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ORIGINAL PAPER

Tracing the geographic origin of poultry meat and dried beef with oxygen and strontium isotope ratios

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Abstract Two promising isotope ratios, the $\delta^{18}O$ of the water fraction, as extracted by azeotropic distillation, and the $^{87}Sr/^{86}Sr$ of the ash fraction were tested for their applicability to determine the geographic origin. In two sampling phases, in total 78 fresh poultry breast meat and 72 dried beef samples, independent from each other and originating from different countries, were analysed. The $\delta^{18}O$ was measured with isotope ratio mass spectrometry and the isotope abundance ratio of $^{87}Sr/^{86}Sr$ with a multicollector inductively coupled plasma mass spectrometer. With $\delta^{18}O$ it was possible to distinguish ($p < 0.001$) poultry and dried beef samples according to their country of origin. The beef data suggests that the procedure of processing is of only low additional influence on $\delta^{18}O$ and, if so, it seems to reduce the initial between-country differences. The $^{87}Sr/^{86}Sr$ ratio did not give sufficient indications for differentiation by geographic origin in either poultry meat or dried beef in the smaller, first phase, data set and was therefore not further tested.

Keywords Stable isotope · Authentication · Traceability · Cattle · Broiler · Meat

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Introduction

Currently the demand of the public to get informed about the geographic origin of food, in particular of meat, is increasing. In order to move beyond simple paper traceability, additional tools for a more fraud resistant control of authenticity statements have to be developed. The abundance ratio of different stable isotopes has already been used to determine the authenticity and trace the origin of different raw and processed foods [1–3]. Several official methods based on oxygen (O) isotope ratios are currently in use for the determination of the authenticity of wine [4], maple syrup [5], and fruit juices [6]. Also for cheese [7–10] and butter [11] the O isotopic ratio has been shown to be a reliable tool to determine the geographic origin. Another promising element in this context is also strontium (Sr) which has already been used to determine the geographic origin of foods including meat [3,

12], cheese [9], butter [9, 11], and wine [13–15]. The basic principle for a potential usefulness of isotope ratios is that certain regions are characterised by specific 'fingerprints' of isotope ratios allowing conclusions to be drawn about origin [16].

Concerning meat, several studies so far have been focusing on the distribution of stable isotopes to analytically trace its geographic origin. Various components (muscle, fat, protein fraction, body water, specific metabolites) and elements (^{13}C, ^{15}N, ^{18}O, ^{34}S, ^{87}Sr) were shown to be amenable to isotope measurements [17–25]. The elements, the focus was put on, were those directly or indirectly related to the feed and water offered to the animals. In the case of strontium, the isotope ratio is directly related to the underlying geology [26–28]. The $\delta^{18}O$ of drinking water and of the water incorporated in feed could reflect a region-specific distribution. Several studies gave indications for the value of oxygen isotope ratio to differentiate beef with respect to geographic origin as long as the regions are large enough or far enough apart [20, 21, 25, 29]. However, in contrast to raw beef, for dried beef and poultry meat to our knowledge no such studies exist. Drying is likely to cause a shift of $\delta^{18}O$ [19, 25, 30] and in case this shift differs among processing plants, the authentication of origins, known to work with raw beef, might either be hampered or facilitated in the processed beef. In the case of poultry meat, the dependence on regional feeds is much less pronounced than for mostly forage-fed cattle, which makes the use of analogies to raw beef results difficult. Similarly, the usefulness of the Sr isotope abundance ratio for meat authentication still has to be proven as no such attempts have been made, maybe because the Sr content in this commodity is typically low [31].

The aim of this study was to evaluate to what extent $\delta^{18}O$ and $^{87}Sr/^{86}Sr$ are useful for the differentiation between meat from different countries of origin. As commodities, poultry breast meat, as an important globally marketed form of meat, and dried beef, a well-known Swiss specialty, were chosen. The latter was produced out of raw meat originating from different countries and processed in part in countries other than those where the raw meat originated from.

Materials and methods

Meat samples

A total of 78 poultry breasts were obtained from Brazil, France, Germany, Hungary, and Switzerland between February 2004 and December 2005. This basically included two sample sets, the first comprising 22 samples from these countries (Phase I), the second 56 samples (Phase II). The authenticity of all samples had been certified with valid custom documents, specifying place and date of slaughter. One breast fillet of each sample (for Phase I, four independent fillets were pooled), deep-frozen at –25 °C, was homogenised (Büchi Mixer B 400, Büchi AG, Flawil, Switzerland), divided in sub-samples of 50 g each, vacuum-sealed and deep-frozen again for analysis.

Totally 72 dried beef meat samples (thereof 21 being collected in Phase I), prepared either from *M. biceps femoris* or from *M. semitendinosus*, were either directly collected from the production places (samples produced in Switzerland) or purchased from producers in Australia (n = 8), Austria (n = 5), Canada (n = 8), and USA (n = 5) between May 2004 and February 2006. The Austrian samples were produced from Brazilian raw meat, for the other non-Swiss samples raw meat originated from the country of processing. Swiss samples were partly produced in the Swiss canton of Valais using Swiss raw meat (n = 14) and partly in the canton of Grisons using Swiss (n = 16) and Brazilian raw meat (n = 16). All dried beef samples were produced by curing and various sequences of drying and pressing. Slight variations in recipes (e.g. amount of salt, kind of herbs) and technology (salt application, curing, drying, pressing, use of moulds, storing, packaging, etc.) may occur within the same type of product, depending on the producer. One slice (ca. 50 g) for each isotope ratio, taken from the centre of the dried beef meat piece of a total weight of approximately 1 kg, was homogenised, vacuum-sealed and stored in a cooling room (2.5 °C) until being analysed.

All individual poultry and dried beef samples were independent from each other by being either obtained from different producers or originating from different production batches when obtained from the same producer.

Strontium isotope analysis

Samples of 20 g (poultry meat) and 5 g (beef), respectively, were weighed into quartz crucibles and ashed in a muffle furnace by stepwise heating to 650 °C. A volume of 2 mL sub-boiled nitric acid (690 g kg^{-1}) was then added to the residue. After 4 h of extraction, 2 mL of water (>18 MΩ, 0.2 μm) were added and the suspension was filtered (membrane filter PTFE 0.45 μm, syringe and filter rinsed previously). The concentration of Sr and the separation of Rb from the filtrate were carried out using a Sr-selective column material (SrResin, 100–150 μ, Eichrom, Darien, IL, USA) according to Horwitz

[32]. The column with a diameter of 3 mm contained 100 mg SrResin, which was cleaned with 10 mL of water (>18 MΩ, 0.2 µm) and 10 mL of nitric acid (504 g kg^{-1}). The filtered sample solution was put through the separating column in small portions. Rubidium (Rb) and other cations (Na, K, Ca, etc.) were then eluted with 10 mL of nitric acid (504 g kg^{-1}). Strontium was desorbed with 1 mL of water (>18 MΩ, 0.2 µm) and collected in a previously cleaned polypropylene vial according to Fortunato et al. [9]. The separation of Rb from the filtrate was then repeated. All vessels had been cleaned by nitric acid (100 g kg^{-1}) for at least 24 h before use and were rinsed with water (>18 MΩ, 0.2 µm). To analyse the samples multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS) (VG Axiom, Thermo-Elemental, Winsford, Great Britain) was used. The instrumental parameters were adjusted according to Fortunato et al. [9], Deniel and Pin [33] as well as Waight et al. [34] (Table 1). Sample port and ICP-MS were optimised for a high sensitivity before each measurement.

Oxygen isotope analysis

Water for oxygen isotope analysis was obtained by azeotropic distillation with toluene (principle described by Matissek et al. [35]). The Bidwell-Sterling apparatus, used in the present study, was built as outlined by Schäfer [36]. Briefly, approx. 10 g of the test samples were weighed into a round-bottom flask, 100 mL of toluene were added, and the flask was boiled afterwards in backflow mode for about 18 h in an oil bath at 130 °C. Although more than 96% of the water theoretically extractable from the samples with the azeotropic distillation can be obtained after only 4 h, we wanted to harvest the maximum possible amount of water and therefore used this extended distillation time. A high water extraction yield is necessary to limit the risk of isotope fractionation processes [30]. Water and toluene were collected and the two phases were separated in a separatory funnel. Based on preliminary tests, the pH of the water extract was reduced by adding 500 mg anhydrous ammonium chloride (NH_4Cl, purity 99%, cat. no. 101141; Merck, Darmstadt, Germany) in order to guarantee sufficient signal intensities during the subsequent analysis of the oxygen isotopes. Afterwards 0.5 mL of the extracted water was pipetted into a 10 mL-vacutainer (BD Vacutainer Systems, Plymouth UK) and equilibrated over night with carbon dioxide (4 mL L^{-1} helium gas mixture). These vacutainers had been previously washed for 10 min in distilled water, rinsed afterwards with acetone, and finally dried in an oven at 50 °C for at least 4 h before use. The determination of the oxygen isotope ratio was carried out with isotope ratio mass spectrometry (IRMS) (Delta-Plus XL, Finnigan, Bremen, D) as outlined by Klimmek et al. [37] and Gremaud et al. [38]. Standard deviations of repeated measurements ($n = 10$) were found to be below $\delta^{18}O = 0.3‰$. Each sample was analysed in duplicate.

The performance of the entire measurement process from sample weighing, extraction, and equilibration to isotopic measurement was validated for its quality by various indicators.

Repeatability

Under repeatability conditions, seven aliquots of the same sample were subjected to the entire analytical procedure including weighing, azeotropic distillation, and equilibration of water with carbon dioxide and isotope measurement. The repeatability standard deviation (SDr) across the entire whole analysis was 0.26‰ ($r = 0.7$).

Robustness

Preliminary experiments have shown that the water yields extracted from meat increased with extraction times from 1 to 4–6 h and stay stable for extraction times >6 h. For practical reason, we decided to extract our samples for 18 h. To assess the impact of varying distillation times, one sample was extracted for periods increasing from 6 to 48 h. No statistically significant

Table 1 Important instrumental parameters for VG Axiom MC-ICP-MS

Rf-power	1250 W		
Plasma gas flow (argon)	14.0 L min^{-1}		
Auxiliary gas flow (argon)	0.58 L min^{-1}		
Nebuliser gas flow (argon)	1.0 mL min^{-1}		
Sampling cone material	Platinum		
Mass resolving power (10% valley) m/Δm	410		
Dwell time	50 ms		
Points per replicate	40		
Mass assignment to Faraday cup detectors	Kr	83	L4
	Sr/Kr	84	L2
	Rb	85	L1
	Sr/Kr	86	Ax
	Sr/Rb	87	H1
	Sr	88	H2
Rosol drying unit	CETAC Aridus		
Nebuliser type	TH 1 concentric		
Typical liquid sample uptake rate	100 µL min^{-1}		
Air temperature in clean hood for instrument	18 ± 1 °C		

changes (regression, test on slope) of the δ-values with increasing extraction times were observed. The impact of a varying sample mass (5, 10 and 15 g) was examined by performing 3×3 independent analyses. Standard deviations and averages were compared using one-way ANOVA, and no significant influence of the sample mass on the δ-values was found.

Efficiency of distillation

The efficiency of the extraction procedure for the $\delta^{18}O$ analysis was evaluated based on residual water in the extracted samples determined by the Karl-Fischer method [35]. This residual water of the sample was found to be less than 1% of the fresh weight. The coefficient of variation of the extracted water mass from repeated extractions ($n = 7$) of the same sample was below 2.5%.

Selectivity of the extraction step

The selectivity of the extraction for the $\delta^{18}O$ analysis was evaluated by NMR, and no toluene was found in the extracted water.

Statistical analysis and calculations

From the oxygen isotopes measured, the δ-values (‰) were calculated as

$$\delta = \left(\left(\frac{R_{\text{sample}}}{R_{\text{std}}} \right) - 1 \right) \times 1,000$$

where δ = relative isotope ratio, $R_{\text{sample}} = {}^{18}O/{}^{16}O$ of the sample and $R_{\text{std}} = {}^{18}O/{}^{16}O$ of the standard (Vienna standard mean ocean water). The strontium isotope ratio was given as $^{87}Sr/^{86}Sr$ as is common.

The significance of origin was tested using ANOVA. Subsequently, Bonferroni adjusted pairwise comparisons were performed in order to determine the significance of the differences between origins. All statistical analyses were performed using Systat (version 11, Systat Software Inc., Richmond, California, USA). At first, Phase I poultry samples (five countries of origin; 2–7 samples per origin) and Phase I beef samples (seven countries of origin; 2–4 samples per origin) were statistically evaluated for differences in origin for both oxygen and strontium isotopes. For $\delta^{18}O$, the isotope ratio with the much higher discriminative power, ANOVA, combined with pairwise comparison, was carried out again for both commodities after including also the Phase II data.

Results

Strontium isotopes

In poultry, for the first set of samples weakly significant country differences ($p = 0.04$) were found in $^{87}Sr/^{86}Sr$ of poultry meat, but the pairwise comparison among means with the conservative Bonferoni adjustment did not specify any individual differentiation. The average values of $^{87}Sr/^{86}Sr$ were numerically lowest for poultry meat samples from Germany and highest for samples from Hungary (Table 2). Comparing dried beef of different origin (Table 3), the $^{87}Sr/^{86}Sr$ did not reveal any significant differences among origins of the beef ($p = 0.12$). Additionally, the values of the isotopic ratio covered only a very narrow range.

Oxygen isotopes

In poultry meat, the first, smaller, set of samples already showed a high potential to distinguish between origins (origin effect: $p < 0.001$, associated with a clear group separation by Bonferoni adjusted pairwise comparison). Therefore the oxygen isotopes were determined on the second sample set to confirm this result. For the combination of both sample sets, the effect of country of origin in $\delta^{18}O$ of the water incorporated in poultry meat was again highly significant ($p < 0.001$). The order of the countries stayed the same as that found with Phase I samples alone. Meat samples from Switzerland and Hungary showed the lowest (most negative) $\delta^{18}O$-values (Table 4), whereas French poultry meat was highest in $\delta^{18}O$. Bonferoni adjusted pairwise comparison distinguished between three sample groups: (1) Switzerland and Hungary, (2) Germany, (3) Brazil and France.

For beef, like in the poultry meat, analysis of variance of the first sample set showed clear differences ($p < 0.001$), and analysis was re-done with the total sample set. This also yielded highly significant differences ($p < 0.001$) in $\delta^{18}O$. With one exception (beef manufactured in Austria compared to that

Table 2 $^{87}Sr/^{86}Sr$ measured in poultry breast meat samples from different countries of origin

Country	n[a]	$^{87}Sr/^{86}Sr$
Germany	3	0.708 ± 0.002
Brazil	4	0.709 ± 0.001
Switzerland	7	0.709 ± 0.001
France	2	0.711 ± 0.001
Hungary	6	0.711 ± 0.002

[a] Number of samples

Table 3 $^{87}Sr/^{86}Sr$ measured in dried beef samples from different countries of raw meat origin and places of processing

Country of origin of the raw beef	Country of beef processing	n^a	$^{87}Sr/^{86}Sr$
Brazil	Austria	2	0.709 ± 0.000
USA	USA	2	0.709 ± 0.000
Canada	Canada	2	0.709 ± 0.001
Australia	Australia	4	0.710 ± 0.001
Switzerland	Switzerland[b]	4	0.710 ± 0.001
Switzerland	Switzerland[c]	3	0.710 ± 0.001
Brazil	Switzerland[b]	4	0.712 ± 0.002

[a] Number of samples
[b] Canton of Grissons
[c] Canton of Valais

Table 4 $\delta^{18}O$ measured in water derived from poultry breast meat samples from different countries of origin

Country	n^a	$\delta^{18}O$	
France	13	–1.79 ± 1.17	C
Brazil	14	–2.56 ± 0.67	C
Germany	15	–4.05 ± 1.22	B
Hungary	16	–5.39 ± 0.87	A
Switzerland	20	–5.69 ± 0.81	A

Countries without a common letter are significantly different ($p < 0.05$)
[a] Number of samples

produced in Switzerland), the order of the groups in terms of $\delta^{18}O$ remained the same as when analysing only the Phase I samples. The results of the pairwise comparison gave four groups: (1) samples from Australia, (2) samples processed in Switzerland and Austria, (3) US samples, samples of Swiss raw meat origin and samples processed in Austria, and (4) samples from the US and Canada (Table 5). Canadian dried beef showed the lowest $\delta^{18}O$, the Australian samples the highest values.

Discussion

Strontium isotopes

The $^{87}Sr/^{86}Sr$ has been described to be an indicator for both geochemical origin and age of the geological material [26]. Through determination of this Sr isotope ratio, a link to the type of geological bedrock, characteristic for geographic regions, and the animals kept on this land might be possible. As stated by Capo et al. [26], bones and teeth of terrestrial herbivores reflect the Sr isotope ratio of their feed that was similar to bedrock values in many cases. This explains the successful application of the Sr isotope ratio in various foods (see "Introduction")

Table 5 $\delta^{18}O$ measured in water derived from dried beef samples

Country of origin of the raw beef	Country of beef processing	n^a	$\delta^{18}O$	
Australia	Australia	8	1.84 ± 1.24	D
Brazil	Switzerland[b]	16	–1.03 ± 1.43	C
Switzerland	Switzerland[c]	14	–1.64 ± 1.31	BC
Switzerland	Switzerland[b]	16	–2.03 ± 1.09	BC
Brazil	Austria	5	–2.40 ± 1.20	BC
USA	USA	5	–3.35 ± 0.55	AB
Canada	Canada	8	–5.59 ± 2.06	A

Countries without a common letter are significantly different ($p < 0.05$)
[a] Number of samples
[b] Canton of Grissons
[c] Canton of Valais

In poultry meat, no clear discrimination between individual countries of origin was possible using $^{87}Sr/^{86}Sr$. This might have had several reasons. The housing conditions of commercially fattened poultry are such that relatively little contact with the environment takes place, especially when they are kept in house with no access to free range. Therefore, the extent to which the body of the birds adjusts to the local Sr isotope ratio is reduced, especially since the feed components used in poultry diets are mostly traded globally. Contaminations with Sr of mineral supplements, which are typically imported into the farms and thus are likely to have a different isotope ratio than home-grown feed and drinking water, also cannot be excluded. Additionally, in poultry the fattening period typically lasts just for some weeks, giving the animals not much time to fully equilibrate with the isotopic ratio of diet or environment.

Beef cattle are normally in closer contact to their environment than commercially fattened poultry. This explains why the likelihood that specific Sr isotope features of the environment reflected by the meat should be higher for cattle than for poultry. In spite of this, in the dried beef samples the discriminative power of the Sr isotopes for countries of origin was even lower than for poultry. Additionally, changes in $^{87}Sr/^{86}Sr$ might occur in the transformation process (e.g. by curing or seasoning), which could mask country differences being present in the raw beef.

The general difficulty to differentiate between countries noted in both commodities might have two major reasons. Firstly, the generally low Sr concentration of meat and its small variation among samples does not facilitate differentiation. Additionally, since

the objective of the present study was to confirm analytically the declaration of the country of origin of raw materials as required by law (Swiss Ordinance on Labeling and Advertisement of Food, LKV Art. [16]), political borders set the boundaries for the sample grouping applied in the present study. These borders did not explicitly follow geological borders. Especially in large countries like Brazil, Canada, USA, France, and Germany the $^{87}Sr/^{86}Sr$ might be characteristic for individual regions and the corresponding soils, where the animals had been reared on, rather than for the entire country.

Oxygen isotopes

The use of $\delta^{18}O$ in the meat's water fraction as an indicator of the geographic origin of beef is based on the experimentally verified evidence that there is a direct relationship between the isotope composition of ingested water and that of the water in the body water pool [18, 20, 21, 39]. In the present investigation, there was a significant between-country variation in $\delta^{18}O$ in both the commodities, poultry meat and dried beef. The reasons for this variability could be manifold. The $\delta^{18}O$ values in the body water pool of living animals are determined by various sources of water [40] including drinking water (from local tap water and local precipitation water) and, to a lesser extent, water in moist feeds [41] (mainly fed to ruminants and not to poultry) as well as the metabolism of the animal. There are distinct geographic differences in the $\delta^{18}O$ of the local water, noticeable in a decrease of the heavier oxygen isotope, ^{18}O, with increasing distance from the sea (continental effect), with increasing distance from the equator and with increasing elevation (effect of altitude) [19, 42–44].

Drinking water supply is almost invariably restricted to regional sources. From German data, it was calculated that the natural isotope gradient of $\delta^{18}O$ in water was about –2.4‰ (domestic use) [41], about –3‰ (groundwater) [42] per 1,000 km distance from the sea as well as between –0.28‰ [41] and –0.44‰ [42] per 100 m of elevation. From Swiss data, a decrease of $\delta^{18}O$ from –0.15 to –0.3‰ per 100 m altitude was determined [44, 45]. The oxygen isotope ratio of meat water used in a study for the authentication of the geographic origin showed that, within the same region (e.g. 'Schleswig-Holstein', ca. 3,000 km^2), the distribution of $\delta^{18}O$ covered a maximal range of up to 4‰ while, on a global level, the total range accounts for up to 12‰ [25]. Therefore, a differentiation of very different geographical origins, such as Germany and Argentina, was still feasible while the separation of more closely related origins, e.g. the region of 'Schweinfurt' ($\delta^{18}O$ = –6.3 to –2.5‰) and the region of 'Kulmbach' ($\delta^{18}O$ = –4.7 to –1.9‰) was not possible [25]. This was confirmed by the study of Lüpke et al. [41], where the isotope ratios of beef samples coming from neighbouring regions defined as circles of 150–200 km diameter were compared, meaning that samples coming from areas close to the border of neighbouring countries were not likely to be differentiated. The $\delta^{18}O$ difference between slaughterhouses located in the same region reached 0.8–1.0‰, whereas the $\delta^{18}O$ differences between region averages were between 0.3 and 1.2‰ [41].

Apart from the drinking water-related dependence, there might be a selective enrichment in meat depending on feed type. Renou et al. [29] found that the ^{18}O enrichment of the water fraction of beef meat samples from various regions in France was different when either maize silage or grass was fed, while it was not significantly related to the geographic origin. If this would really be the overriding effect, this would mean a serious limitation of the geographic traceability, since forage type may change and may actively be changed once such control practices become widely known. The type of feed, given to the animals, is unknown in our case; however, it is known that poultry usually is offered dry feed and local tap or precipitation water. As a consequence the influence of the water incorporated in feed might be smaller for poultry than for beef.

One further relevant factor influencing $\delta^{18}O$ might be the evaporative water losses from the body of livestock (via lung and skin), where ^{16}O is lost at higher rates than the heavier ^{18}O, thus enriching the body water pool of terrestrial animals with ^{18}O relative to that of the ingested water [18, 39, 46]. Moreover, some water is produced from feed nutrient oxidation (metabolic water), and this does influence the ^{18}O enrichments as well. The magnitude and the impact of these phenomena on the $\delta^{18}O$-values are hard to predict. As the body water fractionation relative to the ingested water increases with decreasing body size, this phenomenon is more relevant for poultry than for cattle [40].

Finally, factors independent of the animal may have an important influence as well. For example different carcass treatment and storage conditions after slaughter may also cause shifts in the oxygen isotope ratio [41], and sample storage unprotected against evaporation as well. These are 'man-made' factors, which on the one side can be prevented by defined careful handling, but on the other side could be used to change the isotope ratio of the meat on purpose.

Poultry meat

In the present study, the means of $\delta^{18}O$ were as expected from the global meteoric water isotope ratio for the different origins of poultry (except of the French samples). Accordingly, higher values were observed for countries closer to equator [19, 47] and oceans [41] (Brazil) than for Germany, Hungary and Switzerland. The mean $\delta^{18}O$-value of the German poultry samples of –4.1‰ was, as expected, higher than that of the German groundwater (δ = –6 to –12‰) [42] as a consequence of the process of body water fractionation outlined above. The $\delta^{18}O$-level of the fresh poultry breast was also similar to that observed in fresh German beef samples (δ = –3.8 ± 1‰, n = 175) [25]. Also the Hungarian (–5.4‰) and Swiss (–5.7‰) poultry samples showed higher $\delta^{18}O$ compared with the corresponding groundwater level (–8 to –11‰ in both countries [48]). The Brazilian samples (–2.6‰) were on the upper limit of the value of groundwater (–2 to –10‰ [48]). For unknown reasons, poultry samples from France showed high δ-values (–1.8‰), which were also not comparable to fresh beef samples from France (δ = –5.8 ± 1.8‰, n = 6) [29]. The poultry meat samples came from different slaughterhouses and French regions. Possible explanations might be found in specific feed composition, husbandry conditions or post-slaughter meat treatment. An effect can also be expected when the meat is not frozen immediately after slaughter and thus subject to higher water evaporation. Furthermore, changes might occur when summer precipitation water instead of tap water is supplied as drinking water, which shows different $\delta^{18}O$ values compared with ground or river water (and hence the resulting tap water) as the latter reflects the isotope distribution of the rain water some months ago [44]. However, since the French samples had been collected in different seasons over a period of 1.5 years, the latter source of variation should not have influenced the $\delta^{18}O$ of these samples.

Dried beef

In dried beef, the Australian could be distinguished statistically from all other samples. An important reason for this could have been that Australia has a warmer climate than the countries of the Northern hemisphere. Climate and especially temperature are known to influence $\delta^{18}O$ [29, 45, 49, 50]. Also a higher evaporation from feed or extended periods of grazing may increase the O isotopic ratio [25]. On the other end of the scale of the $\delta^{18}O$ value, the Canadian dried beef was significantly different from all other groups of samples, with the exception of the US samples. The samples with $\delta^{18}O$-values in between these two countries could be divided into two groups with Brazilian raw beef processed in canton of Grissons being different from US dried beef, with all other origins laying in between. Individual reasons for this differentiation between countries remain unexplored, and maybe a combination of factors was responsible.

Concerning the effects of site of processing, as opposed to the importance of the raw beef origin, the comparison within Swiss products and to the Austrian dried beef produced from Brazilian raw beef gives some indications. Due to the fact that ^{18}O preferentially remains and gets enriched during evaporation [19, 30, 44, 46], it was expected that the drying process would affect the final oxygen isotope ratio of the meat product. Unpublished preliminary data suggest that the δ-values increase with drying as do studies describing an elevated $\delta^{18}O$ with increasing storage time in a simulated storage experiment [46]. However, samples of Brazilian raw meat origin (processed in Austria and canton Grissons) could not be separated from dried beef made from Swiss raw meat (processed either in canton of Grissons or Valais). This is astonishing since other studies [20] showed that $\delta^{18}O$ of raw beef meat is different at least in samples from Argentina and Germany. This suggests that drying masked the likely initial differences in $\delta^{18}O$ between Brazilian and Swiss raw beef. This does not seem to be purely a specific effect of Swiss-type of processing as there was also no clear difference between the Austrian product and those Swiss products made from Swiss raw beef. An indication that processing as such does not create new differences but rather blurs existing differences is given by the observation that it was not possible to distinguish $\delta^{18}O$ between the two Swiss processing regions both using Swiss raw beef. For the samples used in the present study raw meat of different origins was separately cured. If this is not the case, initial differences in $\delta^{18}O$ present in the raw meat might be further reduced when curing of meat of different origin is done together in one container. The drawn liquid from the meat will contain a mixture of both isotopic ratios of the raw meat origins. Because the liquid will be reabsorbed, the original isotopic ratio of the raw meat might be changed towards the mean value thus equalising original differences.

Processing conditions apart from drying, which basically could influence the $\delta^{18}O$ of the meat, are stitch pumping or wet curing. For stitch pumping a curing solution is injected in the meat and for wet curing the whole raw meat is placed in a brine. For the preparation of the brine tap water from the area

of the processing (with the $\delta^{18}O$ of this region) is used. Nevertheless, the traditional way is still dry curing where the curing salt is rubbed into the meat manually or by tumbling. From the processing point of view stitch pumping is not useful, because the injected water increases the water content of the meat and so increases the drying time. Since stitch pumping is not forbidden in the specifications for dried beef produced in Switzerland [51, 52], and even no regulations are given about dried beef not produced in Switzerland, its application cannot be excluded, even if it is unlikely.

Conclusion

Especially encouraging results were obtained with this set of samples when poultry was coming from countries with known differences in groundwater isotope composition. This characteristic is difficult to adulterate thus representing a possible solid criterion that is quite resistant to fraud. For dried beef the differentiation was more difficult, but nevertheless possible, when geographic conditions were clearly different (Australia vs. Europe vs. North America), while a gradient with increasing distance from the ocean was not clearly apparent. However, it was possible to differentiate between certain origins using $\delta^{18}O$, even after the assumed changes in the isotopic ratio caused by drying. In this context it seems that the site of processing diminishes original country difference to some extent making authentication of country origin less efficient. The place of processing is difficult to state, as processing effects seem not to be that much different between sites in their effects on $\delta^{18}O$. Although demonstrated to be suitable for various foods, the Sr isotope ratio did not prove useful with the meat sample set investigated to trace its country of origin, which has not to be identical with the geological origin. The low variation in the Sr isotope abundance ratio found between, but also within, countries in both commodities therefore makes this indicator appear less promising than the one based on the oxygen isotopes. Future studies should test combinations of the $\delta^{18}O$ method with multi-element analysis, determined e.g. with ICP-HRMS [53], or with the abundance of distinct stable isotopes of other elements such as ^{13}C, ^{15}N and ^{34}S isotopes which are known to be helpful for the resolution of the various origins. The latter was demonstrated by Boner and Förstel [20] as well as Schmidt et al. [54], where a differentiation of beef between various geographic regions within and between countries was possible.

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