Human exposure to trace elements, aromatic amines and formaldehyde in swimsuits: Assessment of the health risks

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ABSTRACT

Nowadays, most of the swimsuits are mainly made of artificial fibres, which have interesting properties such as water repellence and fast drying. Swimsuits contain a wide range of additives, which can mean a hazard for the environment and/or human health. In this study, the concentrations of formaldehyde (free and water soluble), 24 aromatic amines, and 28 trace elements (Ag, Al, As, B, Ba, Be, Bi, Cd, Co, Cr, Cu, Fe, Hg, Mg, Mn, Mo, Ni, Pb, Sb, Sc, Se, Sm, Sr, Sn, Tl, Ti, V and Zn) were analysed in 39 swimsuits covering a wide range of materials, colours and brands. Dermal exposure and health risks were assessed for adults (men and women) aged > 18 years old, babies between 2 and <3 years old, children (boys and girls) between 3 and <6 years old and 6 and <11 years old, and teenagers (boys and girls) between 11 and <16 years old, wearing swimsuits for 4 h or 8 h. Formaldehyde and aromatic amines were below their respective detection limits in all samples (< 16 and < 1.5 mg/kg, respectively). Regarding trace elements, Ti showed the highest mean levels (1,844 mg/kg), being significantly higher in polyamide (3,759 mg/kg) than in polyester (24.1 mg/kg) swimsuits. These high Ti levels were confirmed by environmental scanning electron microscope in a single sample made of polyamide. Increased concentrations of Cr were also observed, but only in polyamide black fabrics, with values ranging from 624 to 932 mg/kg. Non-cancer risks (hazard quotients) derived from the exposure to trace elements were in a safe zone for all analysed trace elements. Furthermore, the carcinogenic risks were evaluated for As, Cr and Pb, exhibiting values below the 10^{-5} threshold, with the exception of Cr in babies and children-girls. For Ti, health risks could not be calculated due to the lack of information on toxicological data. However, because Ti was the element with the highest concentrations in swimsuits, and taking into account the potential toxicity of TiO2 nanoparticles, further research is needed to assess the migration of this element from fibres to skin.

Keywords: swimsuits, trace elements, formaldehyde, aromatic amines, human exposure, risk assessment

1. Introduction

A number of recent studies have demonstrated the presence of toxic substances in clothing (Avagyan et al., 2015; Gong et al., 2016; Lorenz et al., 2012; Luongo et al., 2014; Rovira and Domingo, 2019). In 2012, Greenpeace launched the campaign "Toxic Threads", which was focused on determining the potential levels of nonylphenol ethoxylates, phthalates, and amines, among other toxic substances in clothing (Brigden et al., 2012). Although the definition of clothing wears refers to any type of garment worn in a particular situation ("Clothing. Cambridge Advanced Learner's Dictionary & Thesaurus online," 1993), people do not take into account the bath clothes.

A swimsuit is defined as a piece of clothing whose purpose is to cover a part of the body while people take a bath in beaches, swimming pools, rivers or lakes, among others. In men, swimwear generally covers the part of the genitals and extends to the legs, to a lesser or greater length. In women, the swimsuit usually covers the entire chest and genitals. Initially, swimsuits covered almost completely the body and were made of natural fabrics, either cotton or wool. At that time, swimsuits were heavy and slow to dry when they got wet. In the 1960s, with the generalization of synthetic fibres and because of its resistance, adaptability, elasticity and fast drying, they started to be used for the manufacture of swimwear. Currently, most swimsuits are made of polyester or polyamide, sometimes combined with elastane, due to the multiple advantages that these two synthetic materials offer: lightness, fast drying, perfect fit, and resistance (Niwa et al., 2014). However, synthetic fibres are also associated with some negative aspects such as the environmental impact derived from their manufacture (De Falco et al., 2019; Van Der Velden et al., 2014), as well as the presence of hazardous substances for human health (Antal et al., 2016; Jung et al., 2019; Rovira et al., 2015).

Polyester is a plastic material that offers multiple advantages over other fibres, such as cotton. It is more economic, resistant, does not deform, and absorbs less moisture, making more difficult the bacterial development (Martins et al., 2013). However, it gets dirty quickly, owning a high affinity for grease, earth and oil, while only dispersed dyes are used for staining water (Freeman and Mock, 2012). In turn, polyamide is a type of polymer that is characterized by having in the chain recurring amide groups, mostly attached to aliphatic or cyclo-aliphatic groups. These types of fibres stand out for being resistant to abrasions, putrefaction, and even seawater. On the other hand, they dry very rapidly and have low humidity absorption capability, characteristics that make this type of fibres suitable for swimsuits (Mukhopadhyay, 2009).

Chemical additives are used in swimsuits to improve or to add some properties to synthetic fibres (Schindler and Hauser, 2004). For example, catalysts make the production process faster and more efficient (James et al., 2001). Other additives are used to obtain fabrics with different finishes in order to satisfy the consumers' demands or needs (Schindler and Hauser, 2004). These substances include flame retardants (Kajiwara et al., 2009), antimicrobial agents (He et al., 2017), dyes (Ramugade et al., 2019), plasticizers such as phthalates (Negev et al., 2018), pesticides (Cai et al., 2013), as well as a variety of trace elements. Moreover, metals have many uses in the textile industry such as complex dyes, antimicrobials (metallic salts) and water repellents (ZnO) (Derden and Huybrechts, 2013; Morais et al., 2016; Xu et al., 2018; Zhang and Jiang, 2018).

Chemicals used in the textile industry can have a negative impact on the environment and also on human health (Saxena et al., 2017). They can migrate from clothes (Herrero et al., 2019; Leme et al., 2014; Rovira et al., 2017a) and enter in contact with the skin, causing dermatitis, allergies and, microflora alterations, among other adverse effects (Mobolaji-Lawal and Nedorost, 2015; Murayama et al., 2013; Walter et al., 2014). Therefore, it is important to know their contents in clothing, as well as the potential health risks.

The present study was aimed at evaluating the presence of formaldehyde, aromatic amines and a number of trace elements in commercially available swimsuits. Moreover, the dermal exposure to each chemical, and the associated human health risks, were also assessed. To the best of our knowledge, this is the very first time that this kind of textile products are investigated.

2. Materials and methods

2.1. Sampling

Swimsuits characteristics regarding material, colour, manufacture location, and density are shown in Table 1. A total of 39 garments were used for analysis, including man swimwear (n=16), woman swimwear (n=16), child swimwear (n=6), and a T-shirt (n=1). The range of colours varied from white to black, containing several shades of blue, fluorescent colours (yellow, orange, and pink) and prints. With respect to the materials, 16 were manufactured with a combination of polyamide and elastane, 15 were 100% polyester, 6 combined polyester and elastane, while two were 100% polyamide. All items were randomly purchased in different stores and supermarkets of Reus and Tarragona (Catalonia, Spain). Most of them were made in Asian countries, with only three manufactured in Spain.

2.2. *Chemical analysis*

2.2.1. Trace elements

Prior to trace element determination, an acidic digestion of each sample was performed. For polyester samples, 0.3 g of each garment was completely digested with 8 mL of HNO_3 (65% Suprapur, E. Merck, Darmstadt, Germany). In turn, polyamide samples were treated differently: 0.2 g of garment was treated with 1.5 mL of H_2SO_4 (96% Suprapur, E. Merck) and 6.5 mL of HNO_3 (65% Suprapur, E. Merck). Irrespective of the material, all samples were digested in a Milestone Start D Microwave Digestion System for 5 min at 105 °C, followed by 15 min at 180 °C, and 20 min at 200 °C. After cooling, extracts were filtered and brought to a volume of 25 mL with ultrapure water. As quality control, blanks were also run during the digestion every 8 samples. Spinach leaves (1570a - National Institute of Standards and Technology, Gaithersburg, MD, USA) were used as reference material every 8 samples (Rovira et al., 2015). All materials in contact with samples and extracts were previously cleaned with a diluted (10%) HNO_3 solution.

The concentrations of aluminium (Al), antimony (Sb), arsenic (As), barium (Ba), beryllium (Be), bismuth (Bi), boron (B), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), magnesium (Mg), manganese (Mn), mercury (Hg), molybdenum (Mo), nickel (Ni), samarium (Sm), scandium (Sc), selenium (Se), silver (Ag), strontium (Sr), thallium (Tl), tin (Sn), titanium (Ti), vanadium (V) and zinc (Zn), were determined by inductively coupled plasma mass spectrometry (ICP-MS, Perkin Elmer Elan 6000) in all swimsuits samples, as well as in standards, blanks and replicates (Rovira et al., 2017a). Detection limits were the following: 0.02 mg/kg for Ag, Be, Bi, Cd, Co, Mo, Sc, Sm and Tl; 0.04 mg/kg for As, Ba, Cr, Cu, Mn, Ni, Pb, Sb, Sn, Sr and V; 0.08 mg/kg for Hg and Ti; 0.21 mg/kg for B and 0.42 mg/kg for Al, Se and Zn; 8.33 mg/kg for Fe; and 20.8 for Mg.

2.2.2. Formaldehyde

The content of free and hydrolysed formaldehyde was determined according to the standard UNE-EN ISO 14184-1:2011, Determination of formaldehyde — Part 1: Free and hydrolysed formaldehyde (water extraction method) (AENOR, 2011; Piccinini et al., 2007). Briefly, an amount of the sample (1.0 g) was incubated with water at 40 °C. The total amount of free and hydrolysed formaldehyde was determined by colorimetry with absorbance at 412 nm after adding acetyl-acetone. Blank and standards were used. The detection limit was 16 mg/kg.

2.2.3. Aromatic amines

The analysis of aromatic amines was carried out according to the UNE-EN ISO 14362-1: 2017 method for determination of certain aromatic amines derived from azo (AENOR, 2017; Piccinini et al., 2008). Samples were extracted and the extracts were then added to a reducing media. After that, a liquid –liquid extraction were performed. Quantification of aromatic amines was performed using gas or liquid chromatography coupled to mass spectroscopy (GC-MS or LC-MS). The content of the following aromatic amines was analysed: 4-aminobiphenyl, benzidine, 4-chloro-o-toluidine, 2-naphthylamine, o-aminoazotoluene, 5-nitro-o-toluidine, 4-chloroaniline, 2,4-diaminoanisole, 4,4'-diaminodiphenylmethane, 3,3'-dichlorobenzidine, 3,3'-dimethoxybenzidine, 3,3'-dimethylbenzidine, 4,4'-methylene-di-o-toluidine, p-cresidine, 4,4'-methylene-bis-(2-chloraniline), 4,4'-oxydianiline, 4,4'-thiodianiline, o-toluidine, 2,4-diaminotoluene, 2,4,5-trimethylaniline, o-anisidine, 2,4-xylidine, 2,6-xylidine, and 4-aminoazobenzene. The limit of detection was set at 1.5 mg/kg.

2.3. Environmental scanning electron microscope (ESEM)

Environmental scanning electron microscope (ESEM) (FEI Quanta 600) attached to an energy dispersive X-ray (EDX) (Oxford Instruments INCA X-Sight, Abingdon, UK), with a backscattering detector (BSD) contrast by atomic number (Z), was used to evaluate the presence and composition of particles in clothing samples (Rovira et al., 2017a). ESEM working parameters were the following: low vacuum, 20 kV accelerating voltage, and 10 mm working distance.

2.4. Exposure and risk assessment

The concentration of the analysed trace elements and organic substances in swimsuit samples were used to calculate the dermal exposure and the associated risks. The exposure was estimated for different age groups: adults (men and women) aged >18 years old, babies between 2 and <3 years old, child (boy and girl) between 3 and <6 years old and between 6 and <11 years old and teenagers (boy and girl) 11<16 years old wearing swimsuits for 8 h. We considered swimming briefs and broads shorts in child boys, teenagers' boys and adults' men and also both bikini and swimsuit for teenagers' girls and adults' women in the analyses. Dermal exposure was calculated with the Equation 1 based on the European Chemical Agency (ECHA) (European Chemicals Agency, 2016).

$$\text{Exp derm} = \frac{F_{cloth}\ x\ d_{cloth}\ x\ A_{skin}\ x\ F_{mig}\ x\ F_{contact}\ x\ F_{pen}\ x\ T_{contact}\ x\ n}{BW} \qquad \text{(Equation 1)}$$

where Exp_{derm} is the dermal exposure (mg/(kg·day)); F_{cloth} is the fraction of element in clothes (dimensionless); d_{cloth} is the density of the clothing (mg/cm^2); A_{skin} is the skin area covered by the clothing (cm^2); F_{mig} is the migration fraction of substance from cloth to skin per day (1/day); $F_{contact}$ is the fraction of contact area for skin (dimensionless); F_{pen} is the penetration rate of the element (dimensionless); $T_{contact}$ is the duration of the clothing skin contact (dimensionless); n is the number of events per day (1/day), and BW is the body weight (kg). A F_{mig} of 0.5% was assumed to calculate dermal exposure (Bundesinstitut für Risikobewertung, 2012). Two scenarios (4 and 8 h) were assumed based on the potential time that swimsuits are worn. The dermal exposure parameters are summarized in Table 2.

Non-carcinogenic risks were determined calculating the hazard quotient (HQ), which is defined as the quotient between exposure and the dermal reference dose (RfD). The carcinogenic risk was calculated multiplying the exposure by the respective dermal slope factor (SF). In turn, as dermal toxicological parameters (RfD and SF) are not available, the dermal RfD was calculated multiplying the respective oral RfD by the gastrointestinal (GI) absorption factor, while dermal SFs were calculated dividing the respective oral SF by the GI absorption factor (US EPA, 1989). RfDs and SFs were obtained from the Risk Assessment Information System (RAIS, 2018). The only exception was the oral RfD for Pb, which is not defined in the RAIS, being taken from Seiler and Sigel (1988). GI absorption factors were obtained from the U.S. EPA Preliminary Remediation Goals (US Environmental Protection Agency, 2016).

2.5. Statistics

Data analysis was carried out using the IBM SPSS Statistics software (version 25.0). The Kolmogorov-Smirnov test was used to assess the distribution of the values. Correlations between metal concentrations were performed by applying the Pearson and Spearman correlation coefficients for parametric and non-parametric data, respectively. The student's t-test was used to compare differences of metal levels between polyester and polyamide, as well as elastane and non-elastane fibres. In turn, the ANOVA test for data following a parametric distribution, or the Kruskal Wallis tests for non-parametric data, were used to assess differences between groups according to the colour. A difference was considered as statistically significant when the probability was lower than 0.05 ($p<0.05$).

3. Results and discussion

3.1. Concentrations of formaldehyde, aromatic amines and trace elements

In all samples, the concentrations of formaldehyde and aromatic amines were below the respective detection limits: < 16 mg/kg for free and hydrolysed formaldehyde, and < 1.5 mg/kg for the 24 aromatic amines. Although previous studies have reported the presence of formaldehyde (Novick et al., 2013; Piccinini et al., 2007) and aromatic amines (Brüschweiler et al., 2014) in clothing and their potential health risks (De Groot et al., 2010; Malinauskiene et al., 2012), studies focused on the determination of these chemicals in swimsuits are missing in the scientific literature. For comparative purpose, reported data on the levels of formaldehyde in cloths made with polyester have typically ranged between <20 and 189.6 mg/kg (United States Government Accountability Office, 2010). In turn, the levels of aromatic amines are between 5 and 65 mg/kg according to a recent study conducted in the Swiss market (Crettaz et al., 2019).

With respect to inorganic chemicals, 28 trace elements were analysed in the current study. The concentrations of Be, Fe, Hg, Sc, Se, Sm and Tl were below their respective detection limits in all the samples. Other elements, such as Ag, As, and Bi, were only detected in a few samples (Table 3). These results are in accordance with those of recent studies conducted in our laboratory (Herrero et al., 2019; Rovira et al., 2017b, 2017a, 2015). Titanium was the element showing the highest mean concentration (1,844 mg/kg), followed by Al (249 mg/kg), Cr (122 mg/kg), Sb (45.4 mg/kg) and Cu (27.9 mg/kg). The highest Ti concentration (6,603 mg/kg) was found in sample #31 (fluorescent pink girl swimsuit, 80% polyamide and 20% elastane). Furthermore, Ti presented significantly higher levels in all the samples made -totally or partially- of polyamide (range 1,382-6,603 mg/kg) with respect to those made -totally or partially- of polyester (range 5.21-70.1 mg/kg) ($p<0.01$).

Chromium also showed high levels in samples #39 (932 mg/kg), #2 (841 mg/kg), #32 (776 mg/kg), and #17 (624 mg/kg), all corresponding to black swimsuits made of polyamide (PA) and elastane (E) (84%PA and 16%E, 81%PA and 19%E, 82%PA and 18%E, and 80%PA and 20%E, respectively) (Table 1). In fact, high levels of Cr in polyamide black garments were already found in previous studies (Matoso and Cadore, 2012; Rovira et al., 2015). Other trace elements with high individual concentrations were Al (770 mg/kg) in sample #17 (black man swimsuit, 80% polyamide and 20% elastane) and Cu (328 mg/kg) in sample #10 (blue lining of a man swimsuit, 100% polyamide). The presence of some metals, such as Co, Cu, Cr or Ni, in clothing may be due to the use of their salts as mordents to improve the dyeing process, or as metal-complex dyes directly (Chakraborty, 2011; Singh and Bharati, 2014).

Some correlations between trace elements were found, being statistically significant those between Al and Mg ($r=0.861$, $p<0.01$), Mn ($r=0.691$, $p<0.01$) and Ti ($r=0.735$, $p<0.01$), as well as between Mg and Ti ($r=0.661$, $p<0.01$), and Ti and Sn ($r=0.706$, $p<0.01$)). Interestingly, when analysing the data according to the material, only the correlation between Al and Mg was kept in polyamide fibres ($r=0.812$, $p<0.01$); suggesting that these elements could be jointly employed in this material manufacturing. It must be highlighted that the levels of both elements were significantly higher in samples made of polyamide than polyester (Fig. 1). The concentration of Al found in swimsuits - with higher levels ($p<0.05$) in polyamide (412 mg/kg) than in polyester (54.6 mg/kg)- was even much higher than that found in other garments previously analysed in our lab (Rovira et al., 2015, 2017b) (Table 4). This may be due to the use of Al oxychloride as a water repellent, both in polyester and polyamide (Bundesinstitut für Risikobewertung, 2012; Textor et al., 2001; Waeber et al., 2006), an important feature in a swimsuit. A similar trend was noticed for Co, whose levels were higher in polyamide swimsuits (34.5 mg/kg) than in other garments analysed in previous studies, where mean levels were between 0.05 mg/kg and 0.79 mg/kg (Rovira et al., 2015, 2017b).

Lead, Sr and Ti also showed significantly higher levels according to the textile material (Fig. 1). It is important to highlight the great difference between the mean values of Ti in polyester and polyamide, being much higher in garments made of polyamide (3,759 mg/kg) compared to polyester (24.1 mg/kg) swimsuits. Similar results were reported by von Goetz et al. (2013), who analysed pants and long sleeved shirts made with polyamide. The presence of Ti in polyamide fibres is due to the use of TiO_2 particles, which make the material much more hydrophobic, and protects against UV (Bossennec and Charbonneaux, 2007; Salman and Razlan, 2018; Shayestehfar et al., 2018). In addition, Ti is a delustering agent and pigment in synthetic fibres in the textile industry (Vann et al., 2009). TiO_2 is added during the polymerization phase and in the form of particles. The presence of Ti was confirmed using ESEM to analyse sample #31, a pink girl's swimsuit (80% polyamide, 20% Elastane) with high contents of Ti (6,603 mg/kg) (Fig. 2). According to the ESEM result, the particles were distributed throughout the fibres, with diameters between 178 and 430 nm.

The levels of Sb in polyester (62.8 mg/kg) were significantly higher than those detected in polyamide (14.5 mg/kg). This was already observed in previous studies conducted in our lab, in which underwear, home textiles (pyjamas or bedclothes) and t-shirts were analysed. The Sb levels of the garments made with polyester, were in the ranges 57.7- 152 mg/kg, 0.20-88.4 mg/kg, and 52.4-204 mg/kg, respectively (Rovira et al., 2017a, 2017b, 2015). The differences in Sb levels may be attributed to polyester manufacturing catalysts, in which Sb is added in the condensation phase in order to improve the reaction (James et al., 2001).

When analysing the data according to the elastane content in swimsuits ($n=22$), significant differences in the concentrations of Al, Mg, Pb, Sr, Sb and Ti were found. Although Ti was the element with the highest mean value in garments containing elastane, our finding is mainly due to fact that most samples containing elastane here analyzed, were made of polyamide (17 out of 19) ($p<0.01$). Swimsuits containing elastane showed higher levels than other samples for Al (408 vs. 10 mg/kg, $p<0.01$), Mg (1.00 vs. 0.10 mg/kg, $p<0.01$), Pb (0.12 vs. 0.04 mg/kg, $p<0.05$) and Sr (0.44 vs. 0.20 mg/kg, $p<0.05$). In contrast, Sb concentrations were significantly lower in elastane samples (22.0 vs 58.9 mg/kg, $p<0.01$), because most of the polyester samples -showing higher Sb levels- did not contain elastane (5 out of 20).

With respect to the colour of the samples, dark coloured garments (e.g., dark blue and black) showed higher concentrations of Al, Cr, Cu, Mg and Mn. However, significant differences were only noticed between dark coloured and fluorescent colours for Mg and Mn, being in both cases the highest concentrations found in dark swimsuits: 0.76 vs. 0.20 mg/kg for Mg ($p<0.05$), and 9.33 vs. 1.62 mg/kg for Mn ($p<0.05$). As above commented, polyamide dark colours showed higher Cr levels (204 to 932 mg/kg) than other swimsuits (polyamide with light colour, or polyester with dark or light colour) ($p>0.05$). The only exception was item #26 corresponding to a black polyester swimsuit with 47% of polybutylene terephthalate (PBT), with 303 mg/kg of Cr. Two out of 15 polyester swimsuits were manufactured with 47% of PBT. This kind of material is characterized by the resistance to chlorine and water repellence, characteristics that are very appropriate for swimming trunks. Chromium compounds can be used as fixing and lighting agents, especially in polyamide fibbers (Matoso and Cadore, 2012). Unlike polyester swimsuits, in polyamide ones some differences between trace elements with respect to coloration were detected. Dark coloured samples showed a significantly higher Mn concentration than light coloured garments (8.44 and 4.05 mg/kg, respectively ($p<0.05$)). Although polyester garments have higher concentrations of Sb, in the case of polyamide garments there are significant differences between colours, being higher in dark swimsuits (19.5 mg/kg) compared to fluorine colours (11.5 mg/kg, $p<0.05$) and light colours (8.30 mg/kg, $p<0.01$). There were also significant differences in the levels of Ti, in which the levels in fluorine garments (6,256 mg/kg) were almost twice the values in dark colours (3,957 mg/kg, $p<0.01$), and triple in those of light colours (2,763 mg/kg, $p<0.05$). The fact that Ti levels are so high in fluorescent coloured swimsuits could be due to the fact that these colours are more sensitive to adverse conditions, since titanium is used to protect the fibres of light and heat (Bossennec and Charbonneaux, 2007).

3.2. Human exposure and risk assessment

Dermal exposure could not be calculated for formaldehyde and aromatic amines since all levels were below their respective limit of detection. Different population groups were considered, according to the age: babies between 2 and <3 years old; children (boys and girls) between 3 and <6 years old, and between 6 and <11 years old; teenagers (boys and girls) between 11 and <18 years old; and adults (men and women) above 18 years old. Furthermore, we considered that men/boys wear either swimming briefs or board shorts and women/girls wear either bikini or swimsuit. For babies we assumed that they wear t-shirts and swimsuits. In terms of exposure time, despite two different scenarios could be considered, half-day (4h) or the whole day (8h) using the swimsuit in the beach or pool and as one scenario is a multiple of another, the exposure has only been calculated for the most hazardous situation, wearing a swimsuit during 8 hours.

In general, the exposure was higher in women and girls (child or teenager) than in men and boys, as well as higher in younger groups (Fig. 3). Babies between 2 and <3 years old presented the highest exposure levels for each element, due the higher ratio between surface area (trunk, genitals and buttocks) and body weight, in addition, we assume that babies wear a T-shirt. Exposure levels for all elements for babies were approximately one order of magnitude higher than adult male exposure, which shown the minimum exposure levels in each age group. Obviously, exposure is higher in men or boys (child or teenager) wearing board shorts than those wearing swimming briefs and in women wearing swimsuits than those wearing bikini. In both cases, this difference is related to the area of skin covered by the swimwear, a swimsuit covers more skin than a brief and a swimsuit than a bikini, in the same way that a woman's or girl swimsuit covers more skin than a man or boy’s board short. Titanium was the trace element with the highest mean exposure in all population groups, being the group of babies between 2 and <3 years old the group with highest exposure ($1.31 \cdot 10^{-4}$ mg/kg/d) and adult men the lowest exposure levels ($4.41 \cdot 10^{-5}$ mg/kg/d).

The HQ was used to assess the non-carcinogenic risks derived from dermal exposure. All values were far below the limit value, which is set to 1, indicating that analysed trace elements exhibited non-carcinogenic risks. The highest mean HQ levels were Sb for all groups (HQ=0.04).

Carcinogenic risks were only calculated for As, Cr and Pb, since they are the only detected elements for which an oral SF is established (RAIS, 2018), (Fig. 4). Babies are the population group with the highest cancer risk for the three elements, with Cr being the element with the highest cancer risk, $1.44 \cdot 10^{-5}$ ($1.91 \cdot 10^{-8}$ for As and $4.38 \cdot 10^{-11}$ for Pb). In the other population groups, Cr is also the element with the highest risk, especially for, child girls from 3 to 6 years old and from 6 to 11 years old, who wear swim suit a ($1.23 \cdot 10^{-5}$ and $1.00 \cdot 10^{-5}$, respectively).

Although Ti has been -by far- the trace element found at the highest concentration, risk assessment could not be carried out due to the lack of toxicological data. Obviously, it does not mean that Ti is not dangerous to health. For example, studies in rat and *in vitro* investigations have demonstrated the negative influence of TiO_2, used in the textile industry, on the reproductive system: spermatogenesis (Fartkhooni et al., 2013), oogenesis (Hou and Zhu, 2017) and suppression of embryonic bone development (Hong et al., 2017). Therefore, in order to establish the potential risk for reproduction of clothes with an important skin contact with reproductive organs, it is important to characterize accurately all the parameters that can affect human exposure to Ti-TiO_2 such as particle size, migration from the fabric, and skin absorption.

As above commented, in our laboratory we have conducted several studies regarding exposure to trace elements through clothes and textiles (Rovira et al., 2017a, 2017b, 2015). For adult men, a cumulative exposure to trace elements approach due to dermal contact, using previous published data (day time clothes, pyjamas and bedclothes while sleeping), was added to swimsuits for two scenarios: summer (wearing swimsuits for 8h/d) and rest of the year (wearing swimsuits 1 h/d). In general terms, exposure through swimsuits was negligible (<10%) with the exceptions of As, Co, Cr, Mo Sn, Ti and V (Fig. 5). In the summer scenario (wearing swimsuit for 8 h/d), As was only detected in swimsuit samples with a contribution of 100%, while the other trace elements contributed between 40 and 65% for Co, Cr, Sn and Ti, and around 20% for Mo and V. However, during rest of the year, assuming only wearing swimsuits 1 h/d, the contribution of swimwear to the total was above 10% only for As (100%), Sn (15%), and Ti (25%).

4. Conclusions

The levels of formaldehyde and aromatic amines were below their respective detection limits for the 39 swimsuits analyzed. Regarding trace elements, high levels of Ti were found especially in polyamide fabrics, while high concentrations of Sb were detected in polyester, probably due to its use as catalyst and delustering agent in these fibres. Dark swimsuits had higher levels of Al, Cr, Mg and Mn than other colours, being remarkable the presence of high concentrations of Cr in black polyamide samples. The times of use, and the surface of skin covered, were two important factors influencing dermal exposure. Titanium was the element with the highest exposure. Non-carcinogenic risks for dermal exposure to trace elements through swimwear were at acceptable levels (HQ < 1). Carcinogenic risks for As and Pb were below acceptable limit (10^{-5}), while Cr(VI) levels were close -or even slightly higher- than this threshold. Despite human health risks could not be estimated for Ti due the lack of toxicological data, the human health risks of this element could not be dismissed. Consequently, further investigations are needed in order to assess the ability of Ti in the migration from the fabric to the skin, as well as to test its potential adverse effects.

A recommendation to minimize the exposure to trace elements contained in swimsuits would be the use of clothes made of natural fibres, such as cotton, instead of clothing exclusively made of synthetic fibres. Unfortunately, there is a lack of natural alternatives in the market, while cotton is less attractive in terms of weight gain or drying time when it is wet. In any case, the textile industry is urged to develop alternative materials which reduce the potential exposure to chemical additives, not only to trace elements but also to a wide range of organic compounds.

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Table 1. Main characteristics of the swimsuits

№	Shop place	Material	Colour	Made in	Density
1	Supermarket	81% P; 19% E	Dark blue	Vietnam	193
2	Supermarket	81% PA; 19% E	Black	Vietnam	217
3	Supermarket	85% PA; 15% E	Green (stamping)	China	194
4	Supermarket	82% PA; 18% E	Purple	Cambodia	240
5	Chain Store	80% PA; 20% E	Dark blue	Vietnam	205
6	Supermarket	85% P; 15% E	Pink + white	China	202
7	Chain Store	82% PA; 18% E	Grey	Vietnam	193
8	Supermarket	85% PA; 15% E	Blue (stamping)	China	223
9	Supermarket	82% PA; 18% E	Blue	China	234
10	Chain Store	100% PA	Blue	Albania	107
11	Chain Store	100% PA	Pink	Sri Lanka	101
12	Supermarket	85% PA; 15% E	Green + white	China	198
13	Chain Store	82% PA; 18% E	Grey	Bangladesh	219
14	Chain Store	100% P	Red	Vietnam	131
15	Chain Store	100% P	Fluorescent yellow	Vietnam	122
16	Chain Store	100% P	Fluorescent blue	Vietnam	124
17	Chain Store	80% PA; 20% E	Black	Tunisia	189
18	Supermarket	100% P	Fluorescent yellow	China	116
19	Chain Store	100% P	Dark blue	Vietnam	143
20	Supermarket	100% P	Black	China	99
21	Supermarket	100% P	Fluorescent orange	China	108
22	Supermarket	100% P	Blue	China	107
23	Chain Store	100% P	Black	Spain	127
24	Chain Store	100% P	Fluorescent orange	Vietnam	216
25	Chain Store	100% P	Fluorescent yellow	Bangladesh	88
26	Chain Store	100% P (47% PBT)	Black	Cambodia	145
27	Chain Store	80% P; 20% E	Navy blue	Cambodia	192
28	Chain Store	100% P	Blue	Albania	213
29	Chain Store	100% P (47% PBT)	Black	Cambodia	217
30	Chain Store	80% PA; 20 E	Fluorescent pink	China	203
31	Chain Store	82% PA; 18% E	Black	China	220
32	Chain Store	82% PA; 18% E	Navy blue	China	210
33	Chain Store	72% PA; 28% E	White	Morocco	126
34	Chain Store	95% P; 5% E	Black	Spain	93
35	Chain Store	88% P; 12% E	White	China	145
36	Supermarket	100% P	Red	Bangladesh	206
37	Supermarket	80% P; 20% E	Navy blue + red + white	China	215
38	Chain Store	83% PA, 17% E	Black	Myanmar	349
39	Chain Store	84% PA, 16% E	Cream	Spain	120

[a]Density in g/m^2; E: Elastane; P: Polyester; PA: Polyamide; PBT: polybutylene terephthalate.

Table 2. Parameters used to assess dermal exposure

Variable	Description	Value	Reference
F_{cloth}	Weight fraction of substance in garments	Table 3	Present study
d_{cloth}	Clothing density	Table 1	Present study
A_{skin}	Men slip swimsuit (Genitals and Buttocks)	1498 cm^2	US EPA, 2011
	Men thighs swimsuit (Genitals and Buttocks + thighs)	2765 cm^2	
	Women bikini (Bosom + Genitals and Buttocks)	2032 cm^2	
	Women swimsuit (Trunk)	6540 cm^2	
	Babies 2-<3 years old t-shirt + swimsuit (Trunk + genitals and buttocks)	2930 cm^2	
	Child boy 3-6 years old slip swimsuit (Genitals and Buttocks)	526.3 cm^2	
	Child boy 3-6 years old thighs swimsuit (Genitals and Buttocks + thighs)	1670 cm^2	
	Child girls 3-6 years old swimsuit (trunk)	3130 cm^2	
	Child boy 6-11 years old slip swimsuit (Genitals and Buttocks)	750 cm^2	
	Child boy 6-11 years old thighs swimsuit (Genitals and Buttocks + thighs)	2575 cm^2	
	Child girls 6-11 years old swimsuit (trunk)	4280 cm^2	
	Teenager boy 11-16 years old slip swimsuit (Genitals and Buttocks)	1117 cm^2	
	Teenager boy 11-16 years old thighs swimsuit (Genitals and Buttocks + thighs)	3957 cm^2	
	Teenager girl 11-16 years old bikini (Bosom + Genitals and Buttocks)	3195 cm^2	
	Teenager girl 11-16 years old swimsuit (Trunk)	6300 cm^2	
F_{mig}	Fraction of substance migrating to skin	0.005	Present study Bundesinstitut für Risikobewertung, 2012
$F_{contact}$	Fraction of contact area for skin	1	Bundesinstitut für Risikobewertung, 2012
F_{pen}	Fraction of penetration inside the body	0.01 0.03 for As	US EPA, 2016
$T_{contact}$	Contact duration between skin-textile	0.33 (8h/24h)	Assumed
n	Mean number of events per day	1/d	Assumed
BW	Adult male	70	Assumed
	Adult female	60	Assumed
	Baby 2<3	13.8	Sobradillo et al., 2000
	Child boy 3<6	19.5	
	Child girl 3<6	17.2	
	Child boy 6<11 years	27.9	
	Child girl 6<11 years	29.0	
	Teenager boy 11<16	54.1	
	Teenager girl 11<16	48.7	

Table 3. Concentrations (mg/kg) of trace elements in swimsuits samples

n=39	% of detection	Mean	SD	Minimum	Maximum
Ag	3	0.04	0.00	<0.02	0.04
Al	90	249	286	<0.42	770
As	8	0.14	0.02	<0.04	0.16
B	18	10.8	16.2	<0.21	43.7
Ba	92	0.52	0.54	<0.04	2.93
Bi	13	0.05	0.02	<0.02	0.08
Co	41	15.5	31.0	<0.02	111
Cr	97	122	253	<0.04	932
Cu	64	27.9	75.7	<0.15	328
Mg	69	0.87	0.55	<0.02	1.62
Mn	100	6.42	9.89	0.06	38.9
Mo	21	0.12	0.11	<0.02	0.40
Ni	44	0.63	0.57	<0.04	2.02
Pb	31	0.21	0.12	<0.04	0.47
Sb	97	45.4	42.7	<0.04	167
Sn	28	1.09	1.38	<0.04	3.75
Sr	85	0.51	0.74	<0.04	4.13
Ti	100	1844	2161	5.21	6603
V	28	0.17	0.20	<0.04	0.79
Zn	62	4.25	4.74	<0.42	24.6

Be, Cd, Fe, Hg, Sc, Se, Sm and Tl concentrations were below their respective detection limits in all the samples.

Table 4. A summary of concentrations (mean and range) of trace elements in textiles from various studies.

	Menezes et al., 2010	Matoso and Cadore, 2012	Rovira et al., 2015	Rovira et al., 2017b	Rovira et al., 2017a	Present study	
	Coloured textile	Polyamide	Cotton, polyester, polyamide and viscose	Cotton, polyester and polyamide	Cotton, polyester and polyamide	Polyester	Polyamide
Ag	-	-	-	0.02 (ND - 0.13)	0.05 (<ND - 0.57)	0.04 (0.04 - 0.04)	ND
Al	38.4 (17.6 - 118)	-	31.8 (1.37 - 198)	14.7 (ND - 108)	28.5 (1.68 - 351)	54.6 (0.68 - 519)	412 (14.3 - 770)
As	-	ND	ND	ND	ND	ND	0.14 (0.12 - 0.16)
B	-	-	0.45 (ND - 2.26)	0.64 (ND - 3.93)	0.30 (ND - 1.27)	1.08 (0.30 - 2.53)	23.9 (0.59 - 43.9)
Ba	14.7 (2.05 - 87.4)	-	1.51 (ND - 9.46)	0.96 (ND - 7.20	2.13 (ND - 19.1)	0.36 (0.08-0.70)	0.71 (0.15 - 2.9)
Be	-	-	ND	ND	ND	ND	ND
Bi	-	-	0.28 (ND - 1.96)	0.01 (ND - 0.08)	0.02 (ND - 0.10)	0.03 (0.02 - 0.03)	0.08 (0.08 - 0.08)
Cd	-	0.2 (ND - 0.2)	0.01 (ND - 0.04)	ND	0.01 (ND - 0.07)	ND	ND
Co	-	ND	0.21 (ND - 5.18)	0.05 (ND - 1.20)	0.79 (ND - 20.4)	0.61 (0.02 - 5.01)	34.5 (0.12 - 111)
Cr	6.39 (ND - 1.61)	234.1 (0.3 - 965)	19.8 (ND - 605)	6.55 (ND - 374)	45.7 (ND - 754)	16.2 (0.12 - 303)	228 (0.13 - 932)
Cu	31.1 (ND - 273)	2.77 (2.2 - 4.0)	20.1 (ND - 287)	32.8 (ND - 1065)	72.8 (ND - 439)	3.67 (0.15 - 39.8)	50.3 (0.13 - 328)
Fe	28.5 (12.1 - 66.1)	-	9.78 (3.38 - 35.1)	12.9 (ND - 40.7)	18.0 (1.09 - 194)	ND	ND
Hg	-	ND	ND	0.04 (ND - 0.13)	ND	ND	ND
Mg	-	-	129 (3.75 - 716)	142 (ND - 889)	114 (ND - 832)	0.41 (0.02 - 1.41)	1.14 (0.38 - 1.62)
Mn	-	-	1.82 (0.10 - 13.3)	0.91 (ND - 7.68)	1.05 (0.07 - 5.77)	6.42 (0.06 - 38.9)	6.41 (1.60 - 14.9)
Mo	-	-	0.03 (ND - 0.38)	0.02 (ND - 0.16)	0.03 (ND - 0.16)	0.07 (0.02 - 0.11)	0.14 (0.05 - 0.40)
Ni	0.69 (ND - 5.10)	1.95 (0.9 - 3.3)	ND	0.19 (ND - 1.20)	0.16 (ND - 1.79)	0.40 (0.05 - 1.24)	0.83 (0.24 - 2.02)
Pb	-	0.31 (ND - 1.4)	0.13 (0.03 - 0.32)	0.13 (ND - 0.90)	0.16 (ND - 1.90)	0.16 (0.04 - 0.31)	0.23 (0.10 - 0.47)
Sb	-	ND	22.3 (ND - 204)	26.9 (ND - 202)	26.0 (ND - 152)	76.4 (16.7 - 167)	14.5 (3.26 - 29.2)
Sc	-	-	0.18 (ND - 0.72)	ND	0.19 (ND - 0.93)	ND	ND
Se	-	-	0.08 (ND - 0.61)	ND	ND	ND	ND
Sm	-	-	0.01 (ND - 0.04)	ND	ND	ND	ND
Sn	-	-	0.05 (ND - 0.20)	0.05 (ND - 0.35)	0.05 (ND - 0.29)	0.19 (0.08 - 0.39)	1.43 (0.11 - 3.75)
Sr	-	-	2.89 (0.05 - 15.1)	5.37 (ND - 33.8)	2.21 (ND - 10.1)	0.22 (0.04 - 0.62)	0.74 (0.15 - 4.13)
Ti	-	-	-	10.9 (ND - 124)	8.34 (ND - 37.8)	24.1 (5.21 - 70.1)	3759 (1382 - 6603)
Tl	-	-	ND	ND	0.01 (ND - 0.03)	ND	ND
V	-	-	ND	0.11 (ND - 0.57)	ND	0.08 (0.04 - 0.21)	0.24 (0.10 - 0.79)
Zn	3.18 (ND - 10.9)	-	12.1 (ND - 256)	1.57 (ND - 16.4)	36.4 (ND - 1185)	3.48 (0.83 - 8.52)	4.91 (1.35 - 24.6)

ND: Not detected

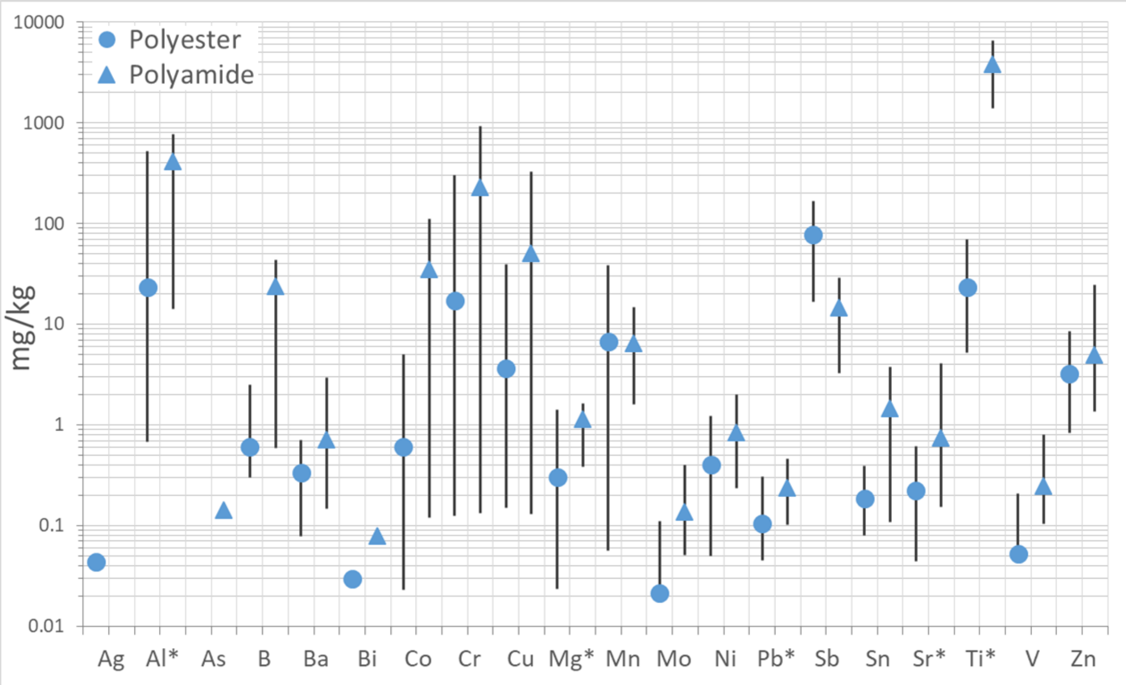
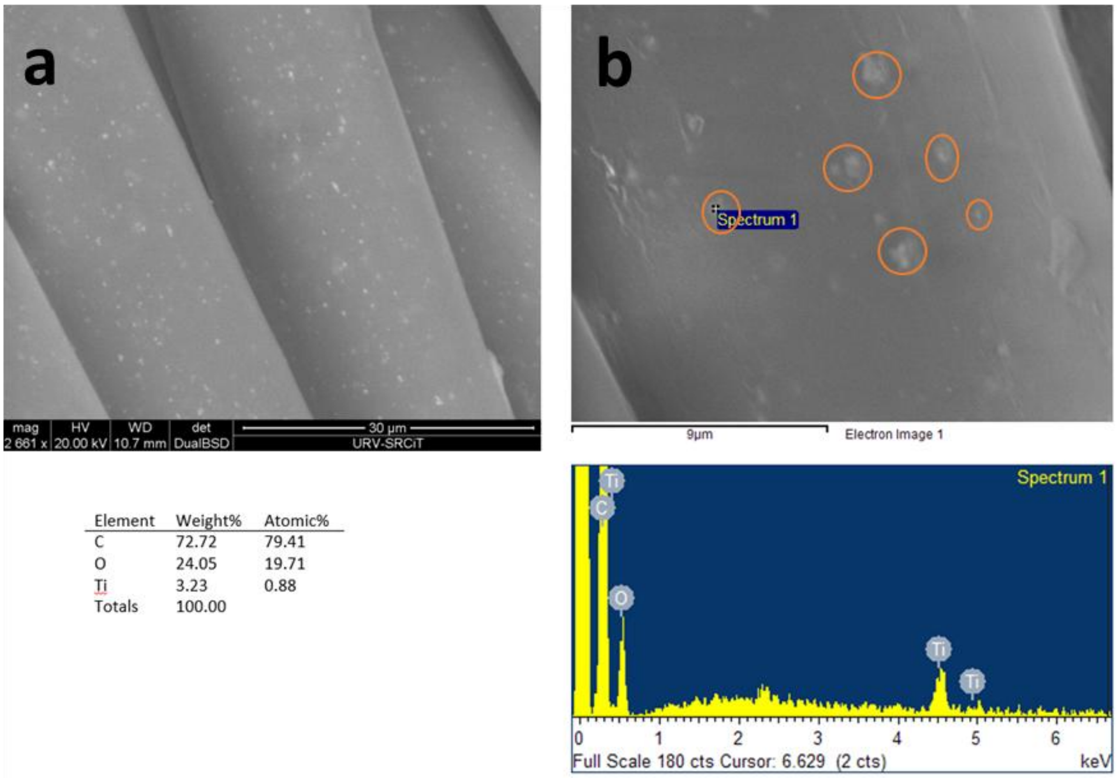


Fig. 1. Concentrations of trace elements (mg/kg) according to the swimsuit material. Blue symbols are mean levels, while black lines are minimum and maximum ranges. Ag in polyamide samples and As in polyester samples were below their respective detection limits. Be, Cd, Fe, Hg, Sc, Se, Sm and Tl levels were below their respective detection limits in all samples. Asterisks (*) indicate significant differences according to the material ($p<0.05$).



Element	Weight%	Atomic%
C	72.72	79.41
O	24.05	19.71
Ti	3.23	0.88
Totals	100.00	

Fig. 2. Backscattering detector (BSD) image (a), and electron microscope image (b) of TiO_2 particles (inside orange circles) found in sample #31. Spectrum 1 indicates where the microanalysis was performed.

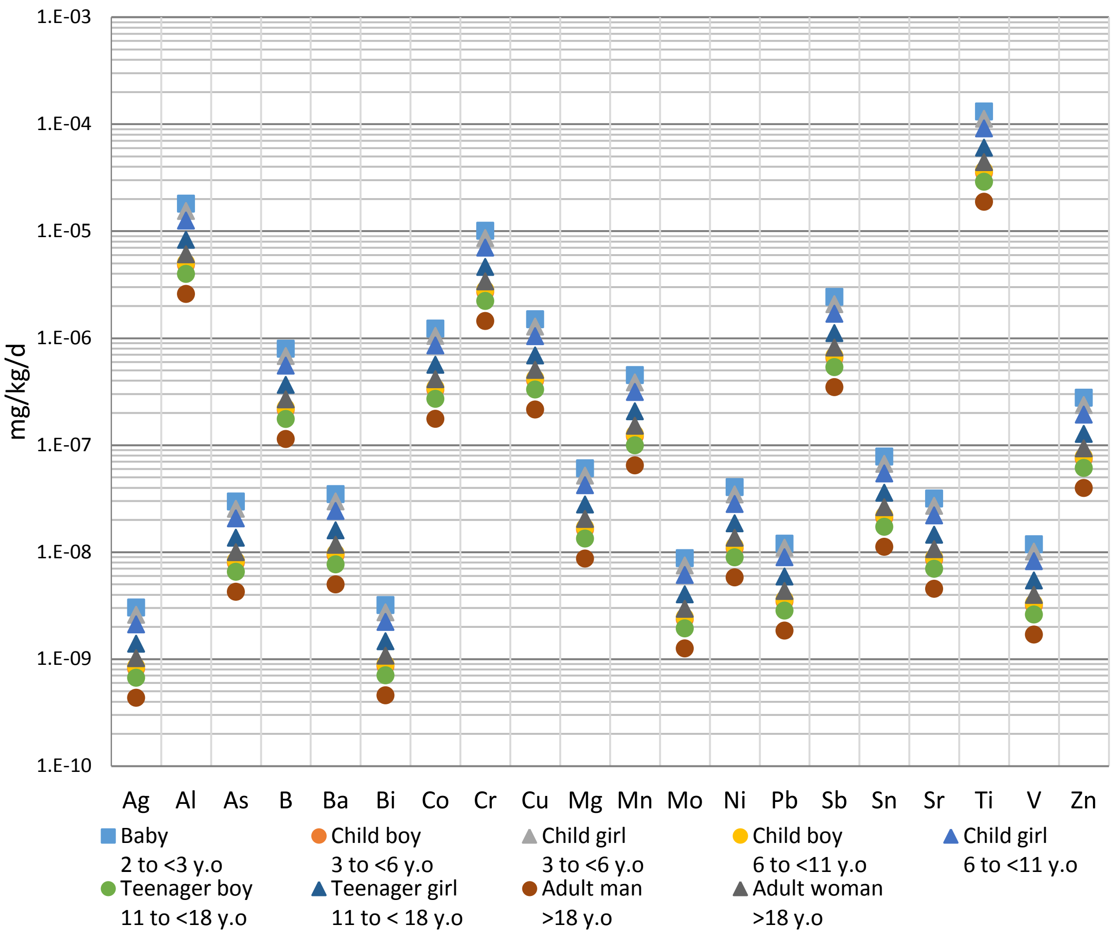


Fig. 3. Dermal exposure (mg/kg/d) due to the use of swimming suits for 8 hours a day for the different groups of population. For males (from child, teenager and adult man) mean exposure between board short and briefs swimsuits and for females (teenager and adult woman) mean exposure between bikini and swimsuit were plotted.

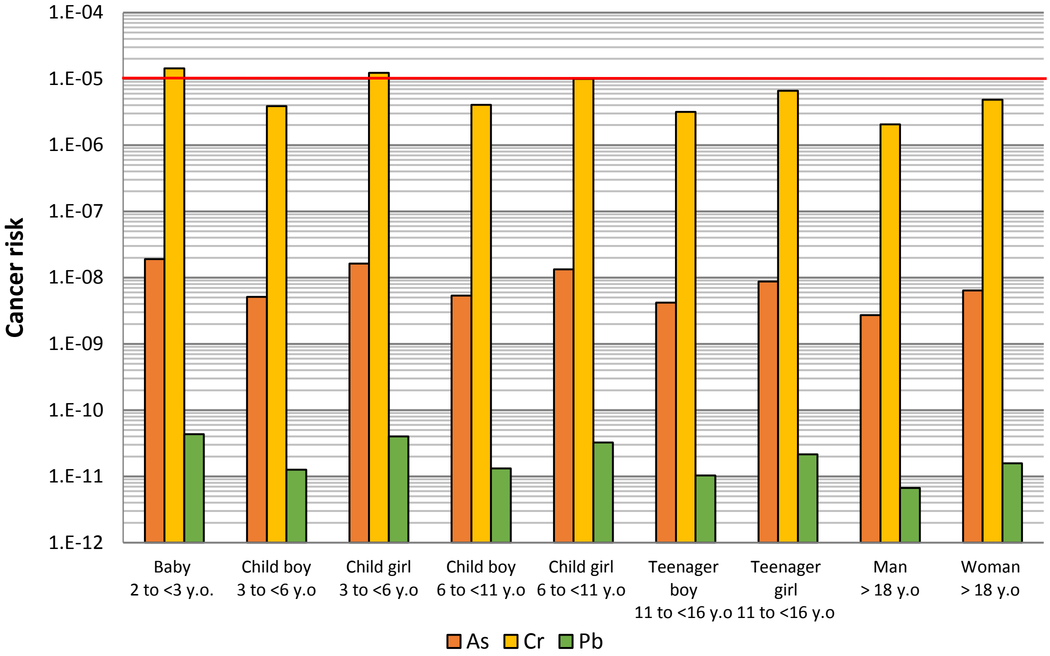


Fig. 4. Cancer risks associated to exposure to trace elements for wearing swimwear.

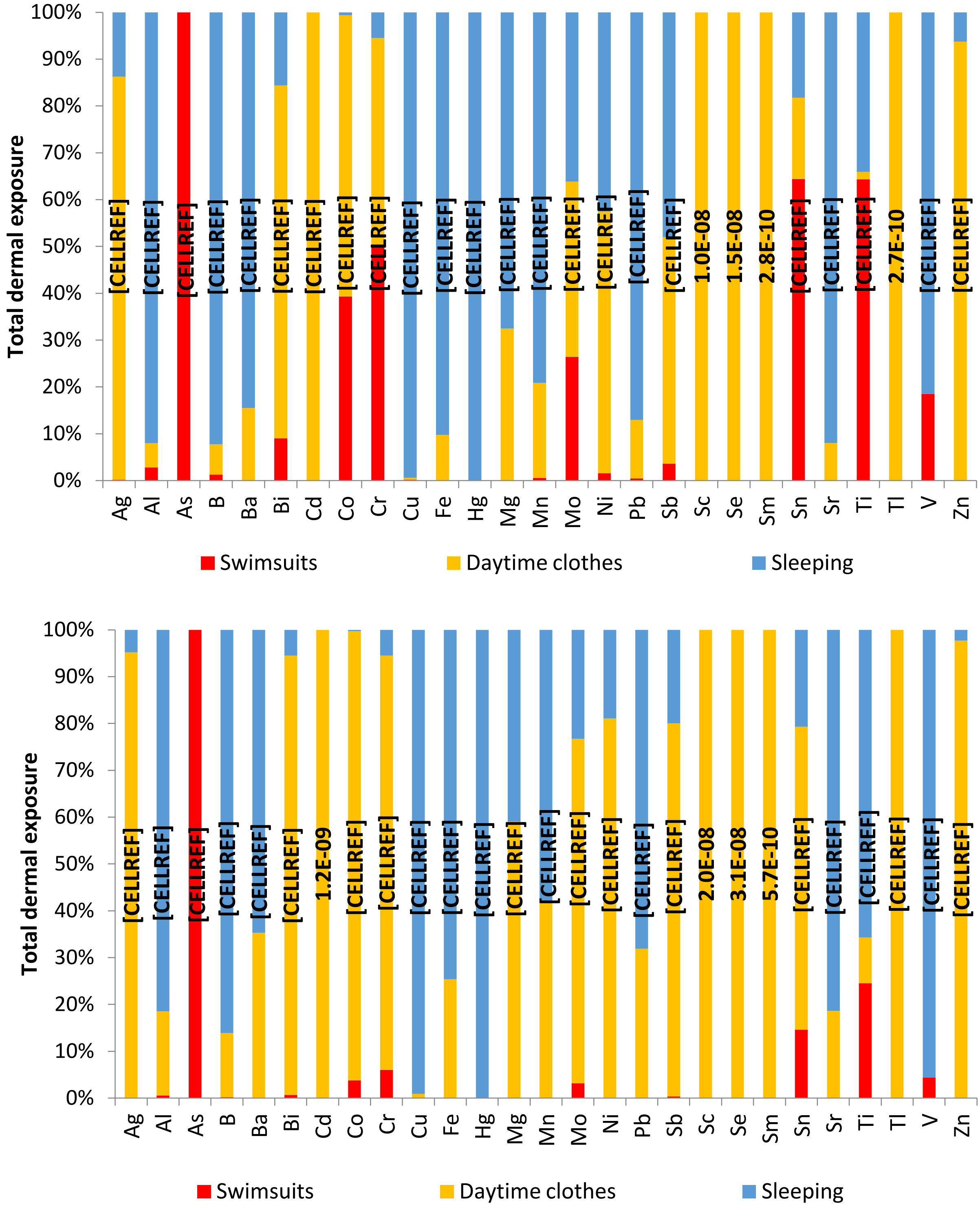


Fig. 5. Contribution (in percentage) of different types of clothes (day time clothes, swimsuits and sleeping wear (pyjamas and bed clothes)) to the total dermal exposure to trace elements during a) summer (8 h/d wearing swimsuits), and b) rest of the year (1 h/d wearing swimsuits). Numbers indicate the total exposure level (mg/kg/day).

***conflict of Interest Statement**

Declaration of interests

XX The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

There is not any potential conflict of interests in this research/manuscript.