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Food chain

Iodine in the feed of cows and in the milk with a view to the consumer's iodine supply

Friedrich Schöne*, Kathrin Spörl, Matthias Leiterer

Thuringian State Institute of Agriculture, Investigation Unit – Laboratory, Naumburger Str. 98, D-07743, Jena, Germany

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ABSTRACT

Milk may be an iodine source for humans, but its magnitude depends on the iodine content of cow feed. The present investigation focused on the iodine level of feed and milk in German feeding practice in comparison with the results of previous milk monitoring and dose-response experiments with dairy cows. In 73 samples of straight feedstuffs (41 concentrates, 32 silages prepared from grass or green maize) and a total of 83 vitamin mineral premixes and compound feeds, respectively, the iodine content was determined and along with the expected daily iodine intake of cows. In six Thuringian cow herds, the total cow diets (51 total mixed rations, TMR) as well as the bulk milk (n = 77) were analysed for iodine. Cereal and legume grains and extracted meals from oilseeds had very low native iodine contents at <9–43 μg iodine/kg dry matter (DM). Silages showed higher contents than the concentrates. In grass silage, the median amounted to 173 μg iodine/kg DM. The significant relationship between the silages' ash and iodine content indicates contamination of grass during harvesting by soil. With regard to supplements, dairy cows received via premixes a mean of 1.2 mg iodine/kg diet DM and this was in the magnitude of 0.8 mg iodine/kg DM determined in TMR *on farm*. However, the resulting mean milk concentration of 105 μg iodine/kg, median 100 μg/kg, is only a half of that in dose-response experiments with 200 μg iodine/kg milk at 1 mg iodine/kg cow feed DM. The decrease of iodine transfer into the mammary gland and milk is caused by rapeseed meal (RSM) with the glucosinolates and their degradation compounds, e.g. isothiocyanates. By compensating for the iodine antagonist effects of diets containing RSM, more iodine should be added. Twice the current mean feed iodine supplement would optimize the contribution of animal-source food to the human iodine supply in Germany.

1. Introduction

Milk may be an iodine source for humans, but its magnitude depends on the iodine content of cow feed. In Germany, up to the middle of the 1980s feed was not regularly supplemented with iodine, resulting in milk that was almost free of iodine (<20 μg/kg) [1]. The native iodine content of plant straight feedstuffs is low. Insufficient iodine ingestion is associated with thyroid iodine depletion, which can cause thyroid hormone deficiency and goiter development, respectively. The longterm lack of feed iodine supplements in cow or sow diets causes hypothyroidism in mothers and offspring, with stillborn and immature calves and piglets [for these and further symptoms of iodine deficiency see [2–4]. An increasing usage of rapeseed meal (RSM) in livestock diets is associated with the intake of glucosinolates which act as iodine antagonists, thereby intensifying the negative effects of insufficient iodine feeding [5].

For the last two decades in the animal husbandry of the EU countries, iodine deficiency conditions have been overcome by the addition of iodine to feed as calcium iodate, sodium iodide and further iodine compounds [6,7]. The maximum level (ML) of 5 mg iodine/kg complete feed for dairy cows and laying hens and 10 mg iodine/kg for fattening pigs, beef and poultry (the complete feed is expressed as grain dry matter, DM, equivalents of 88% DM) exceeds the recommendations of animal nutrition societies [8,9] manifold: 0.15 mg iodine/kg feed DM in fattening pigs to 0.5 mg iodine/kg feed DM in lactating animals. The dimensioning of the ML was less aimed at diminishing risks to animal health than for consumer protection in preventing too much iodine via milk/-products and eggs. According to the German, Austrian and Swiss human nutrition societies, a maximum of 500 μg iodine per adult and day is tolerated [10] which represents the 2.5 fold recommended dietary allowance

* Corresponding author at: Thuringian State Institute of Agriculture, Investigation Unit – Laboratory, Dept. of Feed and Product Assessment, Naumburger Str. 98, D-07743, Jena, Germany.

E-mail address: friedrich.schoene@tll.thueringen.de (F. Schöne).

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(RDA) of 200 μg. Exceeding the upper dietary levels of iodine for humans will increase the risk of hyperthyroidism in connection with autoimmune diseases of the thyroid, [for details see Ref. [10]].

Despite a minor risk of too low or too high milk iodine content, on the German food market, the regular supply of iodine to the farm animals has transformed milk into an iodine carrier. Studies of bulk milk and consumer milk, respectively, from the period 1997–2012 demonstrated mean concentrations of between 94 and 122 μg iodine/kg milk [11–13]. However, there was no information about cows‘ iodine supply via feed in any of these studies. On the other hand, in controlled cow feeding experiments, relationships between milk and feed iodine concentration were found [14,15] which might be used for calculations of dose response in practice.

The present study should focus at the three sources of iodine in cow diets: (1) the straight feedstuffs, consisting of roughage, mainly silages, and concentrates; (2) mineral vitamin premixes; and (3) compound feeds with iodine being supplemented for 2 and 3. Mineral vitamin premixes represent the main iodine source and this should be quantified using the manufacturers‘ labelling of iodine concentrations but also that of the recommended premix administration per cow and day. From iodine intake via feed supplements a mean milk iodine concentration was predicted applying the aforementioned dose-response experiments and milk-monitoring studies. Nowadays, in most cow barns the three feed parts are prepared by so-called mixer feeders and the cow diet is fed as total mixed ration (TMR). This TMR should be analysed to validate the total iodine intake in selected Thuringian cow herds. In this study, the iodine content of bulk milk was also analysed. The iodine status reported for feed and milk has consequences for human iodine supply via milk and milk products which should be discussed in the context with the cited RDA.

2. Material and methods

2.1. Feed and milk samples

The straight feedstuffs analysed in the first part of the study, consisted of 41 concentrate and 32 silage samples. The concentrates sampled in 2015 and 2016 represented cereals (8x barley, 4x maize), couple products from oilseed processing (3x soya-bean meal and 11x RSM in both cases produced after solvent extraction, 7x rapeseed press cake produced by mechanical extraction) and 8x field beans. Samples of the silages, prepared from grass (17x) and from green maize (15x), originated from Thuringian harvest and silage monitoring in 2006 [16]. Hereby, grass silage samples were selected over a high range of ash and soil content, respectively. In the second investigation part, 21 mineral vitamin **premixes** (>40% ash) and 62 **compound feeds** were evaluated for iodine content and for the expected daily intake according to the labelled recommendation. Data originated from the official Thuringian feed surveillance in 2011; manufacturers' names and addresses and brands were anonymized. The premix allowances represented a g or kg range per cow and day in 9 cases – here the mean was used for calculation – or a percentage level of total feed DM in 12 cases. Hereby this "up to level", e.g. 1% of total diet DM, was multiplied with a daily intake of 20 kg DM per cow. Both these principal cases of labelling the recommended premix administration are explained in detail elsewhere [17]. For checking the reliability of labelled iodine concentration, the results of the analyses for the official Thuringian feed surveillance in the time period 2010–2016 were used representing a total of 59 premixes and compound feeds.

In the third investigation part, an *on-farm* study, **iodine in feed and milk** from six Thuringian cow herds was analysed. Herds comprised 350 to 1000 cows of the Holstein breed with 9000 to 12,500 kg mean milk yield per year. In two 4–5week periods during 5 Nov – 10 Dec. 2012 and 5 Nov – 6 Dec. 2013, a total of 51 TMR samples, 8–9 per herd and a total of 77 milk samples, 11–14 per herd were investigated. Bulk milk samples, in Period I 7–10 per herd, in Period II 4–5 per herd, originated from the laboratory of the Thuringian Association of Performance Testing in Animal Production Jena-Göschwitz, Germany, authorized for regular official sampling and official analysis of milk quality criteria, e.g. fat and protein content.

2.2. Analyses

All feed samples were milled through a 0.5 mm screen (Retsch ZM 200, Haan, Germany), the silages and TMR after predrying (1 kg at 60 °C for 96 h). Compound feeds and premixes were analysed on a feed basis. Straight feedstuffs and TMR were additionally analysed for DM by drying at 103 °C to a constant weight to calculate the iodine content in DM. The milk samples were freeze-dried and ground to a powder using mortar and pestle. The fresh milk iodine content was calculated on the basis of lyophilisate weight relative to the milk sample weight.

In all feed and milk samples, iodine was determined by inductively coupled plasma-mass spectrometry ICP-MS [14,18–20]. However, the extraction means differ between the official iodine analysis methods for food and feed: The procedure for iodine-supplemented feed is based on aqueous ammonia (NH_40H) solution [19], which for food [20] with frequently very low native iodine contents prescribes a stronger alkaline digestion with aqueous solution of tetramethylammonium hydroxide [TMAH = $(CH_3)_4N^+OH^-$]. To evaluate an effect of the extraction means, the iodine content of the silage samples was analysed parallel after the two extractions.

In straight feedstuffs, silages, TMR and milk for sample digestion, 500 mg air-dry material was added to 1 ml TMAH (AppliChem, Darmstadt, Germany) and 5 ml distilled deionized water in a 50 ml polypropylene tube with a gas-tight closure [20]. These samples were heated to 90 °C for 3 h for the iodine analysis and after cooling to room temperature 19 ml distilled deionized water was added and the aqueous layer separated by centrifugation at 4000 rpm for 15 min at 15 °C.

The samples of compound feed and premixes from official Thuringian feed surveillance were extracted by aqueous ammonia solution [19] along with, as part of the above mentioned extractionsí comparison, the silage samples. Each ground feed sample, 500 mg, was added to 100 ml of a 0.5% aqueous ammonia solution (made from 25% ammonia [Merck, Darmstadt, Germany] and distilled deionized water) in a vessel that was tightly sealed and kept overnight. Thereafter, the samples were shaken and then the solids were removed by filtration, [for details see Ref. [19]].

For the iodine-content analysis the ICP-MS equipment *NexION® 350X ICP-MS* (Perkin Elmer, Waltham, MA, USA) was used. Using the method of standard addition calibration, three potassium iodide (KI) aqueous standards were prepared (potassium iodide suprapure, Merck, Darmstadt, Germany) for the feed and milk samples [details in Ref. [14]].

This method had a limit of detection (LOD) of 3 μg iodine/kg DM and a limit of quantification (LOQ) of 9 μg iodine/kg DM which was established in low-iodine straight feedstuffs. That means for fresh milk with 130 g DM/kg LOD and LOQ of 0.4 and 1.4 μg iodine/kg fresh matter. The standards for feed analysis represented two samples with defined iodine contents from the official Austrian/German ring trial (Enquete of Arbeitsgemeinschaft für Lebensmittel-, Veterinär- und Agrarwesen, ALVA in 2014). Standard 1 (sample code ALVA PA 14/1) represents 0.61 mg iodine/kg feed, standard 2 (sample code ALVA PA 14/2) 1.39 mg/kg feed. In milk-iodine analysis the certified standard "Skimmed Milk Powder" BCR N 150–major and trace elements (Community Bureau of Reference, Brussels, Belgium) was used. The mean of 1.3 mg/kg iodine