dry weight basis

[b] 2 standard deviations of replicate concentration measurements where available

[c] Number of replicate concentration measurements

[d] Replicate isotope measurements report separate dissolutions, separations, and measurements

[e] 2s error of mass-spectrometric measurement or that calculated from 64 replicate dissolutions (2s $d^{56}Fe$ = 0.59‰, 2s $d^{57}Fe$= 0.107‰), whichever is larger

[f] Number of replicate dissolutions and mass-spectrometric measurements