Arsenic species in poultry feather meal

K.E. Nachman [a,b,c,*], G. Raber [d], K.A. Francesconi [d], A. Navas-Acien [b,e], D.C. Love [a,b]

[a] Center for a Livable Future, Johns Hopkins University, Baltimore, MD, USA
[b] Department of Environmental Health Sciences, Bloomberg School of Public Health, Johns Hopkins University, Baltimore, MD, USA
[c] Department of Health Policy and Management, Bloomberg School of Public Health, Johns Hopkins University, Baltimore, MD, USA
[d] Institute of Chemistry – Analytical Chemistry, Karl-Franzens University, Graz, Austria
[e] Department of Epidemiology, Bloomberg School of Public Health, Johns Hopkins University, Baltimore, MD, USA

ARTICLE INFO

ABSTRACT

Organoarsenical drugs are widely used in the production of broiler chickens in the United States. Feathers from these chickens are processed into a meal product that is used as an animal feed additive and as an organic fertilizer. Research conducted to date suggests that arsenical drugs, specifically roxarsone, used in poultry production result in the accumulation of arsenic in the keratinous material of poultry feathers. The use of feather meal product in the human food system and in other settings may result in human exposures to arsenic. Consequently, the presence and nature of arsenic in twelve samples of feather meal product from six US states and China were examined. Since arsenic toxicity is highly species-dependent, speciation analysis using HPLC/ICPMS was performed to determine the biological relevance of detected arsenic. Arsenic was detected in all samples ($44–4100\ \mu g\ kg^{-1}$) and speciation analyses revealed that inorganic forms of arsenic dominated, representing 37 – 83% of total arsenic. Roxarsone was not detected in the samples ($<20\ \mu g$ As kg^{-1}). Feather meal products represent a previously unrecognized source of arsenic in the food system, and may pose additional risks to humans as a result of its use as an organic fertilizer and when animal waste is managed.

1. Introduction

Arsenic is a naturally-occurring metalloid element that can be found in environmental media as a result of numerous natural and anthropogenic processes. An extensive body of literature links chronic, low-level exposures to arsenic with a spectrum of human morbidities, including cancers of the lung, bladder, kidney and skin (Chen et al., 1988, 1992; Wu et al., 1989), as well as cardiovascular disease (Chen et al., 2011; Medrano et al., 2010; Sohel et al., 2009), type 2 diabetes (Navas-Acien et al., 2008), neurocognitive deficits (Wasserman et al., 2007) and adverse birth outcomes (Ahmad et al., 2001). Research to date strongly suggests that the toxicity of arsenic is highly species-dependent; inorganic species (arsenate and arsenite) are believed to be most potent in inducing toxic effects (National Research Council, 2001). The toxic potential of various organic arsenic species (methylated arsenic acids, arsenobetaine, and arsenosugars) has been less studied (National Research Council, 1999).

Organoarsenical drugs are approved for use in the United States (US) for production of broiler chickens to prevent coccidiosis, enhance feed conversion and promote growth (Silbergeld and Nachman, 2008). The most commonly used organoarsenical drug in poultry production is roxarsone ((4-hydroxy-3-nitrophenyl) arsonic acid, brand name "3-Nitro"), which is approved for use at a rate of 25–50 ppm in feed. It is estimated that 88% of the approximately 8.6 billion broiler chickens produced each year (United States Department of Agriculture, 2011) in the US are fed roxarsone, as stated by a representative of an industry trade group testifying in the Maryland State General Assembly in 2011. Roxarsone is also approved for use in turkeys and swine, of which approximately 244 million and 67 million, respectively, are produced annually (Food and Agriculture Organization of the United Nations, 2011a). Estimates of the frequency of arsenical use in production of these animals are unavailable (Makris et al., 2008).

Arsenic concentrations in the waste of poultry fed roxarsone have been reported in the range of $14–76\ mg\ kg^{-1}$ (Arai et al., 2003; Jackson and Bertsch, 2001; Jackson et al., 2003). Research has demonstrated rapid biotic conversion of roxarsone in poultry waste into inorganic forms (Garbarino et al., 2003), a large fraction of which (75%) is water soluble and capable of horizontal transport from application sites (Rutherford et al., 2003). Surface soils with a long history of treatment with poultry waste were reported to have arsenic concentrations between 12 and $15\ mg\ kg^{-1}$, suggesting either vertical or horizontal arsenic transport (Arai et al., 2003). Other studies have

suggested that accumulation of arsenic can occur over time in agricultural soils amended with poultry waste (Church et al., 2010). Novel methods for disposal and recycling of poultry waste, such as pelletization and waste-to-energy incineration, have highlighted concerns for human arsenic exposure that may result from its management (Nachman et al., 2005, 2008; Stingone and Wing, 2011).

In addition to poultry litter, other process wastes are associated with industrial production of poultry, many of which are rendered, or recycled into products that are used for other purposes. Poultry feathers are a byproduct of the production of poultry for human consumption. The US produced a combined total of 25 billion kg of meat from broiler chicken and turkey in 2010 (United States Department of Agriculture, 2011). Of the live weight of broiler chickens produced, approximately 37% is not consumed directly by humans (Meeker and Hamilton, 2006) (similar estimates are not available for turkeys). This inedible fraction consists of heads, bones, viscera, and feathers, and becomes one source of raw material for the rendering industry. These remaining parts are processed by the rendering industry into feather meal and poultry by-product meal. In some cases, inedible poultry parts are mixed with those of other animals into mixed animal meat and bone meal (Meeker and Hamilton, 2006).

The method for processing feathers into feather meal involves the removal of feathers from poultry carcasses, chopping and rinsing of feathers, autoclaving of feathers (115–140 °C for 45 min), the pressing of rendered material to remove fats, and the drying and grinding of feathers (Elboushy et al., 1990; Meeker and Hamilton, 2006; Papadopoulos, 1985; Yu, 2008). In some cases, viscera, heads, feet, and manure can be added to feather meal before autoclaving to increase certain amino acids (from viscera, heads, feet) and minerals (from manure) that poultry feathers lack (Elboushy et al., 1990). Poultry by-product meal and mixed animal meat and bone meal are produced in a similar manner (Meeker and Hamilton, 2006).

Forty-four percent by weight of US non-tallow rendering is derived from poultry, and over a third of poultry rendering, by weight, is feather meal (Swisher, 2009). In 2008, the US rendering industry produced 604 million kg of feather meal, of which >90% was used domestically (Swisher, 2009). Feather meal is used as an organic fertilizer (Hadas and Kautsky, 1994), a raw material in biodiesel (Kondamudi et al., 2009), and as a feed additive in poultry feed (Elboushy et al., 1990), pig feed (VanHeugten and VanKempen, 2002), ruminant feed (Food and Agriculture Organization of the United Nations, 2011b), and fish feed (Arunlertaree and Moolthongnoi, 2008; Jamil et al., 2007).

Given the many uses of feather meal, including numerous applications in the human food system, the presence of residual arsenic in feather meal may pose human and animal health concerns. Examinations of residual arsenic in the feathers of poultry fed arsenical drugs are limited, but a single evaluation of poultry feathers conducted in the late 1960s found that chickens treated with roxarsone had higher total arsenic concentrations in their feathers than those raised without (Morrison, 1969). This finding is supported by a strong biological basis for arsenic deposition in animal keratinous tissues, including poultry feathers and human fingernails. Fingernails are widely used as arsenic exposure biomarkers in human populations (Slotnick and Nriagu, 2006), and feathers have been used on occasion as arsenic exposure biomarkers in wild bird populations (Dauwe et al., 2000). It is hypothesized that since arsenic accumulates in the feathers of poultry receiving arsenical drugs, feather meal product from those feathers is likely contaminated with arsenic.

An understanding of the significance of feather-borne arsenic to the human and animal food systems requires characterization of the species of arsenic present in feather meal product. An examination of *Clostridium* species, a dominant bacterium in the cecum of chickens, demonstrated the ability of the bacteria to rapidly transform roxarsone into inorganic arsenicals (Stolz et al., 2007). A Food and Drug Administration (FDA)-conducted study of arsenic residues in chicken livers found roxarsone administration to be associated with increases in the inorganic arsenic content of the livers (Food and Drug Administration, 2011a). Taken together, these studies suggest that residual arsenic found in poultry feathers may be in inorganic forms.

To characterize the arsenic in feather meal, we examined samples of feather meal product for the presence and concentration of total arsenic and arsenic species. These data will inform on concentration levels and on the biological relevance of arsenic residues in feather meal, a widely used additive to animal feed and fertilizer products.

2. Material and methods

2.1. Sample collection

We collected samples of feather meal intended for sale as agricultural fertilizer or as animal feed from six US states and through online vendors. Feather meal intended for use as fertilizer was available for purchase in 22 kg bags, whereas feather meal for use as an ingredient in animal feed was not packaged for direct sale. We identified several rendering plants, distributors, or feed mills as vendors from which feather meal samples intended for use as animal feed could be acquired.

For bagged feather meal, upon purchase, bags were turned upside-down five times, opened, and 50 g subsamples were collected with a sterile scoop and bag (Whirl-Pac, Nasco, Fort Atkinson, WI). For unbagged feather meal, which was typically available in large bins from animal feed stores, samples were collected in plastic bags using sterile scoops. Where available, the product brand name, purchase location, distributor, rendering plant and poultry company were recorded. Feather meal products were not labeled as to the use of organoarsenicals during broiler chicken production, nor did product labels note the addition of non-feather biological materials. Samples were shipped by commercial carrier to the Johns Hopkins Bloomberg School of Public Health in Baltimore, Maryland, and stored at room temperature (25 °C) in the dark. The samples were transported to the Institute of Chemistry-Analytical Chemistry, Karl Franzens University Graz, Austria for total arsenic and arsenic speciation analyses.

2.2. Total arsenic measurements

A portion of sample powder (ca 250 mg weighed with a precision of 0.1 mg) or a portion of the extract of the powder was mineralized with nitric acid (2 mL) and water (2 mL) in an Ultraclave III microwave digestion system (MLS, Leutkirch, Germany) under an argon pressure of 4×10^6 Pa at 250 °C for 30 min. After mineralization, the samples were diluted to 10 mL with water/methanol (19 + 1, v/v) The methanol served as the carbon source to enhance the arsenic response, and also eliminated problems with quantification resulting from small differences in residual carbon between samples. The acid and methanol content in the calibration standard solutions were matched to the acid and methanol content in the sample solutions.

The arsenic content in the solutions was determined by inductively coupled plasma mass spectrometry (ICPMS; Agilent 7500ce from Agilent Technologies, Waldbronn, Germany) using helium as collision cell gas (for removing polyatomic interferences). [74]Ge and [115]In served as internal standards whereby a solution containing 500 µg/L Ge and In was added on-line via a T-piece. The calibration standard was Single-Element Arsenic Standard P/N S4400-100031 (CPI International, Santa Rosa, CA, US), As in 2% nitric acid, 1000 ± 3 µg As mL^{-1}. Calibration range was 0.2 µg As·L^{-1}–100 µg As L^{-1}. Certified Reference Material Rice flour 1568a (National Institute of Standards and Technology, Gaithersburg, US) was used to validate the method: we obtained 295 ± 20 µg As kg^{-1}, (mean $\pm$ SD, n = 6); the certified value is 290 ± 30 µg As kg^{-1}. Samples were analysed in duplicate;

when the duplicate results differed by more than 5%, further analyses were performed. This was the case for the two samples from China.

2.3. Arsenic species measurements

To a portion of powder (ca 250 mg to a precision of 0.1 mg) weighed directly into a 12 mL quartz tube, was added 5 mL of 0.1 M trifluoroacetic acid containing 50 μL H_2O_2 (30% v/v); the suspension was sonicated for 10 min and left to stand overnight. Samples were then microwave-extracted in an Ultraclave III microwave digestion system. Extraction was performed under an argon pressure of 4×10^6 Pa; the temperature was ramped from room temperature to 95 °C in 10 min and maintained at 95 °C for 60 min. The use of H_2O_2 ensured that any arsenite in the sample was oxidised to arsenate presenting a single inorganic arsenic species for HPLC analysis. MA and DMA were not affected by this treatment. After cooling to room temperature, portions of the extracts were removed for total arsenic measurement (as described above); another portion was transferred to an HPLC vial for the measurement of arsenic species by HPLC/ICPMS.

An additional extraction procedure was performed for roxarsone measurements because our previous work had indicated that the TFA/H_2O_2 system might lead to low recoveries for this species in certain matrices. Thus, we used an aqueous methanol extraction as reported in Wang et al. (2010). Briefly, a portion (500 mg) of the dried sample was shaken with 10 mL of MeOH/H_2O (1+1, v/v) at room temperature overnight. The suspension was centrifuged at 4700 rcf for 20 min, and the clear supernatant was directly used for HPLC/ICPMS measurements. To test the stability of roxarsone during the sample preparation, portions of the feather meal samples were spiked with 0.5 μg As as roxarsone and treated as described above. The recoveries (extraction plus HPLC) ranged from 70% to 87% for the 12 samples with an average recovery of 81%.

Measurement of arsenic species was performed with an Agilent 1100 series HPLC connected to an Agilent 7500ce ICPMS. The signals at m/z 75 (^{75}As, ^{40}Ar^{35}Cl) and m/z 77 (^{40}Ar^{37}Cl, to ascertain possible chloride interference on m/z 75) were monitored using an integration time of 300 ms. HPLC was performed under both anion- and cation-exchange conditions. For anion-exchange chromatography, used for the quantification of methylarsonate, dimethylarsinate, and arsenate, the conditions were: a Hamilton PRP-X100 column (250×4.6 mm, 5 μm particle size) with a mobile phase of 10 mM malonic acid, adjusted to pH 5.6 using aqueous ammonia, at a flow rate of 1.2 mL min^{-1}; column temperature was 40 °C and injection volume was 20 μL. Under these conditions, however, roxarsone elutes very late (>20 min) as a broad peak. Consequently, for the determination of roxarsone, the following HPLC conditions were employed: PRP-X100 150*4.6 mm; 100 mM malonic acid adjusted to pH 9.5 with aqueous ammonia; 1 ml min^{-1}; 40 °C; 20 μL injection volume. Roxarsone elutes in 4 min under these conditions. For cation-exchange chromatography, a Varian Ionosphere C column (100×3 mm, 5 μm particle size) was used with a mobile phase of 10 mM aqueous pyridine, adjusted to pH 2.5 using aqueous formic acid, at a flow rate of 1.5 mL min^{-1}; column temperature was 40 °C and injection volume was 20 μL. Data evaluation was carried out with Agilent chromatographic software G1824C Version C.01.00. The quantification was done by external calibration, based on peak areas, against standard arsenic species. Stock solutions containing 1000 mg As L^{-1} each of the following species were prepared in water: arsenate prepared from Na$_2$HAsO$_4$.7H$_2$O (Merck, Darmstadt, Germany); methylarsonate prepared in-house from methylarsonic acid synthesised from As$_2$O$_3$ and CH$_3$I; dimethylarsinate prepared from sodium dimethylarsinate (Fluka, Buchs, Switzerland); and arsenobetaine as the bromide salt prepared in-house. Roxarsone ((4-hydroxy-3-nitrophenyl) arsonic acid, grade Vetranal) was purchased from Sigma Aldrich; the powder

was dissolved in methanol to a concentration of 1000 mg As L^{-1} and further dilution to the final concentrations were made in water.

There is currently no reference material certified for inorganic arsenic. Analysis of Rice flour 1568a (National Institute of Standards and Technology) returned the following data: dimethylarsinate (186 μg As kg^{-1}), methylarsonate (13 μg As kg^{-1}), and inorganic arsenic (96 μg As kg^{-1}). Extraction efficiency was 102% and column recovery (anion-exchange) was 99%. These data for arsenic species compare well with those published for Rice flour 1568a by other research groups; for example, Sun et al. (2008) found 110 μg As kg^{-1} as inorganic arsenic and 180 μg As kg^{-1} as organic arsenic in this material.

3. Results

3.1. Sample collection

One or more samples of feather meal were acquired from six US states (one sample each from California, Idaho, Pennsylvania and Tennessee, two samples from Arkansas, and four samples from Oregon) and two samples were obtained from a Chinese internet vendor for a total of twelve samples (Table 1). For states with more than one sample, each sample was purchased from a separate vendor, except for the samples from Arkansas, where samples were purchased from the same vendor at different times. The Chinese samples were purchased from the same vendor, but were purchased as different products (based on differences in protein content). Samples purchased from Arkansas, Oregon, and Pennsylvania were intended for use as fertilizer, whereas the samples from California, Idaho and Tennessee were marked for use in animal feed. A fair degree of heterogeneity in appearance (differing colors and consistencies) was observed across the samples (Fig. 1).

3.2. Total arsenic content and arsenic species

Arsenic was detected in all samples and concentrations spanned three orders of magnitude (44–4100 μg kg^{-1}), with the highest values being found in the Chinese samples (Table 1). The median concentration of total arsenic intended for use as fertilizer was 410 μg kg^{-1} (range 44 to 1280 μg kg^{-1}); the median concentration for samples intended as animal feed additive was 150 μg kg^{-1} (range 93 to 4100 μg kg^{-1}). The two samples from China differed from the other samples by having considerable amounts of insoluble material after the digestion process. These samples also gave a high coefficient of variation for replicate analyses (almost 25% compared with ca 5% for other samples) suggesting that the samples were not homogeneous.

Extraction of samples with trifluoroacetic acid/H_2O_2 prior to HPLC showed generally good extraction efficiencies (Table 1; median value almost 80%). The two samples from China were again different by recording low extraction efficiencies (40% and 53%). Attempts to extract more of the arsenic, and thereby gain a fuller picture of arsenic speciation, by using other solvent mixtures (e.g. aqueous methanol mixtures) were not successful. HPLC recoveries of arsenic had a median value >90%, and this arsenic was distributed among three main species, inorganic arsenic, methylarsonate and dimethylarsinate. Inorganic arsenic (arsenite + arsenate) was the dominant form of arsenic in the samples, accounting for between 37–83% of the total arsenic and 60–100% of the sum of arsenic species quantified by HPLC. Methylated arsenicals accounted for a smaller fraction of the total arsenic present (<5 to 36.8%). Arsenobetaine was below the detection limit (<10 μg As kg^{-1}) for eleven samples, with only one sample showing detectable quantities (but it was less than 5% of the total arsenic). Our preliminary studies with the trifluoroacetic acid/H_2O_2 extraction medium indicated that it was not suitable for roxarsone measurements. Consequently,

Table 1
Sample collection, total arsenic and arsenic speciation results.

Sample	Purchase location	Intended use	Total As	Extraction efficiency	DMA	MA	iAs	Column recovery	% iAs of total As (% iAs as sum of species)
			$\mu g\ kg^{-1}$	%	$\mu g\ kg^{-1}$			%	
1	Arkansas, US	Fertilizer	480	75	80	30	270	106%	56% (71%)
2	Arkansas, US	Fertilizer	440	75	40	20	210	82%	48% (78%)
3	Oregon, US	Fertilizer	52	100	12	<10	40	100%[a]	77% (80%)
4	Oregon, US	Fertilizer	44	131	<10	<10	30	52%[a]	68% (100%)
5	Oregon, US	Fertilizer	410	96	30	30	340	102%	83% (85%)
6	Oregon, US	Fertilizer	90	77	<10	<10	40	58%[a]	44% (100%)
7[b]	Pennsylvania, US	Fertilizer	1280	75	60	60	760	98%	59% (81%)
8	California, US	Animal feed	114	99	35	<10	62	86%	53% (60%)
9[c]	Guangdong, China	Animal feed	4100	40	130	50	1500	104%	37% (88%)
10	Guangdong, China	Animal feed	1500	53	80	20	690	99%	46% (87%)
11	Idaho, US	Animal feed	93	105	20	<10	60	82%	65% (75%)
12	Tennessee, US	Animal feed	150	80	10	<10	80	75%	53% (89%)

[a] Variable and usually low column recoveries are often found when total arsenic concentrations are low, as discussed in Schaeffer et al. (2006).

[b] Arsenobetaine was present in this sample (sample 7, Pennsylvania), at a concentration of 60 μg As kg^{-1}. Arsenobetaine was not detected (<10 μg As kg^{-1}) in the other samples.

[c] Traces of unidentified arsenic cations (ca 30 μg As kg^{-1}) were present in this sample.

we applied a separate (milder) extraction with aqueous methanol, as reported by Wang et al. (2010), followed by HPLC/ICPMS under a set of HPLC conditions optimized for roxarsone. However, we did not detect roxarsone (<20 μg As kg^{-1}) in any of the 12 samples using these arsenic speciation conditions.

4. Discussion

Analyses demonstrated measurable arsenic in all samples of the feather meal product, a significant fraction of which was present in inorganic form. Total arsenic concentrations varied markedly across
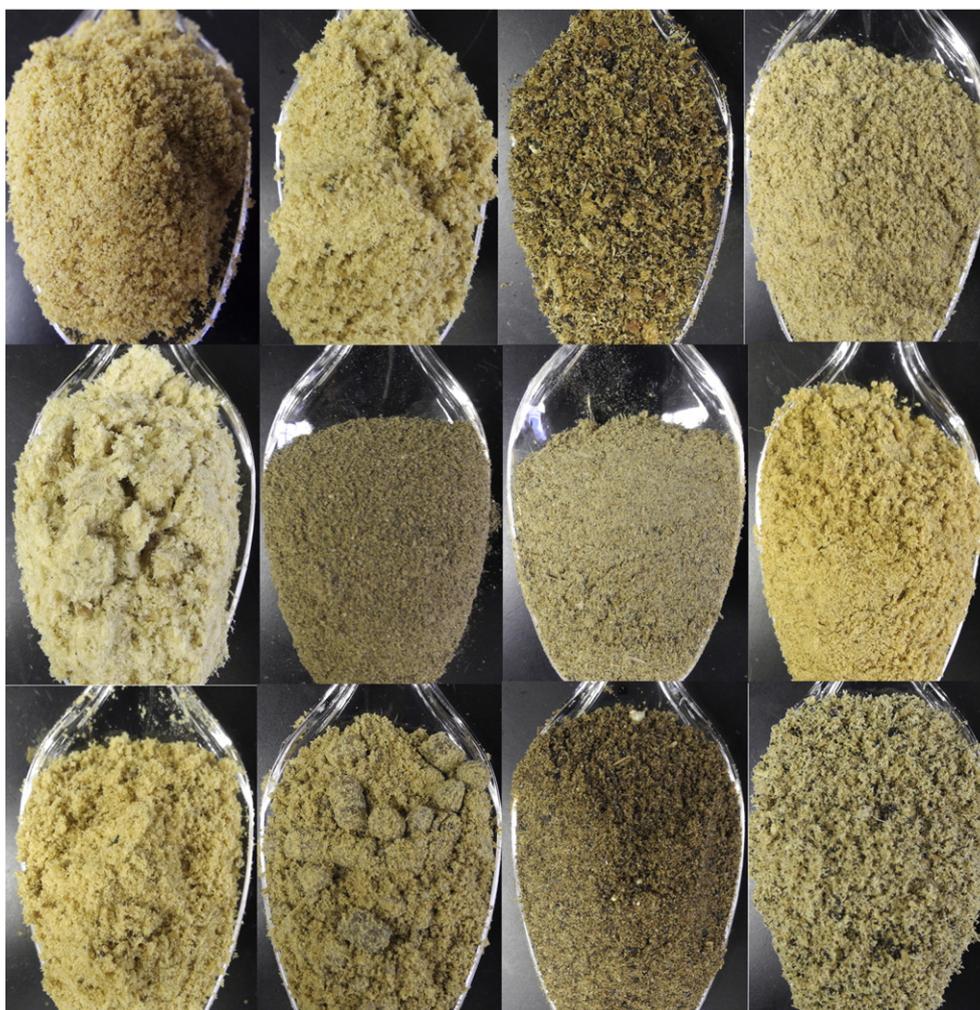

Fig. 1. Feather meal samples from the United States and China.

feather meal samples, including samples used as fertilizer and samples used as animal feed. Also, a considerable fraction of arsenic was non-extractable — up to 60% in the samples from China and up to 25% in the samples from the US. Our experience with other types of samples and matrices, where inorganic arsenic and simple methylated forms are efficiently extracted under the same conditions used for the feather meal, suggests that the arsenic in feather meal, particularly in those samples from China, was likely to be largely present in other forms. This non-extractable arsenic was unlikely to be roxarsone-based on the fact that we could not detect roxarsone in the (native) feather meal samples although we obtained good recoveries of this species (70–87%) when we spiked it to feather meal samples. It remains possible, however, that roxarsone is actually present in feather meal but is so tightly bound within the sample matrix that very forcing conditions are required to release it. Unfortunately, such conditions are also likely to decompose roxarsone. This highlights one of the inherent weaknesses of speciation analysis by HPLC/ICPMS, i.e. the compromise between extraction efficiency and maintaining the true species identity, as discussed in Francesconi and Kuehnelt (2004). For the Chinese feather meal sample, a solid state analytical procedure such as X-ray spectroscopy may provide valuable arsenic speciation data complementary to the data provided by HPLC/ICPMS. The application and benefits of complementary arsenic speciation techniques to biological samples has recently been discussed (Feldmann et al., 2009). It is also possible that roxarsone might be present in feathers of live (or freshly processed) chickens), but is degraded in the production of feather meal which includes an autoclaving step at up to 140 °C.

Feather meal is routinely added to poultry, swine, ruminant and fish feed; its rate of inclusion in chicken feed ranges from 1 – 7% (m/m) (Food and Agriculture Organization of the United Nations, 2011b; Morrison, 1969) and can be even higher in ruminant feed (Aderibigbe and Church, 1983). The presence of quantifiable inorganic arsenic species in feather meal product serves as an additional source of inorganic arsenic to the food system and represents a previously unrecognized source of human arsenic exposure. Arsenic added to poultry, swine and fish feed might contribute to increased levels of inorganic arsenic in food animals. Our understanding of the toxicokinetics of arsenicals in animal foods, including roxarsone used in poultry production and inorganic arsenic measured in feather meals used as animal feed in this study, is limited. This lack of knowledge complicates the determination of the impact of using feather meal as a feed additive on arsenic concentrations in food animals. Also, whether inorganic arsenic detected in feather meal is the result of roxarsone metabolism in chicken is unclear. A recent study, however, concluded that roxarsone administration increased the inorganic arsenic content in the livers of treated broiler chickens relative to controls (Food and Drug Administration, 2011a).

The Animal Feed Safety System is a regulatory program of the FDA that is responsible for ensuring the safety of feed used for raising food animals. A secondary objective of this federal program is to protect humans from illness caused by exposures to hazards in animal feed (Food and Drug Administration, 2010). While the FDA has not developed standards for arsenic residues in animal feed, it recommends using maximum tolerance values of 12.5 to 50 mg total As kg^{-1} diet as proposed by the National Research Council (National Research Council, 2005). There have been no reports of federal or state testing of arsenic in feather meal intended for use in animal feed.

Environmental pathways of arsenic exposure resulting from roxarsone use have been characterized (Silbergeld and Nachman, 2008), and it is expected that arsenic from feather meal that is unconsumed or unabsorbed by poultry and swine would contribute to arsenic loadings in animal waste. This contribution would add to other recognized sources of arsenic contamination of animal waste, including unconsumed roxarsone in spilled animal feed and excreted roxarsone and its metabolites. Methods for management of animal house waste have been demonstrated to transfer waste-borne contaminants to a wide

spectrum of environmental media with which humans have contact, including air, surface and ground water, and soils (Church et al., 2010; Nachman et al., 2005; Silbergeld and Nachman, 2008; Stingone and Wing, 2011).

The use of feather meal as an organic fertilizer may result in additional unanticipated human exposures to inorganic arsenic. Activities related to spreading the feather meal product in the context of gardening may result in unintentional ingestion and inhalation of, or dermal contact with residual inorganic arsenic. Further, certain food crops can accumulate arsenic from soils, and hence the use of feather meal as fertilizer could lead to increased dietary arsenic exposures (Zhao et al., 2010). Some US states have established regulatory policies regarding arsenic content in feather meal used as fertilizer, although only Oregon and Washington report their inspection results online (Association of American Plant Food Control Officials, 2011; Oregon Department of Agriculture, 2011; Washington State Department of Agriculture, 2011). The Oregon Department of Agriculture reported total arsenic concentrations ranging from 690 to 2,900 µg kg^{-1} in seven feather meal products (Oregon Department of Agriculture, 2011), and a similar range in arsenic concentrations was reported by the state of Washington (250 to 2,900 µg kg^{-1} in four products) (Washington State Department of Agriculture, 2011). Our results are consistent with those reports. Additionally, our study was able to show that the majority of arsenic in feather meal is present as inorganic arsenic, whereas arsenic speciation was not performed in the earlier studies in Oregon or Washington.

While our analyses clearly indicate the presence of arsenic contamination in the finished feather meal product, uncertainty remains as to the origin of the detected arsenic. In humans, the accumulation of arsenic in keratinous tissues such as skin, hair and nails has been well characterized (Hughes, 2006). The keratinous composition of poultry feathers (Barone and Schmidt, 2006) may suggest a similar potential for arsenic accumulation. Though product labels do not suggest the intentional addition of materials other than hydrolyzed chicken feathers, it is not possible to rule out contamination of the feather meal product with arsenic-bearing poultry waste (Arai et al., 2003) or other materials. The marked variability in color and consistency may be a reflection of the presence of materials other than feathers in the meal product. It is also possible that the inherent variation in the appearance of chicken feathers may influence the hue of the feather meal product. Consequently, it may be the case that the arsenic recovered from the finished product originates from the feathers, poultry waste, or a combination of the two.

5. Conclusion

The current research demonstrated the presence of inorganic arsenic as the major arsenic species in samples of hydrolyzed feather meal, an ingredient in feed for food animals and a material used as organic fertilizer. Feather meal used as animal feed and as a fertilizer could contribute to inorganic arsenic exposure in persons who consume meat, use organic fertilizers, or come into contact with environmental media impacted by the waste stream from food animal production sites. In this study we had no information on the precise contribution of roxarsone to inorganic arsenic exposure in feather meal. However, given the common use of roxarsone in poultry production, the increase in inorganic arsenic exposure in livers of chicken fed roxarsone, and the likely accumulation of inorganic arsenic in feathers, inorganic arsenic measured in feather meal in this study may have originated from the practice of administering roxarsone to broiler chickens in the context of industrial poultry production. Cessation of arsenical drug usage in poultry production would eliminate an unnecessary contribution of arsenic to the human food supply and would limit the re-introduction of large quantities of arsenic into the natural environment. Such a cessation may be temporarily observed in the US, where the Pfizer pharmaceutical company has

voluntarily suspended domestic marketing of the drug (Food and Drug Administration, 2011b); despite this, no regulatory measures to modify the approval of the drug have been taken, and Pfizer may return roxarsone to the US market without consequence. Further, roxarsone use is likely to persist outside of the US, where Pfizer continues to market the drug (Food and Drug Administration, 2011c).

Acknowledgement

References

Aderibigbe AO, Church DC. Feather and hair meals for ruminants. III. Relationship between enzymatic or in vitro rumen digestibility and in vivo digestibility of diets containing feather and hair meals. J Anim Sci 1983;57:483–94.

Ahmad SA, Sayed MH, Barua S, Khan MH, Faruquee MH, Jalil A, et al. Arsenic in drinking water and pregnancy outcomes. Environ Health Perspect 2001;109:629–31.

Arai Y, Lanzirotti A, Sutton S, Davis JA, Sparks DL. Arsenic speciation and reactivity in poultry litter. Environ Sci Technol 2003;37:4083–90.

Arunlertaree C, Moolthongnoi C. The use of fermented feather meal for replacemnet of fish meal in the diet of oreochromis niloticus. Environ Nat Resour J 2008;6:13–24.

Association of American Plant Food Control Officials. State Internet Sites for Trace Metal Analysis of Fertilizer Products Registered in Those States. Available at: http://www.aapfco.org/metals.htm2011. [accessed November 4, 2011].

Barone JR, Schmidt WF. Effect of formic acid exposure on keratin fiber derived from poultry feather biomass. Bioresour Technol 2006;97:233–42.

Chen CJ, Kuo TL, Wu MM. Arsenic and cancers. Lancet 1988;1:414–5.

Chen CJ, Chen CW, Wu MM, Kuo TL. Cancer potential in liver, lung, bladder and kidney due to ingested inorganic arsenic in drinking water. Br J Cancer 1992;66:888–92.

Chen Y, Graziano JH, Parvez F, Liu M, Slavkovich V, Kalra T, et al. Arsenic exposure from drinking water and mortality from cardiovascular disease in Bangladesh: prospective cohort study. BMJ 2011;342.

Church CD, Kleinman PJA, Bryant RB, Saporito LS, Allen AL. Occurrence of Arsenic and Phosphorus in Ditch Flow from Litter-amended Soils and Barn Areas. J Environ Qual 2010;39:2080–8.

Dauwe T, Bervoets L, Blust R, Pinxten R, Eens M. Can excrement and feathers of nestling songbirds be used as biomonitors for heavy metal pollution? Arch Environ Contam Toxicol 2000;39:541–6.

Elboushy A, Vanderpoel A, Walraven O. Feather meal - a biological waste - its processing and utilization as a feedstuff for poultry. Biol Wastes 1990;32:39–74.

Feldmann J, Salaün P, Lombi E. Critical review perspective: elemental speciation analysis methods in environmental chemistry – moving towards methodological integration. Environ Chem 2009;6:275–89.

Food and Agriculture Organization of the United Nations. FAOSTAT. 2011, Rome, Italy; 2011a. Available at:http://faostat.fao.org/default.aspx [accessed November 4, 2011].

Food and Agriculture Organization of the United Nations. Feather meal, Feathermeal, Hydrolyzed feather meal, Poultry feather meal, Hydrolyzed poultry feather meal, Keratin meal. 2011, Rome, Italy; 2011b. Available at:http://www.fao.org/ag/AGA/AGAP/FRG/AFRIS/Data/326.HTM [accessed November 4, 2011].

Food and Drug Administration. Fourth Draft: Framework of the FDA Animal Feed Safety System;2010. Available at:http://www.fda.gov/AnimalVeterinary/SafetyHealth/AnimalFeedSafetySystemAFSS/ucm196795.htm. [accessed November 4, 2011].

Food and Drug Administration. Provide data on various arsenic species present in broilers treated with roxarsone: Comparison with untreated birds. Study 275.30 Laurel, Maryland;2011a. Available at:http://www.fda.gov/downloads/AnimalVeterinary/SafetyHealth/ProductSafetyInformation/UCM257545.pdf [accessed November 4, 2011].

Food and Drug Administration. FDA: Pfizer will voluntarily suspend sale of animal drug 3-Nitro;2011b. Available at:http://www.fda.gov/NewsEvents/Newsroom/PressAnnouncements/ucm258342.htm. [accessed December 4, 2011].

Food and Drug Administration. Questions and Answers Regarding 3-Nitro (Roxarsone);2011c. Available at:http://www.fda.gov/AnimalVeterinary/SafetyHealth/ProductSafetyInformation/ucm258313.htm. [accessed December 4, 2011].

Francesconi KA, Kuehnelt D. Determination of arsenic species: a critical review of methods and applications, 2000–2003. Analyst 2004;129:373–95.

Garbarino JR, Bednar AJ, Rutherford DW, Beyer RS, Wershaw RL. Environmental fate of roxarsone in poultry litter. I. Degradation of roxarsone during composting. Environ Sci Technol 2003;37:1509–14.

Hadas A, Kautsky L. Feather meal, a semi slow-release nitrogen fertilizer for organic farming. Fert Res 1994;38:165–70.

Hughes MF. Biomarkers of exposure: a case study with inorganic arsenic. Environ Health Perspect 2006;114:1790–6.

Jackson BP, Bertsch PM. Determination of arsenic speciation in poultry wastes by IC-ICP-MS. Environ Sci Technol 2001;35:4868–73.

Jackson BP, Bertsch PM, Cabrera ML, Camberato JJ, Seaman JC, Wood CW. Trace element speciation in poultry litter. J Environ Qual 2003;32:535–40.

Jamil K, Abbas G, Akhtar R, Hong L, Zhenxing L. Effects of Replacing Fishmeal with Animal By-products Meal Supplementation in Diets on the Growth and Nutrient Utilization of Mangrove Red Snapper. J Ocean Univ China 2007;6:292–8.

Kondamudi N, Strull J, Misra M, Mohapatra SK. A green process for producing biodiesel from feather meal. J Agr Food Chem 2009;57:6163–6.

Makris KC, Quazi S, Punamiya P, Sarkar D, Datta R. Fate of arsenic in swine waste from concentrated animal feeding operations. J Environ Qual 2008;37:1626–33.

Medrano MA, Boix R, Pastor-Barriuso R, Palau M, Damian J, Ramis R, et al. Arsenic in public water supplies and cardiovascular mortality in Spain. Environ Res 2010;110:448–54.

Meeker D, Hamilton C. An overview of the rendering industry. In: Meeker DL, editor. Essentials of Rendering: All About the Animal By-product Industry. Arlington, VA: National Renderers Association; 2006. p. 1-16.

Morrison JL. Distribution of arsenic from poultry litter in broiler chickens, soil, and crops. J Agric Food Chem 1969;17:1288–90.

Nachman KE, Graham JP, Price LB, Silbergeld EK. Arsenic: a roadblock to potential animal waste management solutions. Environ Health Perspect 2005;113:1123–4.

Nachman KE, Mihalic JN, Burke TA, Geyh AS. Comparison of arsenic content in pelletized poultry house waste and biosolids fertilizer. Chemosphere 2008;71:500–6.

National Research Council. Arsenic in Drinking Water. Washington, D.C.: National Academy Press; 1999.

National Research Council. Arsenic in Drinking Water: 2001 Update. Washington, DC: National Academy Press; 2001.

National Research Council. Mineral Tolerance of Animals. Washington, DC: National Academies Press; 2005.

Navas-Acien A, Silbergeld EK, Pastor-Barriuso R, Guallar E. Arsenic Exposure and Prevalence of Type 2 Diabetes in US Adults. JAMA 2008;300:814–22.

Oregon Department of Agriculture. Search Registered Products. 2011, Salem, Oregon. Available at:; 2011http://oda.state.or.us/dbs/heavy_metal/search.lasso [accessed November 4, 2011].

Papadopoulos M. Processed chicken feathers as feedstuff for poultry and swine. A review. Agric Wastes 1985;14:275–90.

Rutherford DW, Bednar AJ, Garbarino JR, Needham R, Staver KW, Wershaw RL. Environmental fate of roxarsone in poultry litter. Part II. Mobility of arsenic in soils amended with poultry litter. Environ Sci Technol 2003;37:1515–20.

Schaeffer R, Francesconi KA, Kienzl N, Soeroes C, Fodor P, Váradi L, et al. Arsenic speciation in freshwater organisms from the river Danube in Hungary. Talanta 2006;69:856–65.

Silbergeld EK, Nachman K. The environmental and public health risks associated with arsenical use in animal feeds. Ann N Y Acad Sci 2008;1140:346–57.

Slotnick MJ, Nriagu JO. Validity of human nails as a biomarker of arsenic and selenium exposure: A review. Environ Res 2006;102:125–39.

Sohel N, Persson LA, Rahman M, Streatfield PK, Yunus M, Ekstrom EC, et al. Arsenic in drinking water and adult mortality: a population-based cohort study in rural Bangladesh. Epidemiology 2009;20:824–30.

Stingone JA, Wing S. Poultry litter incineration as a source of energy: reviewing the potential for impacts on environmental health and justice. New solut 2011;21:27–42.

Stolz JF, Perera E, Kilonzo B, Kail B, Crable B, Fisher E, et al. Biotransformation of 3-nitro-4-hydroxybenzene arsonic acid (roxarsone) and release of inorganic arsenic by Clostridium species. Environ Sci Technol 2007;41:818–23.

Sun G-X, Williams PN, Carey A-M, Zhu Y-G, Deacon C, Raab A, et al. Inorganic Arsenic in Rice Bran and Its Products Are an Order of Magnitude Higher than in Bulk Grain. Environ Sci Technol 2008;42:7542–6.

Swisher K. Market Report 2008: Times were good, until prices collapsed. Render Magazine; 2009. April: 10–17.

United States Department of Agriculture. Poultry - Production and Value, 2010 Summary; 2001. Available at:http://usda.mannlib.cornell.edu/usda/current/PoulProdVa/PoulProdVa-04-28-2011.pdf. [accessed November 4, 2011].

VanHeugten E, VanKempen T. Growth performance, carcass characteristics, nutrient digestibility and fecal odorous compounds in growing-finishing pigs fed diets containing hydrolyzed feather meal. J Anim Sci 2002;80:171–8.

Wang P, Zhao G, Tian J, Su X. High-Performance Liquid Chromatography—Inductively Coupled Plasma Mass Spectrometry Based Method for the Determination of Organic Arsenic Feed Additives and Speciation of Anionic Arsenics in Animal Feed. J Agric Food Chem 2010;58:5263–70.

Washington State Department of Agriculture. Fertilizer Product Database. 2011, Olympia, Washington; 2011. Available at:http://agr.wa.gov/PestFert/Fertilizers/ProductDatabase.aspx [accessed November 4, 2011].

Wasserman GA, Liu X, Parvez F, Ahsan H, Factor-Litvak P, Kline J, et al. Water arsenic exposure and intellectual function in 6-year-old children in Araihazar, Bangladesh. Environ Health Perspect 2007;115:285–9.

Wu MM, Kuo TL, Hwang YH, Chen CJ. Dose–response relation between arsenic concentration in well water and mortality from cancers and vascular diseases. Am J Epidemiol 1989;130:1123–32.

Yu Y. Replacement of fish meal with poultry by-product meal and hydrolyzed feather meal in feeds for finfish. In: Lim G, Webster CD, Lee C-S, editors. Ch3. Book title. Alternative Protein Sources in Aquaculture Diets. New York: The Hawthore Press; 2008, p. 51–93.

Zhao F-J, McGrath SP, Meharg AA. Arsenic as a Food Chain Contaminant: Mechanisms of Plant Uptake and Metabolism and Mitigation Strategies. Annu Rev Plant Biol 2010;61:535–59.