

Clothing as a potential exposure source of trace elements during early life

Marta Herrero ^{a,b}, Joaquim Rovira ^{a,b,c,*}, Neus González ^{a,b,c}, Montse Marquès ^{a,b}, Fernando Barbosa ^d, Jordi Sierra ^e, José L. Domingo ^{a,b}, Martí Nadal ^{a,b},
Marília Cristina Oliveira Souza ^{a,b,d}

^a Laboratory of Toxicology and Environmental Health, School of Medicine, IISPV, Universitat Rovira i Virgili, Sant Llorenç 21, 43201 Reus, Catalonia, Spain

^b Institut d'Investigació Sanitària Pere Virgili (IISPV), 43204 Reus, Catalonia, Spain

^c Environmental Engineering Laboratory, Departament d'Enginyeria Química, Universitat Rovira i Virgili, Av. Països Catalans 26, 43007 Tarragona, Catalonia, Spain

^d University of Sao Paulo, School of Pharmaceutical Sciences of Ribeirao Preto, Department of Clinical Analyses, Toxicology and Food Sciences. Analytical and System Toxicology Laboratory, Avenida do Café s/nº, 14040-903, Ribeirao Preto, Sao Paulo, Brazil

^e Faculty of Pharmacy, University de Barcelona, Joan XXIII Avenue s/n, 08028 Barcelona, Catalonia, Spain

*Corresponding author.

E-mail: joaquin.rovira@urv.cat (J. Rovira)

31 ABSTRACT

32

33 In recent years, the interest in determining the chemical composition of textile products
34 has increased among the scientific community and regulatory agencies, driven by
35 toxicological issues and environmental concerns. Chemical elements are naturally present
36 in clothing as natural fibers or intentionally added during manufacture. Some of them
37 show physical-chemical properties that allow their absorption through the skin. In
38 addition, chronic situations increase the dermal exposure capacity. Because of age-
39 specific behavioral characteristics and underdeveloped physiological function, children
40 may be especially sensitive to exposure to trace elements. This study aimed to analyze
41 the levels of twenty trace elements in 120 clothing items commercialized in Spain. Textile
42 products for pregnant women and children <36 months old were included. The potential
43 health implication of this dermal exposure in early life was also evaluated. Aluminum,
44 zinc, and titanium showed the highest concentrations, with median levels of 27.6, 5.6, and
45 4.2 mg/kg, respectively. Since chromium is employed as a metal complex dye in synthetic
46 fibers, high levels of this element were found in black polyester. Dermal exposure to
47 titanium, which is a ubiquitous element in clothes made of synthetic fibers, was associated
48 with a hazard quotient (HQ) higher than the threshold value (HQ>1), with values of 1.13
49 for pregnant women and 1.22 for newborns. On the other hand, HQ values of other
50 elements and cancer risks were lower than the recommended values. Assessing early-life
51 exposure to toxic elements can help to identify potential sources and to prevent or reduce
52 human exposure, mainly in vulnerable groups.

53

54 *Keywords:* clothes, textile, dermal exposure, trace elements, ICP-MS.

55

56

57 **1. Introduction**

58 Dietary intake is the most important source of prenatal exposure to toxicants during
59 pregnancy. Chemicals may accumulate and cross the placenta, exposing the fetus to a
60 mixture of hazardous substances (Bocca et al., 2019; Souza et al., 2020; Urbano et al.,
61 2022). After birth, early exposure continues through the ingestion of breast milk, which
62 may also contain traces of chemicals (Oliveira et al., 2020; Castro et al., 2021; Rovira et
63 al., 2022). In addition, other potential routes, such as air inhalation and skin contact, may
64 also contribute to the uptake of toxicants (Rovira et al., 2015; Herrero et al., 2019; Herrero
65 et al., 2020; Souza et al., 2022).

66 Usually, the skin acts by inhibiting the chemical entrance into the human body.
67 However, some toxic elements present physical-chemical properties that allow their
68 absorption through the skin, with long-term exposure increasing dermal exposure
69 capacity. Dermal absorption is identified as one of these minor exposure pathways.
70 Notwithstanding, the relevance of this route versus other significant sources has not been
71 well assessed, especially in some population subgroups, such as pregnant women and
72 newborns (Rovira and Domingo, 2019; Chen et al., 2022; Souza et al., 2023a,b).

73 In recent years, there has been an increasing trend to use new chemical additives in
74 clothing in the textile industry. Most of these substances are intentionally added to
75 provide additional properties to textile materials, improving their elasticity and
76 breathability (Rovira and Domingo, 2019; Filella et al., 2022). Among them, trace
77 elements may be used as catalysts during fabric manufacture, antimicrobials, water
78 repellents, odor-preventive agents, and complex metal dyes (Sarayu and Sandhya, 2012;
79 Rovira et al., 2015; Rovira and Domingo, 2019; Sima, 2022). For example, approximately
80 80% of leathers are chrome-tanned. Typically, textile dyeing use elements like vanadium
81 (V), chromium (Cr), barium (Ba), lead (Pb), copper (Cu), cobalt (Co), and nickel (Ni),
82 while iron (Fe) and manganese (Mn) are used in bleaching processes (Rujido-Santos et
83 al., 2022). Polyester fibers usually contain traces of antimony (Sb), resulting in their use
84 as a catalyst during polyester polymerization and as a flame-retardant synergist (Rovira
85 et al., 2015; Rovira et al., 2017b; Herrero et al., 2019; Biver et al., 2021).

86 Chemical elements may be toxic to human health at high levels or also in chronic
87 exposure situations, being able to bioaccumulate and form complexes with biological
88 enzymes. Essential elements (i.e., zinc (Zn), Cu, and Mn) show human toxicity when their
89 concentrations exceed the threshold. In addition, when elements assume different

90 chemical forms, their toxicity can be drastically altered. Pb, cadmium (Cd), and Ni may
91 induce carcinogenic events or multi-organ toxicity (Batista et al., 2009; Jaishankar et al.,
92 2014; Rovira et al., 2015; Letinic et al., 2016; Gupta et al., 2019, Chen et al., 2022; Kim
93 et al., 2022; Urbano et al., 2022).

94 Because they do not own entirely developed biological systems, newborns and
95 children are particularly vulnerable to environmental pollutants. In addition, they have a
96 higher food intake per body weight in comparison to adults. Early exposure to metals has
97 been associated with birth outcomes, including preterm birth and impaired fetal growth
98 (Souza et al., 2022). A number of studies confirm the presence of high levels of metals in
99 the body of children (Jaishankar et al., 2014; Chen et al., 2022; Urbano et al., 2022; Souza
100 et al., 2020; Zheng et al., 2023).

101 The Developmental Origins of Health and Disease (DOHaD) hypothesis postulates
102 that exposure to some toxic elements influences the critical development and growth,
103 including fetal and perinatal stages, which may significantly affect an individual's short-
104 and long-term health (Mandy and Nyirenda, 2018). Overall, prenatal and postnatal
105 periods are probably critical windows susceptible to environmental chemical exposure
106 predisposing to non-communicable diseases (Sanders et al., 2018). Concerning this, it is
107 essential to identify and estimate the contribution of all the exposure pathways to toxic
108 metals, either individually or as chemical mixtures. Various metals have been associated
109 with neurodevelopmental disorders. Adverse cognitive function and neurotoxicity effects
110 have been observed after Pb and mercury (Hg) exposure (Chen et al., 2022; Urbano et al.,
111 2022).

112 Chemical elements can occur naturally in the fibers structure (Velusamy et al.,
113 2021). However, applying natural dyes for coloring fibers may be more beneficial
114 compared to synthetic dyes, as the latter can cause toxic effects due to their toxic metal
115 content (Rana et al., 2022). The textile industry is one of the most polluting industrial
116 sectors, being responsible of the release of a wide variety of chemical pollutants into the
117 environment. Regarding this, the use of organic cotton has been demonstrated to
118 contaminate much less than other conventional fabrics, being a sustainable option (Hasan
119 et al., 2022; Goyal and Parashar, 2023). Life cycle assessment studies indicate that
120 organically grown cotton has notable potential impact savings in terms of global warming,
121 acidification, eutrophication, blue water consumption, and primary energy demand (PE
122 International, 2014; Avadí et al., 2020). Unfortunately, and with a few exceptions focused
123 on a selective group of substances (Herrero et al., 2022a,b), there is no reliable

124 information on the health benefits of wearing organic cotton instead of regular cotton
125 regarding the presence of chemical pollutants in the fabrics.

126 The present study aimed to analyze the concentration of twenty trace elements,
127 including both toxic and essential, in the clothing of pregnant women, newborns, and
128 toddlers commercialized in Spain. The dermal absorption of these chemicals through the
129 same skin-contact clothes was also estimated. The associated health risks were also
130 assessed.

131

132 **2. Materials and methods**

133

134 *2.1. Sample selection*

135 Textile samples, currently available in the Spanish market, were purchased in
136 April 2021. Up to 120 clothing items for pregnant women and children <36 months old
137 were bought in hypermarkets, chain stores, small retailers of Tarragona County
138 (Catalonia, Spain), and online shops. The whole set included skin-contact clothing for
139 pregnant women (n=40; i.e., elastic T-shirts, jeans/trousers/leggings, bras and panties),
140 for newborns aged <12 months old (n=30; i.e., bodysuits, pyjamas, and socks), and for
141 toddlers ranging 12–36 months (n=50; i.e., pyjamas, underwear, T-shirts, dresses, and
142 trousers/leggings). The percentage of clothes, according to coloring, fiber material,
143 official certification, place of purchase, and country of manufacture, are summarized in
144 Table 1. Table S1 (Supplementary Information) provides complete information on each
145 sample. Around 50% of the samples were organic, made either of organic cotton or
146 chemical-free. After being purchased, all samples were immediately stored at room
147 temperature until their analysis.

148

149 *2.2. Microwave digestion*

150 Sample preparation for determining trace chemical elements was separately
151 performed according to the fiber type of the manufactured cloth. For cotton samples
152 (natural fibers with >80% of cotton), an amount of 0.3 g of each garment was completely
153 digested with 6 mL of HNO₃ (65% Suprapur, E. Merck, Darmstadt, Germany) and 3 mL
154 of H₂O₂. In turn, synthetic samples made of polyamide or polyester, or mixed samples
155 containing <80% of cotton, were subjected to acid digestion. An amount of 0.2 g of the
156 garment was treated with 1.5 mL of H₂SO₄ (96% Suprapur, E. Merck), and 6.5 mL of
157 HNO₃ (65% Suprapur, E. Merck). Irrespective of the material, all samples were digested
158 in a Milestone Start D Microwave Digestion System for 5 min at 105°C, 5 min at 150°C,

159 5 min at 180°C, and finally 10 min at 200°C, performing temperature exchange in 10-
160 minute ranges. After cooling, extracts were filtered and brought to a volume of 25 mL
161 with distilled water.

162

163 2.3. Instrumental analysis

164 The levels of aluminum (Al), antimony (Sb), arsenic (As), barium (Ba), bismuth
165 (Bi), boron (B), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), lead (Pb),
166 manganese (Mn), mercury (Hg), molybdenum (Mo), nickel (Ni), silver (Ag), strontium
167 (Sr), tin (Sn), titanium (Ti) and zinc (Zn) were determined by inductively coupled plasma
168 mass spectrometry (ICP-MS, Perkin Elmer Elan 6000). Standards, blanks, and replicates
169 of the samples were included during the analysis. The limits of detection (LOD) were the
170 following: 0.04 µg/g for Ag; 0.08 for Ba, Bi, Cd, Co, Mo, Pb, and Sn; 0.17 µg/g for As,
171 Cu, Cr, Hg, Ni, and Sr; 0.42 µg/g for Ti; 0.83 µg/g for B, Mn, and Sb; 1.67 µg/g for Al;
172 and 1.83 µg/g for Zn.

173

174 2.4. Exposure and health risk assessment

175 Concentration data of the trace elements in clothing were used to assess dermal
176 exposure and the associated health risks. Exposure was estimated for three different
177 groups: pregnant women, newborns (<12 months old), and toddlers (boys and girls, 12–
178 36 months old) based on the following equation (Eq. 1) developed by the European
179 Chemical Agency (ECHA, 2016):

180

181 Eq. 1:
$$\text{Exp}_{\text{derm}} = \frac{F_{\text{cloth}} \times d_{\text{cloth}} \times A_{\text{skin}} \times F_{\text{mig}} \times F_{\text{contact}} \times F_{\text{pen}} \times T_{\text{contact}} \times n}{\text{BW}}$$

182

183 Exp_{derm} is the dermal exposure expressed in mg/(kg·day), F_{cloth} is the fraction
184 of element in clothes (mg/mg), d_{cloth} is the density of the clothing (mg/cm²), A_{skin} is
185 the skin area covered by the clothing (cm²), F_{mig} (%) is the migration fraction of
186 substance from cloth to skin, F_{contact} is the fraction of contact area for skin
187 (dimensionless), F_{pen} is the penetration rate of the element (dimensionless), T_{contact} is
188 the duration of the clothing skin contact (day), n is the number of events per day (1/day),
189 and BW is the body weight (kg). All dermal exposure parameters are summarized in Table
190 S2 (*Supplementary Information*).

191 Non-carcinogenic risks were determined by calculating the hazard quotient (HQ).
192 HQ corresponds to the quotient between exposure and the dermal reference dose (RfDd).
193 The carcinogenic risk was calculated by multiplying the exposure by the respective
194 dermal slope factor (SFd). RfDs and SFs were obtained from the Regional Screening
195 Level from the US EPA Preliminary Remediation Goals (US EPA, 2021).

196

197 2.5. Data analysis

198 Data analysis was performed using the IBM SPSS Statistics Software® version
199 28.0 (IBM Corp. Released 2020, Armonk, NY, USA). For statistical purposes, results
200 below the respective LODs were assumed to equal one-half of that limit ($ND = \frac{1}{2}LOD$).
201 The Kolmogorov–Smirnov test was used to assess the distribution of the values. The
202 Kruskal–Wallis tests for non-parametric data were used to assess group differences. The
203 effect size (Epsilon Squared - ϵ^2) approach assessed the statistical difference between
204 groups. The results with ϵ^2 values higher than 0.15 showed significance ($\epsilon^2 < 0.01$ - Very
205 small; $0.01 \leq \epsilon^2 < 0.06$ – Small; $0.06 \leq \epsilon^2 < 0.14$ – Medium, and $\epsilon^2 \geq 0.14$ – Large).

206

207 3. Results

208 3.1. Occurrence of trace chemical elements in clothing

209 The descriptive analysis of the concentrations of twenty trace elements in skin-
210 contact clothing samples ($n = 120$) commercialized in Spain is summarized in Table 2.
211 The individual values of the element levels in all samples are given in Table S3
212 (*Supplementary Information*). The levels of As, Bi, Cd, and Hg were below their
213 respective LODs in all the analyzed clothes. Some elements (i.e., Ag, B, Co, Mo, Sb, and
214 Sn) showed a detection rate lower than 50%, while Al and Ba were detected in all samples.
215 Aluminum, Zn, and Ti presented the highest median concentrations: 27.6, 5.55, and 4.17
216 mg/kg, respectively. The levels of Ag in the current samples were below the LOD (0.04
217 mg/kg), while Zn showed a median concentration of 5.55 mg/kg in all the clothes (Table
218 2). Considering the textile items individually, the highest level of Al was found in sample
219 #91 (toddlers dress, white and blue, 100% organic cotton). Zinc was detected at high
220 levels in sample #21 (pregnant women's bra, black, 100% recycled polyester), while Ti
221 was relatively abundant in sample #104 (toddlers t-shirt, blue and red, 100% organic
222 cotton) (Table 2).

223 In general terms, higher levels were observed in dark colors than in lighter ones
224 for all analyzed elements. Higher Cr levels were found in samples of dark clothing, with
225 a mean concentration of 141 mg/kg in black samples and 2.39 mg/kg in blue clothing. In
226 all cases, Co concentrations were below the LOD (<0.04 mg/kg), except for black clothes
227 (mean: 8.07 mg/kg). Pb showed low and similar values irrespective of the clothing color
228 (range: 0.09 to 0.19 mg/kg). Copper showed a median concentration of 1.16 mg/kg, with
229 a 94% of detection rate. Interestingly, Cu, used in dyes production, presented a greater
230 value (up to 15.7 mg/kg) in printing clothes (a mixture of colors). In turn, differences
231 were found when comparing dyeing and printing clothes. Aluminum, Cu, and Zn showed
232 high values in dyeing clothes (130 vs. 92.1 mg/kg $\epsilon^2 = 0.095$, 17.1 vs. 5.49 mg/kg $\epsilon^2 =$
233 0.161, and 52.0 vs. 10.2 mg/kg $\epsilon^2 = 0.076$, respectively). In contrast, Cr, Sb, and Ti
234 presented higher levels in printing clothes (17.4 mg/kg $\epsilon^2 = 0.075$, 8.39 mg/kg $\epsilon^2 = 0.083$,
235 and 112 mg/kg $\epsilon^2 = 0.079$).

236 The type of material could influence the type of trace elements found. In the
237 current survey, the median concentration of Sb in clothing was lower than its respective
238 LOD, with a mean value of 7.48 mg/kg and a 22% of detection rate. Clothes made of
239 polyester showed an Sb content of 49.4 mg/kg, a value higher than in other fibers (cotton,
240 elastane, and polyamide). The levels of Ti in synthetic fibers were higher than in natural
241 fibers, highlighting items made of polyester and polyamide (mean: 449 and 255 mg/kg,
242 respectively).

243 Barium and Mn showed higher eco-clothes levels than those made of conventional
244 cotton (6.15 vs. 1.22 mg/kg, $\epsilon^2 = 0.275$; and 1.99 vs. 1.23 mg/kg $\epsilon^2 = 0.327$, respectively).
245 Other chemical elements followed the same pattern (Figure 1). On the other hand, Sb was
246 the only element that showed a large difference (considering epsilon squared higher than
247 0.14) according to the place of manufacture, with higher concentrations in clothes made
248 outside the EU (3.03 vs. 8.98 mg/kg, $\epsilon^2 = 0.305$).

249 The levels of trace elements in clothes from pregnant women, newborns, and
250 toddlers are shown in Table 4. Generally, lower values were observed in newborn clothes
251 (12-36 months old). A difference was observed for Al when comparing the clothes of
252 newborns and pregnant women (15.5 vs. 164 mg/kg; $\epsilon^2 = 0.423$) and Cu between garments
253 for newborns and toddlers (3.68 vs. 16.2 mg/kg; $\epsilon^2 = 0.144$).

254
255

3.2. Health risk assessment

Data on human exposure to trace elements according to the three population subgroups and the type of cloth (i.e., t-shirts, jeans, panties, pyjamas, bodies, socks, underwear, and dresses) are shown in Table 5. In general terms, the greater the concentration of the trace element in clothing, the greater the risk of dermal absorption. Consequently, also a higher probability of adverse health effects occurrence.

Children's exposure to trace elements is generally greater than that of adults, considering their high exposure to chemicals and low body weight. A simulation of a real exposure situation was performed by considering the sum of all clothes used by each population group. Higher values of dermal exposure were observed for pregnant women and toddlers than newborns. However, no significant differences were detected between the groups. Regarding the individual target elements, the highest dermal exposure in pregnant women corresponded to Al and Ti ($1.28 \cdot 10^{-5}$ and $1.13 \cdot 10^{-5}$ mg/kg/day, respectively), while As and Hg contribution was notably lower ($6.94 \cdot 10^{-9}$ and $1.30 \cdot 10^{-8}$ mg/kg/day, respectively). Globally, values on dermal exposure were relatively low. However, it must be highlighted that the present study was focused only on dermal exposure through direct-contact clothing. Thus, the potential contribution of other exposure routes was neglected.

Data on dermal exposure were used to estimate the hazard quotient (non-carcinogenic risks) and cancer risk (CR). Chemicals are considered hazardous to human health when the HQ value exceeds 1. HQ values are summarized in Table 6. No data were provided for Bi because a dermal RfD is unavailable yet. In turn, health risks for Hg through clothing were not calculated because this element was not detected in any sample. Titanium showed HQ values higher than 1 for pregnant women (1.13) and newborns (1.22). In contrast, the HQ calculated for other elements was below the threshold.

Cancer risks were calculated for As, Cr, and Pb since they are the only elements for which an SFd value is available. The results of the carcinogenic risks associated with exposure to trace elements through clothing are depicted in Figure 2. In all cases, the risks were below the threshold value of 10^{-5} , which is considered acceptable according to international standards, acceptable below 10^{-6} (US. EPA, 2005).

4. Discussion

Trace elements may come from specific and different stages of industrial production. Some of them are added to textile manufacturing to improve the quality of

290 the material, while others are present in raw fiber or as dyes and pigments. Moreover,
291 using inorganic-based compounds as functional additives in textiles can lead to
292 heterogeneity in the element composition of clothing, being difficult to identify their
293 general usage and co-associations of elements (Rovira and Domingo, 2019; Fillela et al.,
294 2022). Taking this into account, data were also treated according to specific parameters,
295 such as the color of the fiber type.

296 Chromium, Pb, Cu, and Co are often used as metal complex dyes. Specifically,
297 Cr is employed as a metal complex dye (acid dye) in polyamide fibers, mainly for dark
298 clothes (Rovira et al., 2015, 2017a,b; Herrero et al., 2020; Chen et al., 2022). The results
299 of the current survey also support this evidence that dark clothing presented higher levels
300 of Cr, Co and Pb than light coloured clothing. Manganese compounds (such as MnO₂)
301 can effectively remove the color from certain dyes and pigments. Therefore, this element
302 is frequently used to bleach clothes and decolorize raw textile material by removing the
303 coloring components that are inherent or bonded to the fibers. In the present study, the
304 median concentration of Mn found in white clothes was 1.37 mg/kg. Although Mn is an
305 essential element, it also shows a toxicity profile (i.e., neuro- and immune toxicity) when
306 reaching high exposure rates (>3mg/day) (Batista et al., 2009). The essentiality *versus*
307 toxicity of chemical elements is an issue of public health concern, mainly in situations of
308 overexposure in early life. In this survey, the median value of Mn -determined in all
309 clothes- was 0.97 mg/kg.

310 Synthetic and natural fibers showed different elemental compositions due to the
311 difference in the sources of raw materials used (Sungur and Gulmez, 2015; Rovira et al.,
312 2017a; Herrero et al., 2019, 2020). Synthetic fibers are made from petroleum-derived
313 materials, being the elemental composition of these fibers is determined by their
314 production processes, such as polymerization. Antimony trioxide (Sb₂O₃) is a catalyst for
315 synthesizing polyester fibers, being used in the textile industry associated with
316 decabrodiphenyl ether for flame retardant properties. It is therefore expected that their
317 concentration is higher in fibers made from polyester than from other materials (cotton,
318 elastane, and polyamide), as shown by the results of this study. However, Sb is a toxic
319 element that can cause respiratory and skin irritation (Rovira et al., 2015, 2017a; Herrero
320 et al., 2019; Filella et al., 2022). Fortunately, the amount of antimony used in polyester
321 production is typically low, and government agencies, such as the US Environmental
322 Protection Agency, tightly regulate it.

323 Titanium has many applications in the textile industry. Titanium oxide (TiO₂) may
324 be used for antimicrobial activities in synthetic fibers, in clothes with UV filter properties,
325 and also as a pigment (Rovira et al., 2015, 2017a; Herrero et al., 2019; Filella et al., 2022).
326 The levels of Ti in synthetic fibers were higher than in natural fibers. These results
327 confirm the previous findings of Rovira et al. (2017a), who detected levels of 23.6 vs.
328 2.69 mg/kg in synthetic and natural fibers, respectively. Besides the TiO₂, nanoparticles
329 of Ag and the zinc oxide (ZnO) present antibacterial and antifungal activities and UV
330 protection for polyester fiber.

331 The chemical elements present in natural fibers depend on the exposure-grown
332 location and environmental conditions. Zinc and Cu are essential trace elements naturally
333 present in cotton fiber. They are found in the soil, absorbed by the plant, and incorporated
334 into the thread (Filella et al., 2022; Hasan et al., 2022). In this study, Zn and Cu showed
335 contents of 12.9 and 33.9 mg/kg, respectively, in clothes made of cotton. These values
336 were higher than those found in synthetic fibers. Cotton plants can also absorb toxic
337 metals, such as Cd and Pb. Natural fibers were classified into conventional and organic
338 cotton (eco-clothes). Differences in trace element contents -according to cotton
339 production- are depicted in Figure 1. Organic cotton is grown without synthetic pesticides
340 and fertilizers, thus reducing the risk of soil contamination with toxic elements, including
341 Cd, Pb, and As. Moreover, organic farming practices reduce the uptake of metals by
342 plants. It is important to note that metals in cotton do not necessarily mean human health
343 risks. Compared to synthetic fibers, the burdens of trace elements in these fibers are
344 generally low and typically well below the safety limits set by regulatory agencies (Rovira
345 et al., 2017a; Hasan et al., 2022; Goyal and Parashar, 2023).

346 For increased consumer safety and reliability, some textile products may own a
347 specific OEKO-TEX® Standard 100 certificate, indicating that the product has been tested
348 for harmful substances, including toxic elements (Pb, Cd, As, Hg, and Cr). These certified
349 products may be made with natural and synthetic fibers. The threshold of this certificate
350 is applied to the finished product, including all parts that are in direct contact with the
351 skin, besides buttons and zippers. Table 3 summarizes, for each trace element, the
352 threshold established by the Eco Guidelines (European Commission, 2014) and the
353 analyzed concentration on garments of the present study. As expected, the contents of
354 most trace elements were lower than their respective threshold in textiles. In addition, Cu
355 presented lower levels in certified clothes than in non-certified items ($\varepsilon^2 = 0.097$).
356 Similarly, the concentrations of Pb and Ni were below the established thresholds.

357 However, further analyses should be performed since both elements are toxic and showed
358 a high detection rate (71% and 95% for Pb and Ni, respectively). In contrast, the levels of
359 Cr were higher than the threshold for this metal. It must be stated that only Cr(VI) is
360 carcinogenic, and chemical speciation of Cr to differentiate the content of Cr(III) and
361 Cr(VI) was not performed in this study.

362 Another critical variable was the country of manufacture. In 2018, European
363 Union added a new restriction on hazardous substances (carcinogenic, mutagenic, or toxic
364 for reproduction - category 1A or 1B) in clothing, other textiles, and footwear in the
365 Regulation (EU) 2018/1513 (Annex XVII to REACH regulation) (EC, 2018). Hence,
366 some toxic elements, like As, Cd, Cr(VI), and Pb, are restricted in homogeneous textile
367 materials, with a maximum concentration limit by weight of 1 mg/kg. In this study, the
368 levels of As and Cd were below their respective LODs. Pb levels were 0.11 and 0.17
369 mg/kg in clothes manufactured in the EU and non-European countries, respectively.
370 Anyway, both values were lower than the limit established by the REACH regulation.
371 The levels of Cr, not differentiated between Cr(III) and Cr(VI), were higher than the
372 security threshold (6.28 and 35.2 mg/kg in textile items made inside and outside the EU,
373 respectively). It is important to note that the threshold established by OEKO-TEX
374 advocates that lower values of toxic elements should be found in baby clothes. Although
375 not all samples have this certificate, baby clothes contain a lower amount of trace
376 elements. Therefore, they are toxicologically safer (Chen et al., 2022).

377 The levels of trace elements in clothes previously reported by some authors are
378 similar to the results obtained here. Rovira et al. (2017a) analyzed the concentrations of
379 28 trace elements in home textiles. Similarly, the levels of As and Cd were below their
380 respective LODs, while high values of Al and Ti were detected in all the samples (10.9
381 and 1.58 mg/kg, respectively). In turn, Cu and Cr were more frequently detected in
382 colored clothes than in white textile samples. Herrero et al. (2020) also found high levels
383 of Cr and Cu in dark-colored garments. Rovira et al. (2015) also reported that Sb
384 concentrations were higher in clothes made of synthetic fibers than in cotton items (5.78
385 vs. 2.28 mg/kg), while Cd and Pb levels did not exceed the limit established by OEKO-
386 TEX[®]. In contrast, As and Ni levels found in samples from China (Chen et al., 2022) were
387 relatively high (0.07 mg/kg and 5.83 mg/kg, respectively) when compared to our data (<
388 LOD and 0.5 mg/kg, respectively). On the other hand, Filella et al. (2022) also found high
389 levels of Ti in cleaning cloths, with concentrations up to 3.73 mg/kg. Due to its important

390 properties applied in synthetic fibers, Ti is an abundant and ubiquitous element in
391 clothing.

392 Exposure assessment during early life is essential, considering that toxic effects
393 in critical periods of development, such as during fetal development and early childhood,
394 can have significant and long-lasting impacts on health. The levels of inorganic elements
395 determined in clothes were used to estimate the dermal exposure for different population
396 subgroups and to evaluate the associated health risks. In general, a higher dermal
397 absorption occurs for those chemical elements deposited on the surface of the fibers, such
398 as Ti and silver (Ag), compared to those linked to the fibers (Herrero et al., 2019).
399 Consequently, the exposure duration of wearing direct-contact clothes is a fundamental
400 issue.

401 The non-carcinogenic risk was evaluated by calculating the HQ. In the current
402 survey only Ti showed HQ higher than the limit value, than 1, for pregnant women and
403 newborns. It is important to note that the toxic effects of Ti are largely dependent on the
404 form and amount of exposure (Rovira et al., 2017a; Filella et al. 2022). Additionally, there
405 have been concerns about the potential carcinogenic of titanium dioxide (Filella et al.
406 2022). Therefore, since this element is ubiquitous in textile products, Ti levels in clothes
407 should be frequently monitored. Cancer risks were calculated for As, Cr, and Pb and
408 although they were within the acceptable values ($< 10^{-6}$) considering that the exposure
409 occurs during early life, no safety values should be applied to carcinogenic elements.

410 When considering an early life exposure scenario, health risks for pregnant
411 women must be particularly evaluated, as most toxic elements cross the placenta.
412 Exposure during critical time windows leads to adverse health effects throughout
413 childhood and even later in life. In addition, it is fundamental to identify sensitive
414 windows to develop toxic responses in these periods. At last, some factors that may
415 influence chemical elements' exposure effects, including age, sex, nutritional status,
416 combined chemical exposures (including both organic and inorganic chemicals), and
417 aggregated exposure sources (i.e., dermal absorption, soil ingestion, and dietary intake),
418 should be taken into account in health risk assessment (Souza et al., 2020; Souza et al.,
419 2023; Zheng et al., 2023).

420

421 **5. Conclusions**

422 Some trace elements may be both essential and/or toxic to human health,
423 depending on the internal dose, or exposure level. Therefore, it is basic to consider all
424 exposure pathways in risk assessment. However, dermal exposure has still been very little
425 studied, mainly for toxic compounds. Clothing is an important source of exposure to trace
426 elements since its chemical composition is extensive, including toxic elements. A number
427 of factors may influence the elements levels in textile products and their dermal
428 absorption. In the present study, high levels of Ti were found in clothing, being the HQ
429 values of this element, calculated for pregnant women and newborns, above the
430 international thresholds. Current legislation only limits the levels of 4 metals, when
431 several studies show that more metals are present in clothing and could have an adverse
432 effect on health. It would be necessary to review the legislation and non-mandatory
433 certifications in order to include a wider range of substances. Finally, assessing early-life
434 exposure to toxic elements can help to identify potential sources and to prevent or reduce
435 human exposure. In addition, it can also help to identify populations that may be
436 particularly vulnerable and inform public health policies and interventions to protect
437 human health. Unfortunately, clothing does not only contain a toxic substance, therefore,
438 it is necessary that risk assessments are performed under a multi-exposure and multi-
439 chemical scenario.

440

441 **6. Declarations**

442 *Credit author statement*

443 Marta Herrero: Conceptualization; Formal analysis; Investigation; Methodology;
444 Resources; Writing – original draft.

445 Joaquim Rovira: Conceptualization; Writing – review & editing.

446 Neus González: Conceptualization; Writing – review & editing.

447 Montse Marquès: Conceptualization; Writing – review & editing.

448 Fernando Barbosa: Writing – review & editing.

449 Jordi Sierra: Conceptualization; Formal analysis; Writing – review & editing.

450 José L. Domingo: Writing – review & editing.

451 Martí Nadal: Conceptualization; Funding acquisition; Project administration; Writing –
452 review & editing.

453 Marília Cristina Oliveira Souza: Conceptualization; Writing – original draft.

454

455 *Conflict and/or competing interest*

456 The authors declare that they do not have known competing for financial interests
457 or personal relationships that could have appeared to influence the study reported here.

458

459 *Acknowledgments*

460 This study is part of the EarlyCLOTHES study, which has been financially
461 supported by the Spanish Ministry of Science and Innovation through grant PID 2019-
462 104443 GB-I00.

463

464 *Ethics approval and consent to participate*

465 Not applicable

466

467 *Consent for publication*

468 Not applicable.

469

470

471

472 **References**

473

- 474 Avadí, A.; Marcin, M.; Biard, Y.; Renou, A.; Gourlot, J.P.; Basset-Mens, C. (2020). Life cycle
475 assessment of organic and conventional non-Bt cotton products from Mali. *International*
476 *Journal of Life Cycle Assessment*, v.25, p.678–697. DOI:10.1007/s11367-020-01731-x
- 477 Batista, B. L.; Rodrigues, J. L.; Tormen, L.; Curtius, A., L.; Barbosa, F. (2009). Reference
478 Concentrations for Trace Elements in Urine for the Brazilian Population based on q-ICP-MS
479 with a Simple Dilute-and-Shoot Procedure. *Journal of the Brazilian Chemical Society*, v.20,
480 p.1406-1413.
- 481 Biver, M.; Turner, A.; Filella, M. (2021). Antimony release from polyester textiles by artificial
482 sweat solutions: A call for a standardized procedure. *Regulatory Toxicology and*
483 *Pharmacology*, v.119, p.104824. DOI: 10.1016/j.yrtph.2020.104824.
- 484 Bocca, B., Ruggieri, F., Pino, A., Rovira, J., Calamandrei, G., Martínez, M.Á., Domingo, J.L.,
485 Alimonti, A., Schuhmacher, M. (2019) Human biomonitoring to evaluate exposure to toxic
486 and essential trace elements during pregnancy. Part A. concentrations in maternal blood,
487 urine and cord blood. *Environmental Research*, 177, art. no. 108599. DOI:
488 10.1016/j.envres.2019.108599
- 489 Castro, I., Arroyo, R., Aparicio, M., Martínez, M.Á., Rovira, J., Ares, S., Cunha, S.C., Casal, S.,
490 Fernandes, J.O., Schuhmacher, M., Nadal, M., Rodríguez, J.M., Fernández, L. (2021)
491 Dietary habits and relationship with the presence of main and trace elements, bisphenol A,
492 tetrabromobisphenol A, and the lipid, microbiological and immunological profiles of breast
493 milk. *Nutrients*, 13 (12), 4346.
- 494 Chen, H.; Chai, M.; Cheng, J.; Wang, Y.; Tang, Z. (2022). Occurrence and health implications of
495 heavy metals in preschool children's clothing manufactured in four Asian regions.
496 *Ecotoxicology and Environmental Safety*, v.245, p.114121. DOI:
497 10.1016/j.ecoenv.2022.114121.
- 498 EC, European Commission. Commission decision of 9 July 2009 establishing the ecological
499 criteria for the award of the Community Ecolabel for textile products. 2009/567/EC. Off. J.
500 Eur. Union. L197, pp. 70–86.
- 501 EC, European Commission. Commission Decision of 5 June 2014 establishing the ecological
502 criteria for the award of the EU Ecolabel for textile products (notified under document C
503 (2014) 3677) Text with EEA relevance. Off. J. Eur. Union L174, pp.1-83.
- 504 EC, European Commission. Commission Regulation (EU) 2018/1513 of 10 October 2018
505 amending Annex XVII to Regulation (EC) No 1907/2006 of the European Parliament and of
506 the Council concerning the Registration, Evaluation, Authorization and Restriction of
507 Chemicals (REACH) as regard, 2018/1513. Off. J. Eur. Union. L256, pp. 24 – 28.
- 508 ECHA, European Chemicals Agency. Guidance on Information Requirements and Chemical
509 Safety Assessment. Chapter R.15: Consumer Exposure Assessment. Version 3.0; European
510 Chemicals Agency: Helsinki, Finland, 2016; ISBN 978-92-9495-079-6.
- 511 Filella, M.; Brazard, J.; Adachi, T.B.M.; Turner, A. (2022). Hazardous chemical elements in
512 cleaning cloths, a potential source of microfibres. *Science of the Total Environmental*, v.846,
513 p.157419. DOI: 10.1016/j.scitotenv.2022.157419.
- 514 Goyal, A.; Parashar, M.(2023). Organic cotton and BCI-certified cotton fibers. *Sustainable Fibers*
515 *for Fashion and Textile Manufacturing*, book chapter 3, p.51-74. DOI: 10.1016/B978-0-12-
516 824052-6.00011-1.
- 517 Gupta, N., Yadav, K.K., Kumar, V., Kumar, S., Chadd, R.P., Kumar, A., 2019. Trace elements in
518 soil-vegetables interface: Translocation, bioaccumulation, toxicity and amelioration - A
519 review. *Science of The Total Environment*. 651, 2927-2942.
- 520 Hasan, M.M.; Cai, L.; Ji, X.; Ocran, F.M. (2022). Eco-friendly clothing market: A study of
521 willingness to purchase organic cotton clothing in Bangladesh. *Sustainability*, v.14, p.4827.
522 DOI:10.3390/su14084827
- 523 Herrero, M., Rovira, J., Nadal, M., Domingo, J.L. Risk assessment due to dermal exposure of
524 trace elements and indigo dye in jeans: Migration to artificial sweat (2019) *Environmental*
525 *Research*, 172, pp. 310-318.

526 Herrero, M., Rovira, J., Esplugas, R., Nadal, M., Domingo, J.L. (2020) Human exposure to trace
527 elements, aromatic amines and formaldehyde in swimsuits: Assessment of the health risks.
528 Environmental Research, 181, 108951.

529 Herrero, M.; Gonzalez, N.; Rovira, J.; Marquès, M.; Domingo, J.L.; Nadal, M. (2022a). Health
530 risk assessment of polychlorinated biphenyls (PCBs) in baby clothes. A preliminary study.
531 Environmental Pollution, v.307, p.119506. DOI: 10.1016/j.envpol.2022.119506.

532 Herrero, M., González, N., Rovira, J., Marquès, M., Domingo, J.L., Nadal, M. (2022b) Early-life
533 exposure to formaldehyde through clothing. Toxics, v.10, p. 361. DOI:
534 10.3390/toxics10070361.

535 Jaishankar, M., Tseten, T., Anbalagan, N., Mathew, B.B., Beeregowda, K.N., 2014. Toxicity,
536 mechanism, and health effects of some heavy metals. Interdisciplinary toxicology. 7, 60-72.

537 Kim, H. Harrison, F.E.; Aschner, M.; Bowman, A.B. (2022). Exposing the role of metals in
538 neurological disorders: a focus on manganese. Trends in Molecular Medicine, v.28, p.555-
539 568. DOI: 10.1016/j.molmed.2022.04.011.

540 Letinic, J.G.; Saric, M.M.; Piasek, M.; Jurasovic, J.; Varnai, V.M.; Grgec, A.S.; Orct, T. (2016).
541 Use of human milk in the assessment of toxic metal exposure and essential element status in
542 breastfeeding women and their infants in coastal Croatia. Journal of Trace Elements in
543 Medicine and Biology. 38, 117–125.

544 Mandy, M.; Nyirenda, M. (2018). Developmental Origins of Health and Disease: the relevance
545 to developing nations. International Health, v.10, p.66-70. DOI: 10.1093/inthealth/ihy006.

546 OEKO-TEX. OEKO-TEX Standard 100. Limit Values and Fastness. Access in January 2023.
547 Available at: [https://www.oeko-](https://www.oeko-tex.com/en/manufacturers/test_criteria/limit_values/limit_values.html)
548 [tex.com/en/manufacturers/test_criteria/limit_values/limit_values.html](https://www.oeko-tex.com/en/manufacturers/test_criteria/limit_values/limit_values.html)

549 Oliveira, M.M., Trevilato, T.M.B., Segura-Muñoz, S.I., Aragon, D.C., Alves, L.G., Nadal, M.,
550 Marquès, M., Domingo, J.L., Sierra, J., Camelo, J.S., Jr. (2020) Essential and toxic elements
551 in human milk concentrate with human milk lyophilizate: A preclinical study. Environmental
552 Research, 188, 109733.

553 PE International (2014). The life cycle assessment of organic cotton fiber was commissioned by
554 Textile Exchange. – A global average. Accessed in January 2023. Available in:
555 [https://www.ajsosteniblebcn.cat/the-life-cycle-assessment-of-organic-cotton-](https://www.ajsosteniblebcn.cat/the-life-cycle-assessment-of-organic-cotton-fiber_38172.pdf)
556 [fiber_38172.pdf](https://www.ajsosteniblebcn.cat/the-life-cycle-assessment-of-organic-cotton-fiber_38172.pdf)

557 Rana, M.B.; Hasan, M.Z.; Islam, M.A.; Rokonuzzaman, M.; Khandaker, S.; Basha, M.M. (2022).
558 Printing Effects of Different Natural Colourants on Cotton Fabric. Textile & Leather Review,
559 v.5, p.280-295. DOI:10.31881/TLR.2022.30.

560 Rovira, J., Nadal, M., Schuhmacher, M., Domingo, J.L. Human exposure to trace elements
561 through the skin by direct contact with clothing: Risk assessment (2015) Environmental
562 Research, 140, pp. 308-316.

563 Rovira, J., Nadal, M., Schuhmacher, M., Domingo, J.L. (2017a). Home textile as a potential
564 pathway for dermal exposure to trace elements: assessment of health risks Journal of the
565 Textile Institute, 108 (11), pp. 1966-1974.

566 Rovira, J., Nadal, M., Schuhmacher, M., Domingo, J.L. (2017b). Trace elements in skin-contact
567 clothes and migration to artificial sweat: Risk assessment of human dermal exposure. Textile
568 Research Journal, v.87, p.726-738.

569 Rovira, J.; Domingo, J.L. (2019). Human health risks due to exposure to inorganic and organic
570 chemicals from textiles: A review. Environmental Research, v.168, p.62-69. DOI:
571 10.1016/j.envres.2018.09.027.

572 Rovira, J., Martínez, M.Á., Mari, M., Cunha, S.C., Fernandes, J.O., Marmelo, I., Marques, A.,
573 Haug, L.S., Thomsen, C., Nadal, M., Domingo, J.L., Schuhmacher, M. (2022) Mixture of
574 environmental pollutants in breast milk from a Spanish cohort of nursing mothers.
575 Environment International, 166, 107375.

576 Rujido-Santos, I.; Herbelo-Hermelo, P.; Barciela-Alonso, M.C.; Bermejo-Barrera, P.; Moreda-
577 Piñeiro, A. (2022). Metal Content in Textile and (Nano)Textile Products. International
578 Journal of Environmental Research and Public Health, v.19, p.944. DOI:
579 10.3390/ijerph19020944.

580 Sanders, A. P.; Saland, J. M.; Wright, R. O.; Satlin, L. (2018). Perinatal and childhood exposure
581 to environmental chemicals and blood pressure in children: a review of literature 2007-2017.
582 *Pediatric research*, v.84, p.165–180. DOI: 10.1038/s41390-018-0055-3.

583 Sarayu, K.; Sandhya, S. (2012). Current Technologies for Biological Treatment of Textile
584 Wastewater—A Review. *Applied Biochemistry and Biotechnology*, v.167, p.645–661. DOI
585 10.1007/s12010-012-9716-6.

586 Sima, M.F. (2022). Determination of some heavy metals and their health risk in T-shirts printed
587 for a special program. *PLoS ONE*, v.17, p.e0274952. DOI: 10.1371/journal.pone.0274952.

588 Souza, M.C.O.; Saraiva, M.C.P.; Honda, M.; Barbieri, M.A.; Bettiol, H.; Barbosa, F., Kannan, K.
589 (2020). Exposure to per- and polyfluorinated alkyl substances in pregnant Brazilian women
590 and its association with fetal growth. *Environmental Research*, v.187, p.109585. DOI: 10.
591 1016/j.envres.2020.109585.

592 Souza, M.C.O.; Devóz, P.P.; Ximenez, J.P.B.; Bocato, M.Z.; Rocha, B.A.; Barbosa, F. (2022).
593 Potential Health Risk to Brazilian Infants by Polybrominated Diphenyl Ethers Exposure via
594 Breast Milk Intake. *International Journal of Environmental Research and Public Health*, v.19,
595 p.11138. DOI: 10.3390/ijerph191711138.

596 Souza, M.C.O.; Gonzalez, N.; Herrero, M.; Marquès, M.; Rovira, J.; Nadal, M., Barbosa, F.;
597 Domingo, J.L. (2023a). Screening of regulated aromatic amines in clothing marketed in
598 Brazil and Spain: Assessment of human health risks. *Environmental Research*, p.115264.
599 DOI: 10.1016/j.envres.2023.115264.

600 Souza, M. C. O., González, N., Herrero, M., Marquès, M., Rovira, J., Domingo, J. L., Barbosa,
601 F., Jr, Nadal, M. (2023b). Non-regulated aromatic amines in clothing purchased in Spain and
602 Brazil: Screening-level exposure and health impact assessment. *Journal of Environmental*
603 *Management*, 339, 117905.

604 Sungur, Ş.; Gülmez, F. (2015). Determination of metal contents of various fibers used in textile
605 industry by MP-AES. *Journal of Spectroscopy*, v.2015, p.1-5. DOI: 10.1155/2015/640271.

606 United States Environmental Protection Agency (US. EPA, 2005). Guidelines for Carcinogen
607 Risk Assessment. 51(September), 33992–34003. EPA/630/P-03/001F

608 United States Environmental Protection Agency (US. EPA, 2011). Exposure Factors Handbook
609 2011 Edition (Final Report). Washington. DC. EPA/600/R-09/052F.

610 United States Environmental Protection Agency. (US.EPA, 2021). Regional Screening Levels
611 (RSLs)—Generic Tables. Accessed on 15 September 2022. Available online at
612 <https://www.epa.gov/risk/regional-screening-levels-rsls-generic-tables>.

613 Urbano, T.; Zagnoli, F.; Malavolti, M.; Halldorsson, T.I.; Vinceti, M.; Filippini, T. (2022).
614 Dietary intake of potentially toxic elements and childrens chemical exposure. *Current*
615 *Opinion in Environmental Science and Health*, v.30, p.100393. DOI:
616 10.1016/j.coesh.2022.100393.

617 Velusamy, S.; Roy, A.; Sundaram, S.; Mallick, T. K. (2021). A Review on Heavy Metal Ions and
618 Containing Dyes Removal Through Graphene Oxide-Based Adsorption Strategies for
619 Textile Wastewater Treatment. *The Chemical Record*, v.21, p.1570–1610.
620 DOI:10.1002/tcr.202000153

621 Zheng, K.; Zeng, Z.; Tian, Q.; Huang, J.; Zhong, Q.; Huo, X. (2023). Epidemiological evidence
622 for the effect of environmental heavy metal exposure on the immune system in children.
623 *Science of the Total Environment*, v.868, p. 161691. DOI: 10.1016/j.scitotenv.2023.161691.

Table 1. Characteristics of clothing of pregnant women, newborns and toddlers.

| | | Pregnant women (%) | Newborns (%) (<12 months old) | Toddlers (%) (12-36 months old) |
|------------------------|-------------------------------|-------------------------------|---|--|
| | N | 44 | 30 | 50 |
| Coloring type | Dyed | 93 | 67 | 50 |
| | Printed | 7 | 33 | 50 |
| Fiber material | 100% Natural | 2 | 67 | 84 |
| | 100% Synthetic | 14 | 0 | 2 |
| | Mixed - Natural and synthetic | 84 | 33 | 14 |
| Natural fiber | Organic cotton | 52 | 50 | 50 |
| | Regular cotton | 48 | 50 | 50 |
| Synthetic fiber | Elastane | 82 | 7 | 10 |
| | Polyester | 14 | 3 | 6 |
| | Polyamide | 2 | 23 | 0 |
| Colors | White | 11 | 13 | 20 |
| | Black | 20 | 0 | 2 |
| | Grey | 30 | 10 | 2 |
| | Blue | 20 | 3 | 10 |
| | Pink | 5 | 7 | 8 |
| | Others | 9 | 17 | 18 |
| | Mix | 5 | 50 | 40 |
| Official certification | OEKO-TEX® Standard 100 label | 9 | 23 | 2 |
| | OEKO-TEX® Made in Green | 5 | 0 | 0 |
| | GOTS | 0 | 13 | 44 |
| | Without certification | 86 | 63 | 54 |
| Store | Chain Store | 39 | 77 | 58 |
| | Supermarket | 9 | 10 | 10 |
| | Shop | 0 | 13 | 14 |
| | Online | 52 | 0 | 18 |
| Manufactured | European Union | 11 | 27 | 24 |
| | Outside the EU | 89 | 73 | 76 |

Table 2. Descriptive analysis of the trace element concentrations (mg/kg) in skin contact clothing samples (n=120).

| Trace elements | DR ¹ (%) | Mean | SD ² | Min ³ | Median | 95th percentile | Max ⁴ | LOD ⁵ |
|----------------|---------------------|------|-----------------|------------------|--------|-----------------|------------------|------------------|
| Ag | 10 | <LOD | 0.02 | <LOD | <LOD | 0.05 | 0.19 | 0.04 |
| Al | 100 | 104 | 244 | 1.95 | 27.6 | 261 | 1856 | 1.67 |
| As | 0 | <LOD | - | <LOD | <LOD | <LOD | <LOD | 0.17 |
| B | 35 | 0.82 | 0.89 | <LOD | 0.42 | 2.34 | 8.16 | 0.83 |
| Ba | 100 | 3.76 | 17.3 | 0.42 | 0.85 | 8.70 | 185 | 0.08 |
| Bi | 0 | <LOD | - | <LOD | <LOD | <LOD | <LOD | 0.08 |
| Cd | 0 | <LOD | - | <LOD | <LOD | <LOD | <LOD | 0.08 |
| Co | 6 | 0.69 | 7.05 | <LOD | <LOD | 0.20 | 78.5 | 0.08 |
| Cr | 57 | 12.2 | 88.8 | <LOD | 0.21 | 8.74 | 828 | 0.17 |
| Cu | 94 | 9.20 | 21.8 | <LOD | 1.16 | 45.7 | 160 | 0.17 |
| Hg | 0 | <LOD | - | <LOD | <LOD | <LOD | <LOD | 0.17 |
| Mn | 54 | 1.62 | 2.09 | <LOD | 0.97 | 4.95 | 12.8 | 0.83 |
| Mo | 14 | 0.08 | 0.18 | <LOD | <LOD | 0.27 | 1.28 | 0.08 |
| Ni | 95 | 0.81 | 1.22 | <LOD | 0.50 | 2.18 | 7.62 | 0.17 |
| Pb | 71 | 0.15 | 0.13 | <LOD | 0.12 | 0.42 | 0.72 | 0.08 |
| Sb | 22 | 7.48 | 28.1 | <LOD | <LOD | 49.5 | 192 | 0.83 |
| Sn | 15 | 0.05 | 0.03 | <LOD | <LOD | 0.13 | 0.18 | 0.08 |
| Sr | 94 | 2.78 | 3.22 | <LOD | 1.56 | 11.5 | 14.5 | 0.17 |
| Ti | 77 | 80.9 | 225 | <LOD | 4.17 | 634 | 1244 | 0.42 |
| Zn | 76 | 22.8 | 105 | <LOD | 5.55 | 31.6 | 1077 | 1.83 |

¹DR: Detection rate; ²SD: standard deviation; ³Min: minimum; ⁴Max: maximum; ⁵LOD: limit of detection.

Table 3. Threshold (mg/kg) for trace elements established by the Eco Guidelines - OEKO-TEX® Standard 100 certificate and the corresponding level determined on clothes in this study.

| Trace elements | Threshold by OEKO-TEX® ¹ | OEKO-TEX® Standard 100 certificate | Levels (mg/kg) |
|------------------------|-------------------------------------|------------------------------------|----------------|
| As | 0.2 | Yes | < LOD |
| | 1.0 | No | < LOD |
| Ba | 1000 | Yes | 1.95 |
| | 1000 | No | 4.01 |
| Cd | 0.1 | Yes | < LOD |
| | 0.1 | No | < LOD |
| Co | 1.0 | Yes | 0.16 |
| | 4.0 | No | 0.79 |
| Cr ² | 1.0 | Yes | 2.76 |
| | 2.0 | No | 13.40 |
| Cu | 25 | Yes | 1.41 |
| | 50 | No | 10.3* |
| Hg | 0.02 | Yes | < LOD |
| | 0.02 | No | < LOD |
| Ni | 1.0 | Yes | 0.59 |
| | 4.0 | No | 0.84 |
| Pb | 0.2 | Yes | 0.17 |
| | 1.0 | No | 0.15 |
| Sb | 30 | Yes | 0.46 |
| | 30 | No | 8.79 |

¹Threshold by OEKO-TEX®: first value: clothes for babies (< 36 months old); second value: textile products of the direct skin contact (mg/kg) – this explains the two values per chemical element.

² Considering only Cr(VI): 0.5 mg/kg

* $p < 0.05$

Table 4. Mean levels of trace elements (mg/kg) determined in the clothes of pregnant women, newborns, and toddlers.

| | Pregnant women | | | | | Newborn (<12 months old) | | | | Toddlers (12–36 months old) | | | | | |
|------------|----------------|----------------|-------|---------|----------------|--------------------------|----------|-------|----------------|-----------------------------|-----------|-------|---------|----------------|----------------|
| | T-shirt | Leggings/jeans | Bra | Panties | Σ ^a | Pajama | Bodysuit | Socks | Σ ^a | Pajama | Underwear | Dress | T-shirt | Leggings/jeans | Σ ^a |
| Ag | < LOD | < LOD | < LOD | 0.04 | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | 0.03 | 0.03 | 0.03 | 0.03 | < LOD |
| Al* | 117 | 121 | 173 | 260 | 164 | 15.5 | 9.81 | 21.0 | 15.4 | 36.3 | 14.6 | 210.9 | 191.2 | 67.6 | 104 |
| As | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD |
| B | 0.71 | 0.98 | 0.76 | 0.83 | 0.83 | 1.87 | 0.60 | 0.69 | 1.05 | 1.01 | 0.48 | 0.77 | 0.42 | 0.68 | 0.67 |
| Ba | 1.60 | 4.56 | 8.20 | 0.70 | 3.84 | 1.03 | 1.44 | 1.85 | 1.44 | 0.97 | 1.44 | 19.9 | 1.54 | 1.63 | 5.09 |
| Bi | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD |
| Cd | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD |
| Co | < LOD | 0.08 | 8.12 | 0.22 | 1.93 | < LOD | < LOD | 0.04 | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD |
| Cr | 0.33 | 2.96 | 55.6 | 86.9 | 33.4 | < LOD | 0.18 | 1.89 | 0.71 | 0.16 | 0.22 | 0.26 | 0.17 | 0.31 | 0.22 |
| Cu* | 2.72 | 8.67 | 5.40 | 2.02 | 5.06 | 3.01 | 4.22 | 3.83 | 3.68 | 23.5 | 2.92 | 35.6 | 8.15 | 10.6 | 16.2 |
| Hg | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD | < LOD |
| Mn | 2.45 | 2.06 | 2.39 | 1.32 | 2.05 | 0.98 | 1.27 | 2.43 | 1.56 | 0.93 | 0.84 | 1.02 | 1.30 | 2.28 | 1.27 |
| Mo | 0.05 | 0.32 | 0.05 | 0.05 | 0.13 | < LOD | < LOD | 0.06 | < LOD | 0.05 | 0.07 | 0.08 | 0.08 | < LOD | < LOD |
| Ni | 1.09 | 2.12 | 0.40 | 0.24 | 1.07 | 0.62 | 0.54 | 0.95 | 0.70 | 0.52 | 0.44 | 0.58 | 1.37 | 0.31 | 0.64 |
| Pb | 0.18 | 0.18 | 0.12 | 0.06 | 0.14 | 0.20 | 0.11 | 0.11 | 0.14 | 0.29 | 0.10 | 0.22 | 0.11 | 0.15 | 0.17 |
| Sb | < LOD | 23.1 | 3.70 | 0.66 | 8.43 | < LOD | < LOD | 7.67 | 2.84 | < LOD | 1.25 | 39.3 | < LOD | 9.49 | 10.2 |
| Sn | 0.05 | < LOD | < LOD | 0.05 | < LOD | 0.06 | < LOD | 0.05 | < LOD | 0.07 | 0.05 | 0.07 | 0.05 | 0.05 | < LOD |
| Sr | 3.35 | 2.80 | 3.04 | 3.15 | 3.06 | 4.40 | 1.63 | 2.88 | 2.97 | 2.82 | 2.51 | 2.98 | 1.71 | 2.14 | 2.43 |
| Ti | 10.2 | 143 | 176 | 241 | 143 | 0.29 | 2.32 | 260 | 87.5 | 6.29 | 1.02 | 90.5 | 13.3 | 1.71 | 22.6 |
| Zn | 11.2 | 7.05 | 4.86 | 5.01 | 7.03 | 19.9 | 9.56 | 33.18 | 20.9 | 59.4 | 8.34 | 3.92 | 111 | 5.60 | 37.6 |

* $p < 0.05$

^a: Sum of all the clothes used by the group.

Table 5. Human dermal exposure (mg/kg/day) to trace elements through clothing represented by the sum of all of them and the percentage contribution of each cloth.

| | Pregnant women | | | | | Newborns (<12 months old) | | | | Toddlers (12–36 months old) | | | | | |
|-----------|----------------|--------------------|-----|---------|----------|---------------------------|----------|-------|----------|-----------------------------|-----------|-------|---------|--------------------|----------|
| | T-shirt | Leggings/ jeans | Bra | Panties | Σ* | Pajama | Bodysuit | Socks | Σ* | Pajama | Underwear | Dress | T-shirt | Leggings/ jeans | Σ* |
| Ag | 32 | 36 | 10 | 22 | 1.98E-09 | 45 | 49 | 6 | 1.01E-09 | 36 | 4 | 15 | 11 | 35 | 2.38E-09 |
| Al | 28 | 34 | 12 | 26 | 1.28E-05 | 56 | 34 | 10 | 6.37E-07 | 17 | 1 | 34 | 24 | 25 | 7.49E-06 |
| As | 36 | 42 | 11 | 11 | 6.94E-09 | 48 | 46 | 6 | 3.80E-09 | 39 | 4 | 13 | 12 | 32 | 7.02E-09 |
| B | 31 | 50 | 11 | 8 | 7.52E-08 | 74 | 23 | 3 | 5.74E-08 | 51 | 3 | 11 | 6 | 28 | 7.03E-08 |
| Ba | 16 | 51 | 31 | 2 | 3.22E-07 | 38 | 52 | 9 | 6.12E-08 | 10 | 1 | 69 | 5 | 15 | 3.47E-07 |
| Bi | 36 | 43 | 11 | 10 | 2.45E-09 | 46 | 46 | 7 | 1.37E-09 | 39 | 4 | 13 | 12 | 32 | 2.44E-09 |
| Cd | 36 | 42 | 11 | 11 | 3.47E-09 | 48 | 46 | 6 | 1.90E-09 | 39 | 4 | 13 | 12 | 32 | 3.51E-09 |
| Co | 1 | 3 | 92 | 4 | 1.20E-07 | 48 | 46 | 6 | 1.90E-09 | 39 | 4 | 13 | 12 | 32 | 3.51E-09 |
| Cr | 0 | 7 | 34 | 59 | 2.24E-06 | 16 | 34 | 51 | 1.15E-08 | 26 | 4 | 14 | 8 | 48 | 2.05E-08 |
| Cu | 28 | 52 | 16 | 4 | 3.32E-07 | 40 | 54 | 7 | 1.73E-07 | 46 | 1 | 29 | 5 | 19 | 1.60E-06 |
| Hg | 36 | 42 | 11 | 11 | 6.94E-09 | 48 | 46 | 6 | 3.80E-09 | 39 | 4 | 13 | 12 | 32 | 7.02E-09 |
| Mn | 33 | 48 | 13 | 6 | 2.32E-07 | 38 | 48 | 13 | 5.72E-08 | 24 | 2 | 7 | 10 | 57 | 1.37E-07 |
| Mo | 9 | 87 | 2 | 2 | 1.89E-08 | 47 | 44 | 9 | 1.96E-09 | 35 | 5 | 19 | 17 | 24 | 4.64E-09 |
| Ni | 24 | 72 | 3 | 1 | 1.45E-07 | 49 | 41 | 10 | 2.88E-08 | 35 | 4 | 14 | 29 | 19 | 4.64E-08 |
| Pb | 44 | 42 | 9 | 5 | 1.30E-08 | 62 | 33 | 4 | 7.31E-09 | 56 | 2 | 14 | 6 | 22 | 1.79E-08 |
| Sb | 2 | 91 | 6 | 1 | 8.73E-07 | 23 | 22 | 55 | 4.19E-08 | 2 | 1 | 52 | 1 | 44 | 6.85E-07 |
| Sn | 38 | 42 | 10 | 10 | 3.96E-09 | 59 | 35 | 6 | 2.47E-09 | 44 | 4 | 13 | 10 | 29 | 4.86E-09 |
| Sr | 36 | 46 | 10 | 8 | 2.92E-07 | 69 | 25 | 6 | 1.43E-07 | 46 | 4 | 12 | 8 | 30 | 2.23E-07 |
| Ti | 3 | 44 | 21 | 33 | 1.13E-05 | 1 | 6 | 93 | 8.76E-07 | 21 | 0 | 62 | 11 | 5 | 1.08E-06 |
| Zn | 53 | 34 | 7 | 5 | 6.54E-07 | 60 | 28 | 11 | 7.54E-07 | 62 | 1 | 1 | 31 | 5 | 3.34E-06 |

*Σ: Sum of all clothes used by the group.

Table 6. Non-cancer risk (Hazardous quotient – HQ) calculated for pregnant women, newborns, and toddlers.

| Trace Elements | Pregnant women | Newborns (<12 months old) | Toddlers (12-36 months old) |
|-----------------------|-----------------------|---|--|
| Ag | 1.05E-05 | 1.55E-05 | 1.19E-05 |
| Al | 1.28E-05 | 1.34E-05 | 7.49E-06 |
| As | 2.46E-05 | 3.73E-05 | 2.34E-05 |
| B | 4.13E-07 | 7.01E-07 | 3.51E-07 |
| Ba | 2.33E-05 | 2.77E-05 | 2.48E-05 |
| Cd | 3.69E-02 | 5.59E-02 | 3.51E-02 |
| Co | 4.00E-04 | 4.00E-04 | 1.17E-05 |
| Cr | 2.99E-02 | 3.00E-02 | 3.00E-04 |
| Cu | 8.53E-06 | 1.29E-05 | 4.00E-05 |
| Mn | 1.66E-06 | 2.07E-06 | 9.76E-07 |
| Mo | 3.83E-06 | 4.22E-06 | 9.29E-07 |
| Ni | 3.00E-04 | 4.00E-04 | 1.00E-04 |
| Pb | 1.98E-06 | 3.02E-06 | 2.56E-06 |
| Sb | 1.46E-02 | 1.53E-02 | 1.14E-02 |
| Sn | 7.17E-09 | 1.13E-08 | 8.11E-09 |
| Sr | 4.89E-07 | 7.27E-07 | 3.72E-07 |
| Ti | 1.13* | 1.22* | 1.08E-01 |
| Zn | 2.56E-06 | 5.07E-06 | 1.11E-05 |

*Values higher than the threshold.

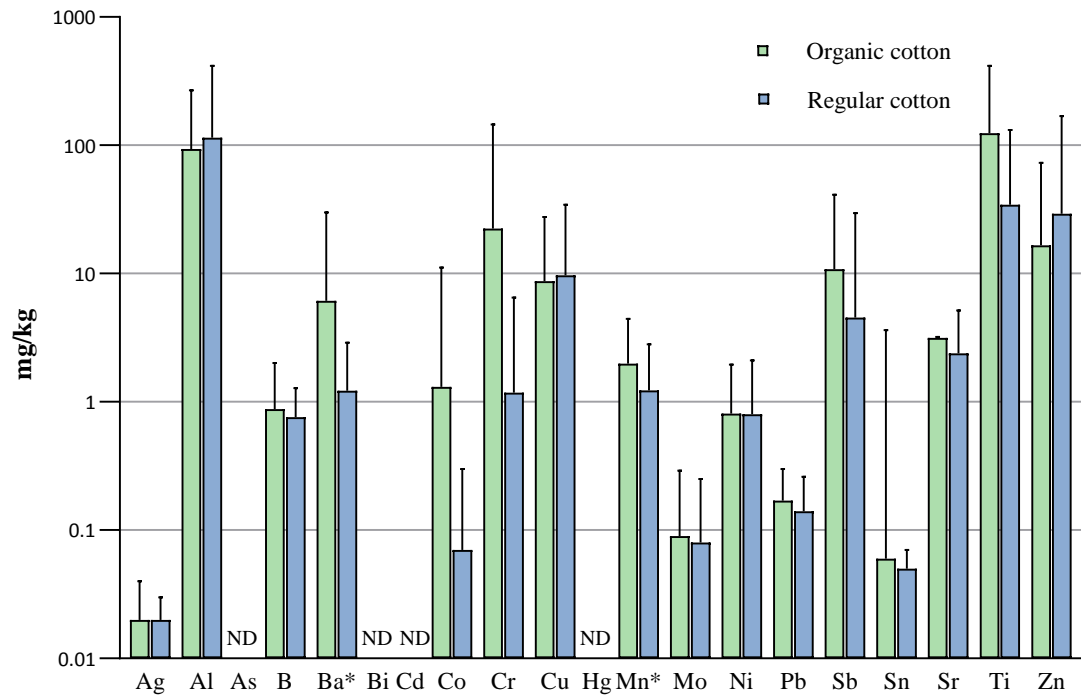


Figure 1. Levels of trace elements in clothes made with organic and conventional cotton. ND: not detected. *: $p < 0.05$

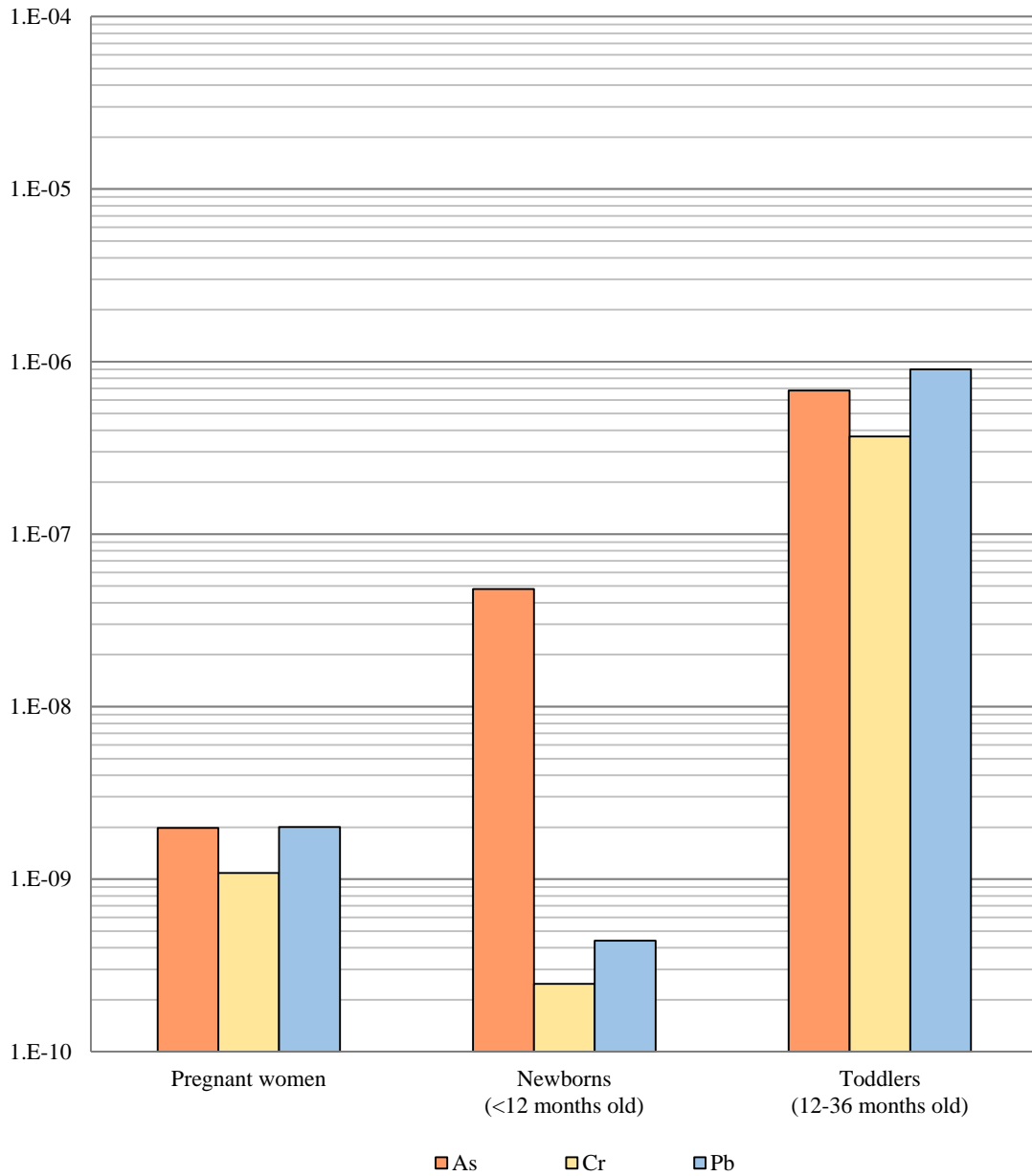


Figure 2. Cancer risk associated to exposure to As, Cr and Pb for pregnant women, newborns and toddlers.



Click here to access/download
Supplementary Material
Supplementary Information.docx



Credit author statement

Marta Herrero: Conceptualization; Formal analysis; Investigation; Methodology; Resources; Writing – original draft.

Joaquim Rovira: Conceptualization; Writing – review & editing.

Neus González: Conceptualization; Writing – review & editing.

Montse Marquès: Conceptualization; Writing – review & editing.

Fernando Barbosa: Writing – review & editing.

Jordi Sierra: Conceptualization; Formal analysis; Writing – review & editing.

José L. Domingo: Writing – review & editing.

Martí Nadal: Conceptualization; Funding acquisition; Project administration; Writing – review & editing.

Marília Cristina Oliveira Souza: Conceptualization; Writing – original draft.

Declaration of interests

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: