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Wholegrains and health: many benefits but do contaminants pose any risk?

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Running title: The safety of wholegrains in our diet

Key Words: wholegrain; contaminant; mycotoxin; heavy metal; health

Abstract

Since prehistoric times, humans have consumed grains as part of their diet. Being rich in carbohydrates, grains typically form a key part of all food-based dietary guidelines, with wholegrains recommended as healthy dietary choices. However, grains are also naturally exposed to contaminants and can be one of the main dietary sources of food borne contaminants by virtue of their frequent consumption. Recent scientific reports positively highlight wholegrains as foods with inherent health properties, which, because they provide fibre and micronutrients such as B vitamins and zinc, can improve the quality of carbohydrate intake as part of sustainable healthy diets. This article describes potential protective properties inherent to wholegrains and contends that the presence of contaminants in wholegrains merits continued monitoring but that any such risk does not outweigh the known benefits of wholegrain consumption.

Key Words: wholegrain; contaminant; mycotoxin; dietary exposure; fibre; health

What are grains and wholegrains?

Cereal grains are food staples and represent the primary carbohydrate source in the human diet worldwide. Today, three grains (rice, maize and wheat) provide about 60% of the world's food energy (FAO 2018a), with the Food and Agriculture Organization (FAO) forecasting that world grain utilisation in 2018/2019 will reach a record level of 2646 million tonnes (FAO 2018b). For wholegrains specifically, several definitions have been suggested (Van der Kamp *et al.* 2014; AACC International, 2018), all of which principally agree that: 'wholegrains consist of the intact, ground, cracked or flaked caryopsis (kernel) after the removal of inedible parts such as the hull and husk. The principal anatomical components—the starchy endosperm, germ and bran—are present in the same relative proportions as they exist in the intact kernel'. The definition by the Healthgrain Forum acknowledges that some parts of the grain, such as the outermost layers, are removed during processing to eliminate potential contamination of the outer bran (Van der Kamp *et al.* 2014).

The most commonly consumed wholegrains in Western Europe are wheat, oats, rye and barley, with wholegrain products including porridge, breakfast cereals, breads, whole wheat pasta and snacks such as oatcakes, popcorn and wholegrain cereal bars (BDA 2016; CPW 2017). Compositionally, wholegrains are nutrient dense providing fibre as well as B vitamins, vitamin E, magnesium, zinc, iron, copper, selenium and other bioactive compounds (EUFIC 2015; BDA 2016). Nevertheless, dietary intakes of wholegrains are typically low, with 18% of UK adults and 15% of UK children and adolescents not consuming any wholegrains in the most recent analysis of wholegrain intakes in the *National Diet and Nutrition Survey* (2008-2011) and only 17% of adult and 6% of children achieving the US dietary

recommendation of 48 g/day wholegrain (Mann *et al.* 2015). This is similar to that reported in other European countries (Bellisle *et al.* 2014; Sette *et al.* 2017; O'Donovan *et al.* 2018), notable exceptions including Denmark where wholegrain intakes have increased to up to 63 g/day as a results of a multi-stakeholder, national campaign (Greve & Ness 2014; Ness *et al.* 2014). Consumer insights suggest such low intakes may be due to lack of awareness of the benefits of wholegrain and how much should be consumed (CPW, 2017), in addition to negative perceptions in relation to their cost, ease of use and flavour (Kuznesof *et al.* 2012; McMackin *et al.* 2013). The latter perhaps routed in the Nineteenth-century industrialisation when new processing techniques, such as rolling mills, more effectively removed the husks of wheat and rice resulting in 'white flour', which was more popular with consumers. Increasing awareness of the health benefits of wholegrains coupled with growing consumer interest in healthier food products in recent years has resulted in food manufacturers including more wholegrains in cereal products, globally (Seal & Thielecke 2018)

Health benefits of consuming wholegrains

High consumption of wholegrains in the human diet has been associated with decreased risk of developing several chronic diseases (Reynolds *et al.* 2019), such as cardiovascular disease (Wu *et al.* 2015), type 2 diabetes (Ye *et al.* 2012) and possibly obesity (Albertson *et al.* 2016) and cancer (Aune *et al.* 2011). Although the exact mechanisms by which wholegrains may exert such a protective effect remain to be elucidated, the generation of betainized compounds in vivo following the consumption of wholegrain-rich diets appears to be protective for glucose metabolism at least (Kärkkäinen *et al.* 2018). Overall, there is a growing body of evidence indicating that people who consume more wholegrains have a lower risk of some chronic diseases compared with people who include few wholegrains in their diet (Wu *et al.* 2015; Benisi-Kohansal *et al.* 2016; Zong *et al.* 2016). The Global Burden of Disease study (2015) estimated that low wholegrain intake resulted in almost 4 million Disability Adjusted Life Years (DALYs) in the European Union (EU) in 2015, and almost 270 000 avoidable deaths from all causes (of which 250 000 were from cardiovascular disease) (GBD, 2018). The growing evidence base on the health benefits of wholegrains has led governmental authorities and scientific organisations to issue specific recommendations for wholegrains, advising that they are consumed in place of foods that are made from refined grains (Seal *et al.* 2016; SACN 2015), for example in the UK wholegrains are mentioned as part of food based dietary guidelines but as yet no quantitative guidelines exist (Seal *et al.* 2015). Practical guidance exists for consumers in the form of on-pack product health claims in the US (FDA 2018) and suggestions for labelling or identifying wholegrain foods including use of logos by expert organisations such as the Healthgrain Forum in Europe (Ross *et al.* 2017) and the Whole Grains Council in the US (Whole Grain Stamp 2005).

The mechanisms underlying the health benefits of wholegrains are not fully understood but may relate to their fibre content and its influence on short chain fatty acid production and the gut microbiome, and/or the presence of bioactive compounds (*e.g.* vitamins, minerals, lignans and phenolics) (Robinson & Chambers). Whilst the influence of a food structure/matrix on health outcomes is increasingly recognised for dairy products (Thorning *et al.* 2017), this is less well characterised for wholegrains (Thielecke & Nugent 2018).

Contaminants in wholegrains

While grains are important carbohydrate sources, they naturally contain contaminants and, as a food category, represent one of the main dietary sources of foodborne contaminants (see review by Thielecke & Nugent, 2018). Food contaminants have been defined as ‘any substance not intentionally added to food which is present in such food as a result of the production, manufacture, processing, preparation, treatment, packing, packaging, transport or holding of such food or as a result of environmental contamination’ (EC 1993). The potential sources of contaminants in grains are mostly environmental and include air, dust, soil, water, insects, rodents, birds, animals, microbes, humans, storage and shipping containers, and handling and processing equipment. Most contamination is of a microbiological nature but heavy metals and process contamination (*e.g.* acrylamide) can also occur. Pesticide residues may also be present, with recent sampling reports from the European Food Safety Authority (EFSA) suggesting that any pesticide residues present are well within the limits permitted in EU legislation (EFSA, 2016). Pesticide residues are not discussed further in this article, with the focus on mycotoxins, heavy metals and acrylamide.

Mycotoxins

Mycotoxins are poisonous metabolites produced by certain species of fungi, which can grow on grains. Such fungi may be airborne or soil borne and can infect plants in the field or throughout the production chain (Scudamore & Patel 2009; Alshannaq & Yu 2017). Early recordings of poisoning by mycotoxins date back to Assyria in 600 BC (Bennett & Bentley 1999). Globally, occurrence of mycotoxins in grains remains a significant concern to human health, particularly in developing countries (Lee & Ryu 2017; Moretti *et al.* 2017). Five groups of mycotoxins which can affect human health affect grains, namely deoxynivalenol /nivalenol (DON), zearalenone, ochratoxin, fumonisins and aflatoxins. Table 1 lists examples of the grains affected by these mycotoxins and their potential impact on human health. Maize, wheat and barley are primarily affected by all mycotoxins, with adverse effects in humans including nausea and fever, and potentially cancer (Freire & da Rocha 2016)).

Levels of mycotoxins tend to be lower in processed foods than wholegrains; however, incidence rates can vary depending on the individual mycotoxin and on selection processes and protocols followed by food manufacturers (Thielecke & Nugent 2018). Ten-year global occurrence data reveal a consistent presence of mycotoxins in the food supply, with occurrence of samples positive for the individual mycotoxins and the maximum levels in raw grains of 55% and 1642 µg/kg for aflatoxins, 29% and 1164 µg/kg for ochratoxin A, 61% and 71 121 µg/kg for fumonisins, 58% and 41, 157 µg/kg for deoxynivalenol, and 46% and 3049 µg/kg for zearalenone (Lee & Ryu 2017). Further, while climate change and associated changes in humidity, temperature and water availability is suggested to have a direct influence on presence of mycotoxins, no clear consensus exists. Whilst some climate change modelling indicates no influence on DON contamination of wheat and corn in Europe (Van der Fels-Klerx *et al.* 2013), this is in contrast to other modelling of DON contamination in North West Europe (Marvin 2012), and for aflatoxin in maize (Battilani *et al.* 2016; Moretti *et al.* 2019) and *F. graminearum* in Central and North Europe (Moretti *et al.* 2019).

Metals as contaminants

Heavy metals such as arsenic, cadmium and lead are naturally found in the earth and most relevant with respect to grain intake (Thielecke & Nugent 2018). When ingested, such metals can compete with minerals such as calcium, magnesium and iron for absorption but can also bind to cellular components (*e.g.* structural proteins, enzymes and nucleic acids) disrupting their function (Andrade *et al.* 2017; Kosek-Hoehne *et al.* 2017). Chronic exposure to heavy metals leads to wide-ranging health problems affecting the immune system, nervous system and systems relating to reproductive, metabolic and skeletal health (Tchounwou *et al.* 2012). Grains such as wheat and rice are the major contributors to dietary cadmium intake, while rice contributes to dietary arsenic intake (EFSA 2009, EFSA 2012, EFSA 2014).

The storage sites within grain for cadmium and arsenic are different. Whereas cadmium is primarily stored in the endosperm (the endosperm being the main source of white flour) arsenic accumulates mostly in the outer layer of rice. Hence, wholegrain rice with its bran intact can have up to 80% more arsenic than white rice (Greenerchoices 2019). Nevertheless, rigorous safety assessments by EFSA have concluded that rice is safe to eat by all population groups as part of food-based dietary guidelines, including infants and young children (EFSA 2009; EFSA 2014). Results of ongoing pan-EU monitoring programmes during the period 2017-2018 of the arsenic content of a wide variety of foodstuffs, including grains, will inform the scientific discussion as to whether further safeguards are needed (EC, 2019).

Acrylamide

Acrylamide is a chemical that naturally forms in starchy foods when they are cooked at high temperatures such as through roasting, frying or baking, with the general rule of thumb that the browner the food after cooking, the more acrylamide will be present. Grain products, such as bread and breakfast cereals, including wholegrain versions, can also contain acrylamide, level of which will be affected by baking and toasting. . Acrylamide has been classed as potentially carcinogenic (IARC 1994) with the EU Commission updating benchmark values for acrylamide levels in various food categories in 2017 (EC 2017) and proposing levels should be as low as reasonably achievable (ALARA principle).

Human dietary exposure to contaminants in grains

Using data from the UK *National Diet and Nutrition Survey*, the relative (%) contribution of grain (cereal) and grain products to energy and macronutrient intakes can be viewed as considerably greater than for meat, milk, or fruit and vegetables (see Table 2). For example, cereal and cereal products contribute 2-3.5% more to total energy intakes in the total population than each of the other food groups listed. Hence, while grain-based foods can contain low levels of contaminants because they are consumed in significant quantities by large sections of the population, they can make significant contributions to dietary contaminant exposure assessments. This contributions by grains is reflected in recent EFSA Opinions for metals such as cadmium (EFSA, 2012) and arsenic (EFSA 2009, 2014). To refine risk assessments and ensure accurate estimates of contaminant intake there has been calls by EFSA for occurrence data on the presence of acrylamide, mycotoxins and heavy metals (EFSA 2018). Further refinements of mycotoxin intake may also need to account the possibility also exists for carry-over of contaminants in grains fed to animals as part of 'rations' or concentrates' to foods of animal origin which are then consumed by humanse.g. the mycotoxin, aflatoxin B1 in dairy cattle feed which can be metabolised to aflatoxin M1 and subsequently occur in milk (Veldman *et al.* 1992). For the assessment of contaminants in heavy metals, difficulty also arises from the fact that bioaccumulation of heavy metals differs between plant species and depends on concentrations of metals present in the soil (Khan *et al.* 2015). Overall, in Europe, exposure to contaminants in wholegrains remain low but there is a need for continued monitoring, particularly in areas of increased risk such as developing countries (Thielecke & Nugent 2018). This is true not only for grains but all foods.

Strategies to mitigate risk of contaminants to human health

There are a number of steps in the food chain which can influence exposure to contaminants in grains. Considered here is the influence of regulators, farming or production practices, processing and manufacturing, as well as the actions of consumers.

Role of farming and production sectors

Farming and production practices can directly influence the presence or absence of contaminants within grains, with different influences for each type of contaminant. For example, the temperature, grain moisture content and relative humidity of grain storage conditions are critical factors favouring fungal growth and subsequent mycotoxin production, as is grain damage from pests or birds in the field (FAO; WHO 2003). Whereas, the presence and concentration of heavy metals in grains is directly influenced by those in the soil. For the process contaminant acrylamide, the presence of the amino acid asparagine within the grain is a critical component (Curtis & Halford 2016). Hence, multiple strategies are needed to ensure the presence of contaminants are minimised within the final grain product. Examples include following best practice guidelines for storage conditions, minimising insect and bird damage, following appropriate crop rotation, appropriate fertilisation and/or crop selection and milling (for review see Thielecke & Nugent 2018).

Does post-harvest treatment work?

There are some suggested methods for treating or removing contaminants in grains (*e.g.* use of gases such as ammonia, chlorine dioxide, sulphur dioxide or ozone to treat mycotoxin-contaminated grains). However, as yet such approaches are restricted in their reach and efficacy and each with limitations for use and currently prevention remains a more efficient method than treatment (Karlovsky *et al.* 2016).

Do grains themselves provide protection?

There is some evidence that the nutritional composition of wholegrains may in itself provide protection. Wholegrains are more nutrient dense than refined grains as the bran and germ present in wholegrains contains much of the fibre and micronutrients (Papanikolaou & Fulgoni 2017; Robinson

& Chambers 2018; Thielecke & Nugent 2018). Refining removes the bran and germ, and up to 80% of minerals, such as iron and zinc, leaving only the endosperm (which contains mostly carbohydrate, protein and small amounts of some B vitamins and minerals). There is some evidence to suggest that the combination of nutrients present in wholegrains can help mitigate against the negative effects of contaminants with scope for further research in this area (Thielecke & Nugent 2018). For example, the presence of vitamins, carotenoids and antioxidants (*e.g.* ferulic acid in wholegrain wheat) has the potential to reduce the impact of mycotoxins by protecting cell membranes from mycotoxin-induced damage (Atroshi *et al.* 2002; Gross-Steinmeyer & Eaton 2012).. Another potential strategies to mitigate the effects of mycotoxins includes the binding of probiotic micro-organisms to mycotoxins to decrease their gastrointestinal absorption (Thielecke & Nugent 2018). This has been described in milk (Wochner *et al.* 2018) and there is some limited evidence for kefir grains (Taheur *et al.* 2017), corn, sorghum and rice (Kim *et al.* 2017). Further research is needed to explore the potential of probiotics to influence the adsorption of mycotoxins.

For toxic metals, the primary line of defence is at the site of absorption. As with essential minerals, toxic metals must bind with transporters in the small intestines prior to absorption, with empirical data suggesting optimal levels of nutrients such as magnesium, zinc, selenium, calcium, iron and dietary fibre may reduce toxicity (Elsenhans *et al.* 1991). This may imply that if consumption of minerals is optimal, it might mitigate against the toxicity of heavy metals (Thielecke & Nugent 2018). There is some evidence from cellular and animal studies for a protective effect of selenium, magnesium, calcium, zinc, iron and dietary fibres in the absorption of heavy metals but more research is needed to understand the role of these bioactives when present naturally within a grain matrix (Thielecke & Nugent, 2018). Evidence for supporting mechanisms of action is limited but for cadmium absorption at least may involve the metal binding proteins metallothioneine and/or glutathione (Grosicki *et al.* 2015; Shabb *et al.* 2017). Unsurprisingly, evidence from human trials is limited due to ethical reasons. However, there is epidemiological evidence suggesting that iron deficiency can lead to increases in lead absorption (Kordas 2010; Wright *et al.* 2003) and an inverse relationship has been noted between calcium deficiency and blood lead levels (Mahaffey *et al.* 1986). It has been suggested that people eating a diet deficient in micronutrients will be predisposed to toxicity from nonessential (heavy) metals (Peraz *et al.* 1998). So, while there is limited evidence to suggest that the presence of micronutrients in wholegrains may help mitigate the risk of heavy metal contamination, there is comfort in the lack of evidence to suggest that consuming wholegrains increases any such risk (Thielecke & Nugent 2018).

Role of the regulator

Regulatory agencies both globally, regionally and nationally have clear strategies in place to ensure that levels of contaminants in food or feed are as low as reasonably possible through Good Agricultural Practice and Good Manufacturing Practice (EC 1993; FAO/WHO 1995), codes of practice and risk assessment and risk management strategies. Since many contaminants are naturally occurring it would be impossible to impose a total ban on their presence; hence most Regulatory Agencies have developed, and subsequently monitor and enforce, maximum acceptable levels of contaminants in foods that are scientifically based and technologically practical (*e.g.* for mycotoxins) (EC 2006). Within Europe, EFSA provides scientific advice and guidance on suitable maximal levels of contaminants but it is the responsibility of individual Member States to ensure compliance, with responsibility for imported foodstuffs resting with the country of origin. The US Food and Drug Administration and Food Inspection Agency in Canada hold similar roles to EFSA in the EU. While there are some differences in approaches and regulatory limits applied between jurisdictions, the public can be reassured by the rigorous monitoring and enforcement strategies that are in place which ensure the highest food safety standards and protection to public health.

What about the consumer?

, There are several ways consumers can reduce the potential risk to health of contaminants in wholegrain foods. First by following basic good hygiene and correct food storage practices. For example, adhering to 'use by' dates to avoid consumption of bread on which fungal moulds have grown, or avoiding over-toasting bread or wholegrain products to minimise exposure to acrylamide. Such steps are of course more difficult when individuals or populations are experiencing food poverty and/or food insecurity and especially so where presence of contaminants (particularly mycotoxins) is less routinely monitored than in Western Societies. Whilst opting for processed foods might protect as contaminants tend to be lower in these foods, this is not always the case (*e.g.* cadmium levels are higher in refined than unrefined flour) (Thielecke & Nugent 2018) and it is well described that processed foods can be higher in less desirable nutrients from a public health perspective (*e.g.* salt, sugar and/or fat). and wholegrains are recommended as part of a healthy, balanced diet. Rinsing and cooking can reduce arsenic levels in rice, with cooking in excess water suggested as helpful at reducing arsenic content (Raab *et al.* 2009). However, this can also result in losses of nutrients such as iron, folate, niacin and thiamine, but more so for polished and parboiled varieties than for wholegrain brown rice (Gray *et al.* 2016). Hence, consumers may wish to consider both the variety of rice and cooking methods if attempting to reduce arsenic concentrations in rice (BNF 2018).

Currently, consumers are advised to consume wholegrains as part of food-based dietary guidelines. For example, the UK Eatwell Guide states '*Choose wholegrain or higher fibre versions (of starchy carbohydrates)*' (PHE 2016). Support for such a recommendation arises from an ever growing number of scientific reports suggesting wholegrains in place of refined grains improves the nutritional quality of carbohydrate intake (SACN 2015; Robinson & Chambers 2018), with wholegrains also recommended as part of sustainable, healthy diets (Willett *et al.* 2019). Thus, consumers will benefit most by choosing a healthy balanced diet rich in whole grains rather than avoiding grain intake.

Conclusion

Globally, grains are an important dietary source of energy and many nutrients, particularly carbohydrate. They can be also a noteworthy source of dietary contaminants, particularly mycotoxins and heavy metals. However, there are number of important assurances which ensure a place of wholegrains in our diets. Nutritionally, diets rich in wholegrains have been associated with a more nutrient dense diet than those rich in refined grains and with reduced risk of many chronic diseases. They are also promoted as a sustainable food choice. The importance of ongoing activities by Regulatory Authorities in providing guidance and assessing risk cannot be overstated in ensuring the safety of our food supply. In addition, we suggest that based on the observational and preclinical evidence described here, there may be potentially protective effects of the phytonutrients in grains against contaminants (*e.g.* mycotoxins and heavy metals) which merit further research. The consumer is best protected by eating healthy balanced diet, rich in nutrient dense foods, including wholegrain options.

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Table 1: Summary of the main mycotoxins in grains, their sources and effects on human health as modified from Thielecke & Nugent 2018

Mycotoxin	Fungal source(s)	Grain affected	Effects of ingestion for humans	
Deoxynivalenol/nivalenol	<i>Fusarium graminearum</i>	Wheat, maize, barley	Human toxicoses (e.g. nausea, vomiting, diarrhoea, headache, fever)	
	<i>Fusarium crookwellense</i>			
	<i>Fusarium culmorum</i>			
Zearalenone	<i>F. graminearum</i>	Maize, wheat	Possible human carcinogen	
	<i>F. culmorum</i>			
	<i>F. crookwellense</i>			
Ochratoxin A	<i>Aspergillus ochraceus</i>	Barley, wheat, other commodities	Suspected human carcinogen	
	<i>Penicillium verrucosum</i>			
Fumonisin B1	<i>Fusarium moniliforme</i> plus several less common species	Maize	Suspected human carcinogen	
Aflatoxin B1, B2	<i>Aspergillus flavus</i>	Maize, peanuts, other commodities	Potent human carcinogens by IARC	
Aflatoxin B1, B2, G1, G2	<i>Aspergillus parasiticus</i>	Maize, peanuts		

Table 1: Percent contribution of key commodities to energy, macronutrient and fibre intake of UK adults aged 19-64 years. Data from National Diet and Nutrition Survey years 7 and 8, 2014/2015 and 2015/2016. (Public Health England, 2018)

	Energy (%)	Protein (%)	Fat (%)	Carbohydrate (%)	Fibre (%)
All Cereal and cereal products	32	23	21	46	38
Meat and meat products	17	37	24	6	12
Milk and milk products	9	13	12	5	1
Fruit, vegetables and salad vegetables	9	7	6	11	28