How Best Can We Utilize Our Solid Tannery Waste?

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Abstract. The enormous amount of solid tannery waste that is generated in Bangladesh has been being used as the major component of poultry/fish feed since long. Recent studies on the presence of chromium in solid waste, in poultry/fish feed, and even in chicken, eggs and fish suggest that in the interest of public health solid tannery waste should be avoided while producing poultry/fish feed. The present paper examines the toxicity of Cr(III) and Cr(VI) separately and their maximum allowable dose levels. Two approaches have been considered for using solid tannery waste in poultry/fish feed. In the first one, the tanned part of the waste should undergo elaborate chemical treatment to reduce the amount of chromium to a minimum. In the other approach a proposal has been made to decrease the formation of highly toxic Cr(VI) by taking proper steps at different stages of production of leather and feed. It is assumed that the amount of Cr(III) that enters chicken, eggs and fish through the feed would cause no health risk to consumers. The second approach may be appropriate for Bangladesh. The dumping of solid waste in landfills is not practicable and not in conformity with the present-day idea of industrial symbiosis.
Keywords: Public Health, Solid Waste, Tannery, Chromium Toxicity, Animal feed.

1. Introduction

The leather industry is one of the biggest in the world, with an estimated global trade value of more than US$ 100 billion per year (UNIDO, 2010). However, this industry is also considered one of the most polluting industries in the world (WorstPolluted.org, 2016). There is considerable use of chemicals and water in the conversion of hides into finished leather product and this generates a large amount of solid and liquid waste (Paul *et al*., 2013). The amount of solid waste that is generated from tannery processes all over the world is estimated as 6 million tons per year (Rajamani, 2010). The developing world has been the source of more than half of the world supply of leather raw material since 2010 (UNIDO, 2010). The leather industry in Bangladesh already occupies the second position in the export sector and there are around 270 tanneries in Bangladesh - 90% of which are located in the Hazaribagh area of the capital, Dhaka (WorstPolluted.org, 2016). It is estimated that more than 20,000 m^3 of effluent is generated every day in Hazaribagh and the untreated effluent is discharged to a channel leading to the Buriganga River, the main river of Dhaka. Part of the river closest to the effluent discharge points has virtually turned into sewage and it is no wonder that the Blacksmith Institute branded Hazaribagh as 5th among the top ten toxic threats in 2013 (WorstPolluted.org, 2016). Very few of the tannery industries in Hazaribagh operate effluent treatment plants because of the high costs associated with the installation and maintenance of suitable effluent treatment plants. The Government of Bangladesh has recently started to force the tanneries to shift to a different location where a central effluent treatment plant would be set up for treating the liquid waste before discharge. This is similar to the strategy adopted in countries like Italy where tanneries can only be established in organized industrial districts (Lofrano *et al*., 2013).

2. Management of solid tannery waste

Price and Joseph have suggested five strategies for the reduction of the amount of any waste generated from industry (Price and Joseph, 2000). They are, in decreasing order of preference - waste minimization, material reuse, material recycling, energy recovery and waste disposal. It is clear from this list that the dumping of waste in a landfill is the least desirable option for waste management, although this happens to be the most widely used practice. Not the least utilization of solid waste is achieved in this method. The other methods such as thermal incineration, anerobic digestion, bioremediation, vermicomposting, etc have their advantages and disadvantages and are mostly concerned with the organic part of the waste (Sekaran *et al*., 2007).

In a country like Bangladesh, the huge amount of solid tannery waste has been traditionally disposed off through landfills. Most of these landfills were not constructed properly and therefore toxic metals, especially chromium, have leached out into the adjacent soil making it unfit for cultivation and other uses. Poultry and fish feed manufacturers have discovered the potential of these wastes to be the principal component of animal feed because of their protein content (Hossain *et al*., 2007). Solid tannery waste contains untanned components like raw trimmings and fleshings, which are mostly protein contaminated with some salts (Lofrano *et al*., 2013). The tanned component of the solid tannery waste comprises of splits, shavings and crusts and they contain a significant amount of inorganic materials in addition to the proteinous organic compounds. To use the solid waste in poultry feed, fish feed and fertilizers, both the groups of components are boiled for a few hours and then sun-dried. The dried material is then ground, producing what is called protein-concentrate (Hossain *et al*., 2007). When the idea of using this protein-concentrate as animal feed was put into practice, the result was highly encouraging, as the chicks grew quite rapidly with these feeds (Paul *et al*., 2013). This is a perfect example of industrial symbiosis, an association between companies in which the wastes or byproducts of one become the raw materials for another (Chertow, 2000). The tannery owners could now earn money by selling their wastes instead of spending money on land filling (Alam, 2010). Owners could sell around 5 tons of tannery waste every day and the demand for this tannery waste is so high that a feed factory is ready to make an advance payment to ensure its annual supply of this proteinous waste . However, the question of the toxicity of this waste and possible transfer to common food items like chicken, eggs and fish arose quite strongly (Hossain *et al*., 2007, Bari *et al*., 2015, Hossain and Hasan, 2014, Hossain *et al*., 2009, Mazumder *et al*., 2013, Parvin and Rahman, 2014). The heavy metal concentrations of 18 samples of solid tannery waste at Hazaribagh collected from different spots and from different stages of production indicated that chromium was present in all the solid tannery waste samples in the highest quantity (a maximum of 3.2% of the solid tannery waste) followed by Cd, Pb, As and Hg in trace amounts in some samples (Hossain *et al*., 2007). Bari et. al. measured the chromium content in different body parts of chickens and the values range from less than 0.10 to 2.44 ppm indicating that the chromium is entering into the meat of the chicken, possibly because of the chromium in the feed. Another study was done where the chickens were fed commercially available poultry feed mixed with leather shaving dust and skin cut waste for a 1 or 2 month period (Hossain and Hasan, 2014). The concentrations of chromium found in different body parts of such chickens were similar to those obtained by Bari *et al*. . Total amount of chromium has also been measured in different egg samples and the chromium content in albumen and yolk varies from 1.05 to 2.72 ppm (Hossain *et al*., 2009). It is important to note that all of these studies emphasized on the measurement of total chromium rather than in the form of Cr(III) or Cr (VI) which we feel is a serious drawback of these studies. If these amounts of chromium were assumed to be in the Cr(III) form, would these pose any health threat? To find the answer we have to consider the toxicity of Cr(III) and Cr(VI).

3. Toxicity of chromium

Chromium exists in several oxidation states, the most common and stable forms in nature are Cr(III) and Cr(VI). As is well known, trivalent chromium in the form of chromium sulfate is used for chrome tanning. Let us first examine the usefulness and toxicity of trivalent chromium. Cr(III) in trace amounts is useful for human health. It forms an organic complex called Glucose Tolerance Factor (GTF) that interacts with the pancreatic hormone, insulin, and regulates the uptake of glucose by cells (Barrett *et al*., 1985). It is essential for lipid, protein and fat metabolism in animals and humans. Chromium deficiency may be responsible for maturity-onset diabetes, cardiovascular diseases and nervous system disorders (Grevatt, 1998). According to the National Research Council (NRC) of USA, the safe and adequate daily dietary intake (ESADDI) for chromium is 50-200 μg/day (Allowances *et al*., 1980). The Food and Drug Administration (FDA) has selected a Reference Daily Intake of 120 μg/day, which is within the above range (FDA, 1995). Recently a Panel on Food Additives and Nutrient Sources constituted by the European Food Safety Authority has recommended a maximum Cr(III) intake of 250 μg/day (Additives, 2010) in agreement with the WHO recommendation (United Nations Environment Programme *et al*., 2015). The panel has considered the available literature on Cr(III) and come to the conclusion that it is not possible at the moment to decide on the Tolerable Upper Intake Level (UL) of Cr(III). The UK Expert Group on Vitamins and Minerals (EVM) is also in agreement with the US Food and Nutrition Board that overall there are insufficient data from human and animal studies to derive a safe upper level for chromium. However, EVM opines that about 0.15mg/kg bw/day equivalent to 10 mg/person should have no adverse health effects (Additives, 2010). In an attempt to establish an Oral Reference Dose (RfD) which is an estimate of a daily exposure to the human population that is likely to be without an appreciable risk of deleterious effects during a lifetime, an expert group selected by EPA reviewed the existing literature on the subject (Grevatt, 1998). They found the work of Ivankovic and Preussman (Ivankovic and Preussmann, 1975) most appropriate for development of the RfD. The reviewers determined the NOAEL (No-Observed-Adverse-Effect-Level) based on this study, which turned out to be 1468 mg of Cr(III) per kg of body weight per day. A 100-fold uncertainty factor and a 10-fold modifying factor were introduced to get the reference dose of 1.5 mg per kg of body weight per day (Grevatt, 1998). Nocarcinogenic effect has been observed with Cr(III) in any study (Additives, 2010).

Unlike Cr(III), Cr(VI) has neither any usefulness nor can it be tolerated nearly at as high levels as Cr(III). Rather, its toxicity is some 500-1000 times higher than that of Cr(III) and it is capable of causing mutation, cancer and cell damage (Costa, 1997). Its extreme toxicity is attributed to its easy penetrating ability into cells in the form of negative ions (chromate, dichromate) replacing phosphate and sulfate anions (De Flora *et al*., 1997). Once inside the cell Cr(VI) undergoes step-by-step reduction to Cr(III). It is these unstable intermediate species such as Cr(V) and Cr(IV) that are supposed to be responsible for causing oxidative damage of lipids and proteins, mutation and cell transformation leading to cancer (Das and Singh, 2011). That Cr(VI) is a human carcinogen has been determined by the National Toxicology Program (NTP), the International Agency for Research on Cancer (IARC), the UK Health Protection Agency, and Office of Environmental Health Hazard Assessment (OEHHA) (Program *et al*., 2011, Smoke and Smoking, 2004, Assem and Zhu, 2007).

Perhaps the comparison of the toxicity of Cr(III) and Cr(VI) would remain incomplete without the mention of the work of Kerger *et al* (Kerger *et al*., 1996) who had four adult humans ingested with a single dose of 5 mg chromium in 0.5 L de-ionized water in three chromium mixtures: (i) Cr(III) chloride, (II) potassium dichromate reduced with orange juice and (III) potassium dichromate.

$CrCl_3$ was poorly absorbed (estimated 0.13% bioavailability) and rapidly eliminated in urine (excretion half-life, about 10hr) contrary to potassium dichromate which had the bioavailability of 6.9% and the half-life, about 39 hr. This means that Cr (VI) has more than 50 times the bioavailability of Cr (III) and that its half- life is around 4 times longer. If we calculate the average life-time of Cr (VI), it turns out to be 56 hr, about 2 $^{1}/_{3}$ days. Thus if a chicken is on feeds containing Cr (VI) regularly, there would be bioaccumulation of Cr (VI), as has been found in our study (Parvin and Rahman, 2014). The office of Environmental Health Hazard Assessment (OEHHA) (OEHHA, 2010) has derived the value of 8.2 µg/day for the Maximum Allowable Dose Level (MADL) for hexavalent chromium by the oral route of exposure. Amongst many studies the office chose the work of Murthy *et al* (Murthy *et al*., 1996) that found a NOEL (No observable Effect Level) value of 0.142 mg/kg bw/day for female reproductive toxicity in mice. This value was multiplied by 58 to make it applicable for a 58 kg woman and then divided by 1000 to be on the safe side.

Let us examine in some more detail whether the amounts of total chromium (assumed to be all in +3 state) found in different body parts of chicken fed on tannery-based poultry feed do pose any real threat to public health. The amount of chromium is different for different body parts. Usually it is the maximum in liver. Bari *et al* have measured chromium content in liver, gizzard, meat and skin of three varieties of chicken – broiler, native and free-ranging (Bari *et al*., 2015). Average minimum and maximum values (in mg/kg) were 0.202 & 0.780 for broiler, 0.138 & 0.440 for native and 0.111 & 1.400 for free-ranging chickens. If we take the average of the maximum values only, it turns out to be 0.873 mg/kg. A careful examination of the safe dose levels given by different groups for Cr(III) would reveal that consumers are quite safe even if they ingest as much as 1 kg of chicken per day according to the values given in references, (Grevatt, 1998) & (Additives, 2010). We presume that the low values recommended by other authors are meant for adequate dietary intake, as Cr(III) is essential for health. Even then, with the value of 250 µg/day as recommended by the European Food Safety Authority, a person should have no problem if he does not consume more than 285 g of chicken per day. In view of the fact that chicken meat which constitutes the maximum proportion of edible chicken contains less than this amount (0.873 mg) of chromium, it can be reasonably concluded that consumption of chicken containing only Cr(III) should not be of particular concern to public health.

Let us now turn our attention to Cr(VI).Unfortunately not much work has been done on the presence of Cr(VI) in food items of Bangladesh. By using spectrophotometric method involving diphenylcarbazide, Mazumder et. al. has recently looked at Cr(VI) in the protein concentrate and in chickens of Dhaka, Bangladesh and the maximum value of chromium in the chicken was 177 µg/kg in the liver (Mazumder *et al*., 2013). This work was further extended by Parvin and Rahman who collected chickens from almost all over the country and analyzed different body parts such as liver, gizzard, flesh and brain (Parvin and Rahman, 2014). Like Mazumder *et al* they also used the spectrophotometric method for the analysis of most of the samples. In order to be sure about the reliability of the method some samples were run through ion chromatography. Cr(VI) was found in albumen and yolk of eggs by both the methods. Extracellular fluids are believed to reduce most of the Cr(VI) to Cr(III) before they reach the site of absorption (Kerger *et al*., 1996). The small remnants of Cr(VI) may perhaps bioaccumulate otherwise high amounts would not have been obtained for older chickens (Parvin and Rahman, 2014).

Following the publications of Islam *et al*, (2007), Hossain *et al*, (2007 & 2009) and Hossain & Hasan (2014), local newspapers of Bangladesh made headlines about the ill effects of the consumption of chicken containing large amount of chromium that has come through chrome tanning (Alam, 2010). The newspapers recommended not using tannery waste at all for the manufacture of feeds and in fact the law-enforcing authorities went to the extent of destroying some of these feed factories. Recently, the Bangladesh Supreme Court has banned the use of solid tannery waste in the production of poultry feed.

4. Management of chromium in animal feed

Generally, 1 kg of poultry feed is composed of 600 g of meat bones and 400 g of other ingredients like soya oil cake, ground rice and dry fish (Alam *et al*., 2010). This 400 g of other ingredients should contain little or no chromium. The meat bone part can be divided into untanned and tanned waste. The untanned waste comprising of raw trimmings and fleshings, which constitutes 66% of the solid waste should again contain little or no chromium. It is this 34% of the solid waste (around 20% of the poultry feed), composed of split, shavings and crust of finished leather that contains chromium if the leather is made through chrome tanning. In order to be able to use solid tannery waste for the production of animal feed that would not cause harm to human health, the amount of chromium especially the hexavalent form should be present in the feed in as little quantity as possible. Chrome tanning being the most popular form of tanning, it would be almost impossible to completely get rid of chromium. Paul et. al. (Paul *et al*., 2013) have advocated the removal of chromium altogether from the solid waste before using it in the poultry feed. An oxidation method was used to achieve 95% removal of chromium from tannery solid waste and after further thermal and enzymatic treatment, the amount of Cr in the feed was low enough to be used. Whereas this is safer from the point of view of public health, the cost of production of poultry feed would go up significantly if their elaborate procedure for dechroming is followed. One way of reducing chromium would be to mix chrome leather with vegetable-tanned leather. Another approach to the problem is to change the ratio of different components in the feed so that chromium-containing leather part is at a minimum. The maximum allowable level for hexavalent chromium, as mentioned above is 8.2 µg/day, which is more than 1000 times lower than the safe level of Cr(III) suggested by Expert Group on Vitamins and Minerals. Thus from the point of view of public health, what is more important is the amount of hexavalent chromium present in feeds rather than trivalent chromium. To minimize the amount of Cr(VI) in the feed, one should look into the possible sources of Cr(VI). Amongst the direct sources, Cr(VI) contamination in Cr(III) tanning agent, certain class of metal complex dyes, and inorganic pigments based on lead chromate may be mentioned (Devikavathi *et al*., 2014) . Apart from these direct sources which can be avoided, there are steps in the tanning process itself where Cr(III) is oxidized to Cr(VI). Devikavathi et. al. have used antioxidants like vitamin C and other natural biochemicals to arrest the conversion of Cr(III) to Cr(VI) at different stages of chrome tanning process. They claim that by using a combination of antioxidants they have been able to obtain leather almost free from Cr(VI). Solid waste is boiled and then dried in the sun before using it as a component of poultry/fish feed. Boiling favors oxidation of Cr(III) to Cr(VI) and ultraviolet radiation from the sun is quite effective in causing the oxidation (Başaran *et al*., 2008). It is not surprising, therefore, that Mazumder et. al. have found more Cr(VI) in the boiled and dried solid waste than in the original solid waste. We believe it is possible to control the formation of Cr(VI) in these steps too. Boiling and drying which are essential for preventing putrefaction can be limited to untanned part (fleshings and trimmings) alone. Because this part is chromium free, the question of oxidation does not arise. The tanned part has already been made durable through tanning, so no further boiling is necessary. This would save both time and money. The two parts can be ground together to be used in the animal feed. It has been found that storage conditions also promote oxidation through formation of free radicals (Devikavathi *et al*., 2014). Solid tannery waste should thus be used as fresh as possible. By adopting these measures it should be possible to control the formation of Cr(VI) to a great extent so that the poultry feed and the poultry become no more concern for public health. Naturally the adoption of this management approach would push the cost of poultry feed up a bit but not as much as the other methods.

5. Conclusion

Land filling the enormous solid tannery waste that is generated by the tannery industry is not an acceptable option. The high protein content of this waste should somehow be utilized. Two alternative approaches have been discussed here. The alternative authored by Paul et. al. proposes to get rid of total chromium from the solid waste and the other, by the present authors, puts more emphasis on the prevention of formation of Cr(VI) taking effective measures at different stages of production of leather and poultry feeds, assuming that the amount of Cr(III) present would be harmless. More research is needed to find out which alternative is more convenient and less expensive. In either case the price of poultry feed will go up with concomitant increase in the price of chicken and eggs. Cooperation of all the stake holders including tannery owners, feed producers, poultry farmers and consumers will be needed in the greater interest of survival and flourishing of our tannery industries. This approach of utilizing the solid tannery waste is also in conformity with the modern concept of industrial symbiosis (Bain *et al*., 2010).

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