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# RISK PROFILING OF EXPOSURES TO POTENTIALLY TOXIC METALS PTM(S) THROUGH NOODLES CONSUMPTION. A CASE STUDY OF HUMAN HEALTH RISK ASSESSMENT

– Research paper –

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**Abstract:** This study aimed to quantify levels of the toxic heavy metals metalloid arsenic (As), cadmium (Cd), lead (Pb) and mercury (Hg) in commercially available noodles. Analysis was performed via the acid microwave digestion method followed by inductively coupled plasma mass spectrometry (ICP-MS). Furthermore, the potential health risks resulting from the consumption of noodles by polish consumers were evaluated using the estimated average daily intake of heavy metals (ADIHMs; $\mu g \cdot kg^{-1}$ bw$\cdot day^{-1}$). Additionally, to assess the potential health consequences due to noodle consumption, the total hazard quotient (THQ) and cancer risk (CR) were calculated. The average contents of Pb, As, Cd, and Hg in instant noodle samples were determined as 9.241, 8.483, 29.39, and 1.661 $\mu g \cdot kg^{-1}$, respectively. THQ values for heavy metal toxicity were found to be below 1, indicating that the consumer is not exposed to carcinogenic health risks associated with the analyzed metals. Additionally, the rank order of max THQ was Pb < Cd < As < Hg, and the mean THQ was Pb < Hg < As < Cd, while both the max and mean CR rank orders were Pb < Cd < As.

**Keywords**: dietary intake; food contamination and analysis; risk assessment; ICP-MS.

## INTRODUCTION

It is well known that food is fundamental of human life. Cereal products are the basis of the food pyramid, and, therefore, their participation in the daily human diet is important. Grain-based products are a source of carbohydrates, fibre, protein, vitamins, and minerals, which are mostly ingested with food. The human body itself is not able to produce these nutrients; therefore, they should be provided in the right amounts and proportions. For this reason, the consumption of instant flour-based products, including noodles, as parts of one's daily diet is increasing worldwide (Tajdar-oranj et al., 2018). The consumption of pasta in Europe increased from 3,199,000 tonnes in 2019 to 3,322,000 tonnes in 2020, according to data provided by Union of the Organizations of Manufacturers of Pasta Products in the EU (UN.A.F.P.A.). The World Instant Noodles Association (WINA) estimated that the consumption of instant noodles in the world grew from 100,110 million servings in 2017 to 118,180 million servings in 2021. The largest amount of instant noodles was consumed in China/Hong Kong, with 43,990 million servings in 2021.

Poland was ranked 28$^{th}$ in the world, with the consumption of 330 million servings of instant noodles in 2021. Italian citizens eat 23.5 kg of pasta per year, which is the highest amount in the world and significantly higher than the amount eaten by Polish citizens at 4.2 kg per year, according to data provided by Union of the Organizations of Manufacturers of Pasta Products in the EU (UN.A.F.P.A.). Other countries known for eating pasta include Tunisia (17.0 kg per person), Venezuela (15.0 kg per person), Greece (12.2 kg per person), and Peru (9.9 kg per person). Pasta is not a popular choice in El Salvador (1.0 kg per person), Ireland (1.0 kg per person), Japan (1.7 kg per person), South Africa (1.9 kg per person), and Libya (2.0 kg per person). On average, a Polish citizen eats 11.5 grams of pasta per day. The popularity of noodles is related to their composition and ease of preparation. Noodles are sources of nutrients through the inclusion of, e.g., wheat flour. which contains proteins,

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carbohydrates, minerals, fatty acids, and sometimes additives, including stabilizers, emulsifiers, enzymes, phosphates, antioxidants, and colorants (Idrees, 2020). It is well known that pasta is one of the primary constituents of the Mediterranean diet, which represents a healthy dietary pattern. Pasta is cheap, convenient, versatile, filling and nourishing. With its long shelf life, pasta plays a major role in meeting the nutritional needs of the world (Webb, 2019). Unfortunately, noodles can also contain heavy metals (HMs), the main source of which is wheat. Soil contamination, the use of contaminated irrigation water, and pesticide- and HM-contaminated fertilizers are the main vectors through which heavy metals enter wheat and then noodles. Moreover, the determination of metals plays a major role in analyses of food, because this type of determination is crucial for the control and maintenance of the quality of products and processes, during both manufacture and research, and is needed to determine possible toxicity (Karaś et al., 2020). Taking into account data from the World Health Organization, which reported in 2010 that more than 0.4 million people died, and over 600 million were sick, from 31 of the most common food borne diseases, food safety is very important, not only locally but also worldwide (Akhtar et al., 2020). HMs are found in almost all types of food. Food and water are estimated to be the sources of over 90% of exposure to potential toxic metals (Pirhadi et al., 2022). HMs such as As, Cd, Hg, and Pb can be absorbed and accumulated by plants in the process of cultivation, owing to their unique characteristics, such as persistence and non-biodegradability in environmental media, high environmental stability, bio-accumulation, and bio-magnification in organisms and food chains (Zioła-Frankowska et al., 2021). In addition, chemical residues in food, as well as food production processes, have increased human exposure to heavy metals. Heavy metals such as Pb, Hg, Cd, and As, which have cumulative properties, can cause serious human health issues such as diseases, disorders, and the malfunctioning and malformation of organs due to metal toxicity, especially over a long period of heavy metal (HM) ingestion through high-content food products (Idrees, 2020; Karaś et al., 2021; Zioła-Frankowska et al., 2021). For example, cadmium can lead to cardiovascular disease and bone pain; lead can cause neurotoxicity affecting cognition, cerebral blood disease, gastrointestinal colitis, and damage to kidneys; mercury causes hearing, sight, and cognition impairment; and arsenic causes skin, bladder, and lung cancer, as well as keratosis (Moradi et al., 2021; Sigrist et al., 2016). Moreover, metals such as Pb and Cd were classified as human carcinogen compounds by the International Agency for Research on Cancer (IARC) (Akhtar et al., 2020; Idrees, 2020; Moradi et al., 2021; Tajdar-oranj et al., 2018). In addition, the Agency for Toxic Substances and Disease Registry (ATSDR) confirmed that Pb, As, and Hg affect human organ systems such as the neurological, cardiovascular, gastrointestinal, reproductive, renal, and respiratory (Charles et al., 2018). Considering both the direct and indirect effects of heavy metals on the human body, it is important to estimate the potential risks associated with the consumption of heavy metals in food, including noodles. Therefore, it is reasonable to assess the potential risk of toxic elements available in pasta, such as arsenic, cadmium, mercury, and lead, to human health. The list of approved food additives in the European Union is regulated by Commission Regulation (EU) No 1129/2011 of 11 November 2011 amending Annex II to Regulation (EC) No 1333/2008 of the European Parliament and the Council by establishing a Union list of food additives. The maximum levels for certain contaminants, including arsenic, cadmium, mercury, and lead, are regulated by Commission Regulation (EC) No 1881/2006 of 19 December 2006, which establishes the maximum levels for certain contaminants in foodstuffs. Estimating the risks of HM intake from noodles on human health is mostly based on the Target Hazard Quotients (THQ). This method does not give a quantitative approximate for the likelihood of an exposed population experiencing an adverse health effect, but can indicate the risk level associated with pollutant exposure over a specified period of time (Charles et al., 2018). The U.S. EPA (Environmental Protection Agency) defines human health risk assessment as "the process to estimate the nature and probability of adverse health effects in humans who may be exposed to chemicals in contaminated environmental media, now or in the future". The ATSDR explains that the hazard quotient (HQ) is "calculated to evaluate the potential for non-cancer health hazards to occur from exposure to a contaminant" and that cancer risk (CR) is calculated for carcinogens. Values of HQ below 1 indicate that there should be no non-cancer hazard risk to human health (Charles et al., 2018). The range for generally acceptable carcinogenic risk (CR) established by the U.S. EPA is $10^{-6}$ to $10^{-4}$. Carcinogenic risks with calculated values lower than $10^{-6}$ should not be of concern, while values higher than $10^{-4}$ are often considered unacceptable (USEPA – DRAS).

The present study was designed to analyze the content of Pb (TPb), Cd (TCd), As (TAs), and Hg (THg) in commercial noodles available on the polish market. This research was undertaken to estimate the daily intake rate of metals and the potential health risks associated with the intake of toxic contaminants from the consumption of noodles in Poland.

## MATERIALS AND METHODS

### 1. Chemicals and standards

The chemicals used for sample digestion, such as 70% nitric acid (for trace analysis), were purchased from Sigma-Aldrich, Poland. The standard solutions of Pb, Cd, As (concentration: 1.000 $mg \cdot L^{-1}$), and Hg (concentration: 1.000 $mg \cdot L^{-1}$) were also obtained from Sigma-Aldrich, Poland. The mixed solution including Sc, Rh, and Tb in 1% $HNO_3$ was used as an internal standard, and MilliQ Direct 8 (Millipore, USA) water was used for preparing the solution and pre-treating the sample.

### 2. Analytical method and quality control

HMs, including Pb, Cd, As, and Hg, in the digestion after the mineralization procedure and the blank tests for digestion reagents were analyzed using an ICP-MS 2030 inductively coupled plasma mass spectrometer (ICP-MS) (Shimadzu, Japan). The main operating parameters were the radio frequency power of the plasma, which was 1200 W, and the gas flow rates of the plasma gas, carrier gas, and auxiliary gas, which were 8.0, 1.10, and 0.7 $L \cdot min^{-1}$, respectively. The rate of helium achieved in the collision cell was 9.0 $mL \cdot min^{-1}$. The depth of sampling and the rate of sample injection were 5.0 mm and 0.6 $mL \cdot min^{-1}$, respectively. For the analysis of As, Cd, Hg, and Pb, an internal standard (IS) (using a T shape) was introduced with the sample via a peristaltic pump. As was analyzed using a 75 u mass, Cd using 114 u, Hg using 202 u, and Pb using 208 u. In addition, the selected elements were connected in pairs with IS elements (Rh, Rh, Tb, and Tb, respectively). The Limit of Detection for As, Cd, Hg, and Pb was 0.0063, 0.0031, 0.0009, 0.0030 $\mu g \cdot L^{-1}$, respectively. The accuracy of the analyses was verified using two certified reference materials (CRMs): Trace metal ICP — sample 1 and Trace metal ICP — sample 2, with recoveries of 94 - 105 % (certified values include all elements except Hg). For the dietary health risk exposure calculations in this study, when the analyzed heavy metal concentrations were below the LOD, (i.e., not detected (N.D.)), the values were assumed to be equal to half of the LOD.

### 3. Material and sample preparation

A total of 20 samples of noodles in commercially available sizes (500 g) were collected from different markets in Poland. The characteristics of the type of noodles are presented in Table 1.

The following method was used for the pre-treatment and digestion of all noodles samples: Firstly, the samples were crushed by hand by grinding in an agate pot. Next, 1000 ± 5 mg of the noodle sample was weighed directly in a Teflon digestion tube. Then, we added 8 mL of $HNO_3$ (70% suprapure, Sigma-Aldrich) and allowed the solution to rest for 1 h without the cap. Then, the samples were placed in a microwave digestion system (Microwave Reaction System Anton Paar, Multiwave PRO, Austria) under a power-controlled program. The microwave oven program with eight vessels (rotor 8NXF100) was as follows: 15-minute ramp time with a 20-minute hold time, followed by cool down from 190 to 70 °C. After digestion, the Teflon tubes were cooled to ambient temperature and then extracted to eliminate the gas that was generated during the digestion process, the caps were removed. Lastly, the prepared solutions were transferred to 8 mL volumetric flasks and diluted 10 times with Milli-Q water prior to analysis.

### 4. Evaluation of the potential health risks for people due to noodle consumption

#### 4.1. Average Daily Intake

The health risk assessment of heavy metals due to noodle/pasta consumption was marked based on dietary exposure to each metal. The average daily intake of heavy metals (ADIHMs) for adults was estimated using the equation (1):

$$ADIHMs = Cm \times IRn \times EFh \times EDf / Baw \times ATd \quad (1)$$

where Cm is the heavy metal concentration in the noodle/pasta samples ($\mu g \cdot kg^{-1}$); IRn is the daily intake of noodles in kg/person/day based on data from the Polish Statistical Book, 2020; Efh is the exposure frequency (365 days/year); Edf is the exposure duration (70 years); Baw is the average body weight (in this study, 60 kg was considered average); and ATd is the carcinogenic effect based on average exposure time (Efh x Edf) (Charles et al., 2018).

Table 1. Characterization of the analyzed noodles.

| Sample number | Name and type of pasta | Ingredients |
|---|---|---|
| 1 | Pastani Gluten Free Penne Pasta | Corn flour |
| 2 | Combino Gluten Free Spaghetti Pasta | Corn flour, water |
| 3 | Czaniecki 5-Egg Nest (shaped) Pasta | Wheat groats, semolina, eggs, water, turmeric |
| 4 | Combino Lasagne Pasta | Semolina (durum wheat), eggs, soy |
| 5 | Pastani Spaghetti 100% Durum Pasta | Semolina (durum wheat), eggs |
| 6 | Pastani Fusilli Pasta | Durum wheat flour, eggs |
| 7 | Tiradell Wholemeal Penne Pasta | Whole meal flour from durum wheat, eggs |
| 8 | Lubella Whole-grain Fusilli Pasta with Spelt | Whole-grain flour: wheat, spelt |
| 9 | De Luca Spaghetti Al Nero Di Seppia Pasta | Durum wheat, water, squid ink 3% |
| 10 | LubellaTagliatelle Mosse („Wave") Pasta | Wheat flour |
| 11 | Melissa Primo Gusto Tagiatelle Pasta | Semolina (durum wheat), water, eggs |
| 12 | Dalla Costa Spaghetti All Aglio e Peperoncino | Durum wheat semolina, dehydrated spices 3.9% (chili, garlic, paprika), garlic essential oil 0.2% |
| 13 | Tiradell Whole-grain Buckwheat „Tube" Pasta | Whole-grain buckwheat flour, wheat, gluten and eggs |
| 14 | Asia Flavours Spirulina Pasta | Wheat flour, water, powdered spirulina 2%, salt |
| 15 | Bartolini Fusilli Pasta | Semolina (durum wheat flour), dried tomato, dried spinach, dried beetroot, turmeric |
| 16 | Halloween Tricolore Pasta | Semolina (durum wheat), water, powdered tomatoes, squid ink, eggs, soy, fish and celery |
| 17 | Barilla Gnocchi Pasta | Semolina (durum wheat), water, soy |
| 18 | Lubella Mini „Tubes" Pasta with Vegetables | Wheat flour, powdered spinach, powdered carrot, powdered green peas, powdered broccoli, powdered pumpkin, eggs |
| 19 | Primo Gusto Farfalle Pasta | Semolina (durum wheat), water |
| 20 | Lubella Tagiatelle Rigate Pasta | Wheat flour |

### 4.2. Target Hazard Quotient — non-carcinogenic health effects

In evaluating the non-carcinogenic health risks due to the consumption of noodles/pasta, the THQ was calculated based on the equation (2):

$$THQ = ADIHMs / RfD_{ing} \quad (2)$$

where $RfD_{ing}$ is the oral/ingestion reference dose ($\mu g \cdot kg^{-1} \cdot day^{-1}$), which is used by the EPA for non-cancer health assessment (for Pb 3.5, Cd 1, As 0.3, and Hg 0.1) (WHO, 2011).

### 4.3. Carcinogenic Risk — carcinogenic health effects

The carcinogenic risk of heavy metals caused by the probability of getting sick from cancer due to the direct ingestion of noodles was calculated by the equation (3):

$$CR = ADIHMs \times CSF_{ing} \quad (3)$$

where $CSF_{ing}$ is the ingestion cancer slope factor $(mg \cdot kg^{-1} \cdot day^{-1})^{-1}$. The permissible range for carcinogenic risk over a lifetime of 70 years was recommended as $10^{-6}$ to $10^{-4}$ by the USEPA (CSF for Pb $85 \cdot 10^{-4}$, Cd $38 \cdot 10^{-2}$, As $15 \cdot 10^{-1}$, and Hg 0, respectively) (WHO, 2011).

### 4.4. Hazard Index

To estimate the cumulative potential health risk caused by a mixture of heavy metals due to the consumption of noodles, the hazard index (HI) based on the sum of the particular THG of each HM was applied as indicated in equation (4):

$$HI = THQ_{Pb} + THQ_{Cd} + THQ_{As} + THQ_{Hg} \quad (4)$$

## RESULTS AND DISCUSSION

The analysis of HMs was carried out for 20 samples of various types of pasta available on the polish market. The lowest arsenic content of 1.09 μg·kg$^{-1}$ was measured in sample 2 made from corn flour. The highest arsenic content of 33.02 μg·kg$^{-1}$ was found in sample 14 made from wheat flour with added spirulina and salt. The cadmium content in the analyzed pasta ranged from 1.255 to 55.33 μg·kg$^{-1}$. The lowest value was found in the sample made from corn flour, and the highest value was found in whole grain buckwheat tube pasta (sample 13). Mercury content was not observed in all pasta samples. The lowest mercury content, 0.070 μg·kg$^{-1}$, was found in wheat tagliatelle mosse ("wave") pasta (sample 10), and the highest value, amounting to 13.55 μg·kg$^{-1}$, was found in gluten-free penne pasta (sample 1). In the sample containing the highest mercury content, we also measured the lowest cadmium content. Likewise, for the two samples that contained the lowest mercury content, we measured the highest cadmium content. The lowest and highest amounts of lead were measured in the same samples that contained the lowest and highest amounts of arsenic, ranging from 0.960 to 52.13 μg·kg$^{-1}$ (samples 2 and 14, respectively). Considering the composition of the analyzed pasta, for pasta in which the main ingredient was wheat, the highest concentrations were measured for Cd and the lowest for Hg. For the samples in which ingredients were wheat and vegetable additives, the highest concentration was found for Pb and the lowest for Hg. Notably, in this group, the measured concentrations, especially for Hg, As, and Pb, were several times higher than those found in pasta produced only from wheat. For the group of pasta that did not contain wheat, the highest concentrations were again obtained for Cd. Notably, the concentrations obtained for Hg were the highest among the different types of analyzed pasta. Ghanati et al. (2019) analyzed heavy metals such as As, Cd, Co, Cr, Cu, Hg, Ni, Pb, and Zn in Iranian urban and rural wheat products, including wheat, wheat flour, bread, pasta, and sweets. In six analyzed pasta samples, the authors observed much higher quantities of elements such as arsenic (3404 - 6453 μg·kg$^{-1}$), cadmium (300 - 401 μg·kg$^{-1}$), mercury (15 - 22 μg·kg$^{-1}$), and lead (136 - 289 μg·kg$^{-1}$) compared to the results obtained in this research. Cubadda et al. (2002) analyzed the metal content in commercially available pasta and long pasta (spaghetti) manufactured from semolina using a pilot pasta-making apparatus. The authors found that the arsenic content ranged from 6.0 to 6.3 μg·kg$^{-1}$ in dry pasta, 5.4 to 5.7 μg·kg$^{-1}$ in fresh pasta, and 4 to 36 μg·kg$^{-1}$ in commercially available pasta. The determined cadmium content ranged from 16.4 to 44.4 μg·kg$^{-1}$ in dry pasta, 14.8 to 40.0 μg·kg$^{-1}$ in fresh pasta, and 22 to 103 μg·kg$^{-1}$ in commercially available samples. The determined lead content ranged from 18.4 to 26.8 μg·kg$^{-1}$ in dry pasta, 16.6 to 24.2 μg·kg$^{-1}$ in fresh pasta, and 2.6 to 9.6 μg·kg$^{-1}$ in commercially available pasta. These results are mostly comparable to the values obtained in this research from pasta samples available in Poland. In another study focused on products available on the polish market, Bielecka et al. (2020) determined the contents of toxic elements in various rice products, including pasta. The determined element contents in the analyzed rice pasta ranged from 48.0 to 65.2 μg·kg$^{-1}$ for arsenic, 8.0 to 37.3 μg·kg$^{-1}$ for cadmium, 1.1 to 7.3 μg·kg$^{-1}$ for mercury, and 18.9 to 60.1 μg·kg$^{-1}$ for lead. These results are comparable to the values obtained in this research for pasta samples available in Poland but without arsenic, whose content was much higher. Punshon and Jackson (2018) conducted a multielemental (Mn, Fe, Ni, Cu, Zn, Cr, Co, Se, Cd, Sb, Pb, As, Hg) market-based study of GF and gluten-containing ingredients and prepared foods. The authors determined the mean element contents for rice-based prepared foods, which, in pasta, was 141 μg·kg$^{-1}$ for arsenic, 15.9 μg·kg$^{-1}$ for cadmium, 3.4 μg·kg$^{-1}$ for mercury, and 22 μg·kg$^{-1}$ for lead. The determined mean element contents for non-rice-based prepared foods and pasta was 15 μg·kg$^{-1}$ for arsenic, 52.4 μg·kg$^{-1}$ for cadmium, 1.0 μg·kg$^{-1}$ for mercury, and 7 μg·kg$^{-1}$ for lead. These results are comparable to the values obtained in this research for pasta samples, especially non-rice pasta. Marín et al. (2018) tested metal levels in various foodstuffs from the Region of Valencia (Spain). The determined total arsenic content in pasta ranged from 11.2 to 39.0 μg·kg$^{-1}$, cadmium content from 14.2 to 24.0 μg·kg$^{-1}$, and lead content from 29.0 to 111.0 μg·kg$^{-1}$, which was higher compared to the present results. Collumb et al. (2019) tested trace elements (Al, Ba, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Mo, Ni, Ti, and Zn) in gluten-free pastas and flours from markets located in the Las Vegas, Nevada Area. Cadmium content was determined to range from 150 to 400 μg·kg$^{-1}$ in rice pasta from the US market, 250 to 500 μg·kg$^{-1}$ in rice pasta from the Asian market, and 200 to 350 μg·kg$^{-1}$ in gluten free pasta. Importantly, these results represent on average 10 times higher than the results obtained for pasta samples available in

Poland. Additionally, in this study, the average content for gluten pasta was only 1.77 μg·kg$^{-1}$. The cadmium content in pasta measured by the FASFC (Federal Agency for the Safety of the Food Chain) in 2006, 2007, and 2008 ranged from 11 to 130 μg·kg$^{-1}$. In this study, the average content was much lower (29.38 μg·kg$^{-1}$). Malavolti et al. (2020) determined the cadmium content in pasta, the most frequently consumed food in a typical Italian diet, from two Northern Italian provinces of the Emilia–Romagna region. The mean cadmium content in pasta and other grains was 11.41 μg·kg$^{-1}$, which is lower than the mean value obtained in this research from pasta samples available in Poland. Tajdar-oranj et al. (2018) investigated the concentration of heavy metals, including lead, chromium, cadmium, and aluminum, in commonly consumed instant noodles in Iran. The determined cadmium and lead contents in instant noodle samples ranged from 30 to 140 μg·kg$^{-1}$ for cadmium and from 1004 to 1570 μg·kg$^{-1}$ for lead. These results are higher than the values obtained in this research for pasta samples available in Poland, especially for lead. Wei and Cen (2020) determined the concentration of heavy metals in cereals, legumes, and their corresponding products. The mean concentration of cadmium and lead in dried noodles was 12.36 μg·kg$^{-1}$ for cadmium and 58.96 μg·kg$^{-1}$ for lead. The obtained cadmium concentration was lower than the mean cadmium concentration obtained in this study, but the mean lead concentrations were comparable.

**1. Evaluation of potential human risk from noodle consumption**

It is well known that there are many sources of human exposure to heavy metals, such as water, air, soil, and food, the last of which is a major exposure route. Characterisation of potential toxic effects caused by the excessive presence of heavy metals, such as As, Pb, Cd, and Hg, all throughout the food chain, is measured by human health risk assessment. However, it's not possible to estimate the risks without prior determination of metal concentration in food. In this study, only the exposure route of noodle consumption was considered for the results of human health risk calculations (Table 2).

However, the estimated health risk based on various factors was found to be acceptable even for a single exposure route. The average daily intake of heavy metals in pasta obtained in this study was 0.007 μg·kg$^{-1}$·day$^{-1}$ for arsenic, 0.012 μg·kg$^{-1}$·day$^{-1}$ for cadmium, 0.003 μg·kg$^{-1}$·day$^{-1}$ for mercury, and 0.012 μg·kg$^{-1}$·day$^{-1}$ for lead. All obtained values were lower than the RfD for those elements, which means that they are not likely to be associated with adverse health risks. Additionally, the ADIHMs of heavy metals (μg·kg$^{-1}$ bw·day$^{-1}$) were compared with the Provisional Tolerable Weekly Intake (PTWI), which is 25, 7, and 15 μg·kg$^{-1}$bw·week$^{-1}$, respectively, for Pb, Cd, and As (Akhtar et al., 2020). The weekly intake values for noodles obtained in the current study were much lower than the PTWI values, which confirms that noodles do not have significant health risks for consumers. However, this result does not mean that these content values are acceptable and risk-free. Ghanati et al. (2019) calculated the Chronic Daily Intake (CDI) as 0.2 μg·kg$^{-1}$·day$^{-1}$ for arsenic, 0.1 μg·kg$^{-1}$·day$^{-1}$ for cadmium, 0.1 μg·kg$^{-1}$·day$^{-1}$ for mercury, and 7.9 and 9.8 μg·kg$^{-1}$·day$^{-1}$ for lead and other elements in pasta. These values are higher than the ADI values obtained in this study but still lower than RfDs. Bielecka et al. (2020) calculated the Estimated Daily Intake (EDI) as 0.27 μg·kg$^{-1}$·day$^{-1}$ for arsenic, 0.10 μg·kg$^{-1}$·day$^{-1}$ for cadmium, 0.02 μg·kg$^{-1}$·day$^{-1}$ for mercury, and 0.16 μg·kg$^{-1}$·day$^{-1}$ for lead in rice pasta; these values are also higher than the ADI values obtained in this study. Malavolti et al. (2020) calculated the Daily Dietary Intake (DDI) of lead in food. The mean DDI for all subjects (types of food) was 0.155 μg·kg$^{-1}$·day$^{-1}$, which is higher than the ADI values obtained in this study for lead. In addition, we compared the ADIHM values with the values of MRLs (Minimal Risk Levels), which for As is 5 μg·kg$^{-1}$·day$^{-1}$ for acute and 0.3 μg·kg$^{-1}$·day$^{-1}$ for chronic oral ingestion and for Cd is 0.1 μg·kg$^{-1}$·day$^{-1}$ for chronic ingestion. In both cases our results were below the permitted MRL values. It should be underlined that the MRL factor is defined as "an estimate of the daily human exposure to a substance that is likely to be without an appreciable risk of adverse, non-cancer effects over a specified duration of exposure" (Geronimo et al., 2021). The various factors were calculated to assess the potential risk associated with heavy metal contamination in samples of various noodles types. For all analyzed metals, the maximum THQ values were below 1 in the following order: Hg > As > Cd > Pb. The average THQ values for metals were much lower and in a different order: Cd > As > Hg > Pb. This result indicates that there is no risk to human health from the daily consumption of pasta at the determined levels of metal concentrations.

Table 2. Results of content, average daily intake (ADI), target hazard quotient (THQ), and carcinogen risk (CR) of each element in pasta and the corresponding hazard index (HI).

| Element | Content [μg·kg$^{-1}$] Mean (min – max) | ADI [μg·kg$^{-1}$ per day] Mean (min – max) | THQ Mean (min – max) | CR Mean (min – max) |
|---|---|---|---|---|
| **As** | 8.483 (1.087 – 33.02) | 0.0019 (0.0002 – 0.0073) | 0.0063 (0.0008 – 0.0244) | 2.820 x $10^{-6}$ (3.613 x $10^{-7}$ – 1.098 x $10^{-5}$) |
| **Cd** | 29.39 (1.255 – 55.33) | 0.0065 (0.0003– 0.0123) | 0.0065 (0.0003 – 0.0123) | 2.475 x $10^{-6}$ (1.058 x $10^{-7}$ – 4.660 x $10^{-6}$) |
| **Hg** | 1.661 (<LOD – 13.55) | 0.0004 (1.534 x $10^{-5}$ – 0.0030) | 0.0037 (0.0002 – 0.0300) | - |
| **Pb** | 9.241 (<LOD – 52.13) | 0.0020 (0.0002 – 0.0116) | 0.0006 (6.072 x $10^{-5}$ – 0.0033) | 1.741 x $10^{-8}$ (1.806 x $10^{-9}$ – 9.822 x $10^{-8}$) |
| **HI Mean (min – max)** | | | 0.0171 (0.0077 – 0.0331) | |

Lead presented the lowest potential risk to human health compared to other analyzed metals. In pasta samples made with wheat flour, the highest maximum THQ value was obtained for arsenic in the following order: As > Cd > Hg > Pb. In pasta samples made with corn flour and whole grain flour (buckwheat and spelt flours), the order of maximum THQ values was different: Hg > Cd > As >Pb.

The Hazard Index was also determined for all analyzed metals. For the highest mean concentrations, the HI values were 0.070 for all pasta samples, 0.021 for wheat pasta, 0.046 for wheat pasta with additional ingredients, and 0.052 for corn and whole grain pasta. The percentage of each analyzed metal with the mean highest HI determined for all pasta was As (34.85%), Cd (17.52%), Hg (42.91%), and Pb (4.72%). For the mean concentrations of metals, the HI values were 0.017 for all pasta, 0.012 for wheat pasta, 0.022 for wheat pasta with additional ingredients, and 0.020 for corn and whole grain pasta. The percentage of each analyzed metal with the mean HI determined for all pasta was As (36.76%), Cd (38.21%), Hg (21.60%), and Pb (3.43%). Both HI values (highest 0.070; mean 0.017) were significantly lower than 1, which indicates minimal exposure to risk, with no significant health risk for people who consume pasta. Carcinogenic Risk was found for arsenic, cadmium, and lead in Polish pasta. The maximum values were 1.097 x $10^{-5}$ for arsenic, 4.660 x $10^{-6}$ for cadmium, and 9.822 x $10^{-8}$ for lead. The highest CR values for arsenic and lead were determined for the sample made from wheat flour and spirulina. The highest CR value for cadmium was found in pasta made from buckwheat flour. All determined CR values were below or within the permissible range for carcinogenic risk over a lifetime of 70 years (average range 1.741 x $10^{-8}$ to 2.820 x$10^{-6}$), as recommended by USEPA, at levels from $10^{-6}$ to $10^{-4}$. Ghanati et al. (2019) analyzed heavy metals such as As, Cd, Co, Cr, Cu, Hg, Ni, Pb, and Zn in Iranian urban and rural wheat products including wheat, wheat flour, bread, pasta, and sweets. For all analyzed elements, the determined HI was 1.83 for urban samples and 2.28 for rural samples. These values were significantly higher than the values obtained in this research for only As, Cd, Hg, and Pb. Bielecka et al. (2020) determined the contents of the toxic elements As, Cd, Hg, and Pb in rice products available on the Polish market. The obtained THQ values ranged from 0.0114 to 0.0155 for arsenic, 0.0006 to 0.0027 for cadmium, 0.0003 to 0.0017 for mercury, and 0.0004 to 0.0012 for lead. These values are mostly comparable to the values obtained in this research because in this study, we obtained higher values for mercury (max. 0.0300). Tajdar-oranj et al. (2018) calculated the THQ for Pb, Cr, Cd, and Al for both adult and child consumers due to the ingestion of instant noodles. The THQ for Cd in adults was 0.010, and the THQ for Pb in adults was 0.012. For children, the THQ was 0.035 and 0.044, respectively. The THQ for cadmium is comparable to that of lead, although the THQ for lead is slightly higher than the values obtained in this research. Charles et al. (2018) performed a health risk assessment on instant noodles commonly consumed in Port Harcourt, Nigeria. The analyzed metals were Pb, As, Ni, Hg, Cu, Cd, Al, and Cr, which were analyzed in six brands of noodles. The HI values were, depending

on the brand, 5.004, 1.680, 0.150, 0.135, 0.104, and 0.179; these values are higher than the values obtained in this research. The THQ values were also determined as follows: 0.018 for arsenic, 0.005 for cadmium, 0.054 for mercury, and 0.002 to 4.91 for lead. The lowest THQ values are comparable to the values obtained in this research, while the values of 1.59 and 4.91 obtained for lead are significantly higher. In this research, the highest concentrations of mercury were found in pasta made from corn flour. Xu et al. (2022) aimed to determine the distributions of Hg, Cd, As, Pb, Cr, and Cu in corn and rice samples collected from nine locations in Guizhou. The obtained mean values of EDI for arsenic, cadmium, mercury, and lead, in corn were, respectively, 13, 34, 3, and 142 $\mu g \cdot kg^{-1} \cdot day^{-1}$, which were significantly lower than the EDI values obtained for rice. The mean values of THQ in corn were 0.004 for arsenic, 0.068 for cadmium, 0.011 for mercury, 0.094 for lead, 0.0002 for Cr, and 0.03 for Cu. The mean HI value in corn was 0.20, which was higher than the mean HI obtained in this study for pasta (0.017). The mean THQ of mercury obtained for corn was comparable to the THQ of mercury obtained for corn pasta in this study. The highest concentration of cadmium in this study was found in the buckwheat pasta sample. Bielecka et al. (2022) studied gluten-free cereals and pseudo cereals as a potential source of exposure to the toxic elements arsenic, cadmium, mercury and lead. The obtained THQ values of these elements in buckwheat groats were 0.0004 for arsenic, 0.00209 for cadmium, 0.0001 for mercury, and 0.00037 for lead. The accumulative HI was 0.00295, which is lower than the HI obtained in this study for buckwheat pasta 0.01617. The THQ of cadmium in buckwheat groats was also lower than that determined in buckwheat pasta in this study. One of the highest concentrations of mercury and cadmium was also found in wheat pasta made with powdered vegetables such as tomatoes and carrots, green peas, spinach, broccoli, and pumpkin. Mawari et al. (2022) analyzed the accumulation of heavy metals (arsenic, cadmium, mercury, and lead) in fruits and vegetables and assessed the corresponding human health risk. The Hazard Index values obtained were 0.1359 for tomatoes, 0.3659 for carrots, and 0.0997 for green peas. These values are higher than those obtained for pasta in this study. Ahmed et al. (2022) analyzed heavy metal accumulation in leafy vegetables grown on contaminated and non-contaminated soil. The HI obtained for Cr, Zn, Ni, Cd, and Cu in spinach grown on non-contaminated soil was 0.1725, and the Hazard Quotient (HQ) just for cadmium was 0.0024, which is lower than the THQ value obtained for cadmium in pasta with the addition of vegetables, including spinach. It is well known that toxicity of arsenic is connected to the chemical forms of this metalloid, where inorganic arsenic (i-As) compounds are more toxic than organic forms. In this study, the contribution of noodles to inorganic arsenic was estimated by applying a conversion factor of 70% to the total arsenic data from the noodles. The chosen value of the conversion factor is the most common value reported in the literature (Sigrist et al., 2016). According to our findings, the highest inorganic arsenic concentration was found in pasta made from wheat flour with spirulina, and the lowest concentration was obtained in a sample made from corn flour. Moreover, taking into consideration the type of flour, the largest contribution of the i-As form was obtained in noodles made from wheat flour with additional ingredients, and the lowest contribution was estimated for noodles made from different types of flour other than wheat (Fig.1). In addition, the calculation of the contribution to the inorganic arsenic intake of pasta per day confirmed that values lower than those of RfD for arsenic (the mean estimated ADI for i-As is 0.0013 $\mu g \cdot kg^{-1} \cdot day^{-1}$) are safe for humans. The results for inorganic arsenic in this study are similar to those obtained for wheat flour products (a mean i-As of 25 $\mu g \cdot kg^{-1}$) by others (Sigrist et al., 2016). Moreover, our findings confirm the previous statement by the EFSA that the primary contribution to i-As dietary intake for adults is connected to the consumption of wheat products (Sigrist et al., 2016).

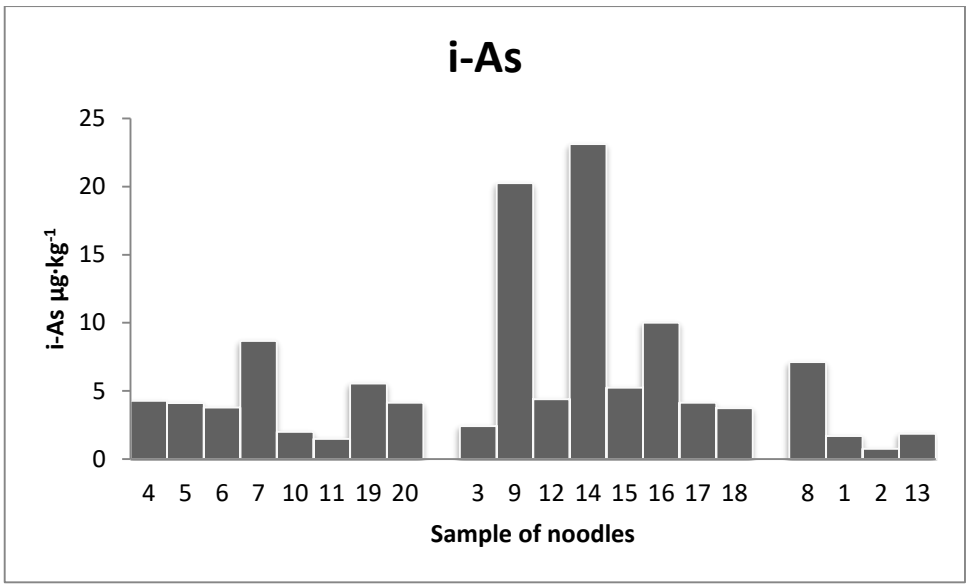



Figure 1. Results for the i-As inorganic form in pasta samples (first group — wheat flour; second group — wheat flour with additional ingredients; third group — different types of flour other than wheat).

## CONCLUSIONS

In this research, the contents of arsenic, cadmium, mercury, and lead were analyzed in pasta samples with various ingredients available on the polish market, and their health risks were assessed, including the ADI, THQ, CR, and HI. The rank order of both the max and mean heavy metal content was Hg < As < Pb < Cd. The ADI values were calculated to be within the permissible limit. The THQ and HI values calculated for each sample were below 1. For all analyzed metals, the maximum THQ was observed in the following order: Hg > As > Cd > Pb. The average THQ values for metals were much lower, however, and in a different order: Cd > As > Hg > Pb. The HI percentage of each analyzed metal was, in descending order, As (34.86%), Cd (17.52%), Hg (42.91%), and Pb (4.72%). All calculated values for CR were also below the permissible limit, which ranged from $10^{-4}$ to $10^{-6}$. The order rank for both the max and mean CR rank was Pb < Cd < As. Based on an estimation of risk factors, it can be stated that the consumption of noodles is associated with very few health risks. However, the possible health risks of heavy metals may become elevated for individuals with increased daily consumption. It is essential to conduct more extensive research focusing on the mitigation measures for the control of HM contamination in pasta to further reduce cancer risks, especially for unexpected contaminants in wheat, which are the main ingredients of noodles. Additionally, since this study only analyzed pasta and other routes of heavy metal exposure, such as various types of food, environmental and occupational sources, weren't considered, results cannot be regarded as comprehensive assessment of overall heavy metal exposure.

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A.Z.F. and D.J.; methodology, A.Z.F and M.F.; validation, A.Z.F. and M.F.; formal analysis, D.J. and M.F.; investigation, A.Z.F. and D.J.; writing—original draft preparation, A.Z.F. and D.J.; writing—review and editing, A.Z.F., D.J.,I.K. and M.F.; visualization, D.J.; supervision, A.Z.F..; funding acquisition, D.J. All authors have read and agreed to the published version of the manuscript.

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