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## **Environmental determinants of different Blood Lead Levels in children: a quantile analysis from a nationwide survey.**

Anne Etchevers, Alain Le Tertre, Jean-Paul Lucas, Philippe Bretin, Youssef Oulhote, Barbara Le Bot, Philippe Glorennec

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1 **Environmental determinants of different Blood Lead Levels in children: a**  
2 **quantile analysis from a nationwide survey.**

3

4 **Authors and affiliations:** Etchevers Anne<sup>a</sup>, Le Tertre Alain<sup>b</sup>, Lucas Jean-Paul<sup>c,d</sup>, Bretin  
5 Philippe<sup>e</sup>, Oulhote Youssef<sup>f</sup>, Le Bot Barbara<sup>g,a</sup>, Glorennec Philippe<sup>g,a</sup>.

6 <sup>a</sup> INSERM U1085, IRSET- Environmental and Occupational Health Research Institute, 2  
7 avenue du Professeur Léon Bernard, 35043 RENNES Cedex, France

8 <sup>b</sup> InVS - French Institute for Public Health Surveillance, 12 rue du Val d'Osne, 94415 Saint-  
9 Maurice Cedex, France. E-mail address: a.letertre@invs.sante.fr

10 <sup>c</sup> CSTB - Centre Scientifique et Technique du Bâtiment, 84 avenue Jean Jaurès, 77447 Marne-  
11 la-Vallée Cedex 2, France. E-mail address: [JeanPaul.LUCAS@cstb.fr](mailto:JeanPaul.LUCAS@cstb.fr)

12 <sup>d</sup> University of South Brittany, UMR 6205, LMBA, F-56000 Vannes, France

13 <sup>e</sup> Ministry of Health, 14 avenue Duquesne, 75350 Paris, France. Email address:  
14 philippe.bretin@sante.gouv.fr

15 <sup>f</sup> Montreal University, Department of Environmental and Occupational Health, CHU Sainte-  
16 Justine, Chemin de la Côte Sainte-Catherine, Montreal, H3T1A8 Canada. Email address:  
17 youssef.oulhote@umontreal.ca

18 <sup>g</sup> EHESP, Rennes, Sorbonne Paris Cité, Avenue du Professeur Léon-Bernard, 35043 Rennes  
19 Cedex, France. Email adress : [Philippe.Glorennec@ehesp.fr](mailto:Philippe.Glorennec@ehesp.fr); Barbara.LeBot@ehesp.fr

20

21 **Corresponding author:** Anne Etchevers, anne.etchevers@inserm.fr

22 INSERM U1085, IRSET- Environmental and Occupational Health Research Institute

23 2 avenue du Professeur Léon Bernard, 35043 RENNES cedex, France

24 Tel: +33 (0)2 23 23 69 11 / Fax: +33 (0)2 23 23 50 55

25 **Abstract**

26 **Background:** Blood Lead Levels (BLLs) have substantially decreased in recent decades in  
27 children in France. However, further reducing exposure is a public health goal because there  
28 is no clear toxicological threshold. The identification of the environmental determinants of  
29 BLLs as well as risk factors associated with high BLLs is important to update prevention  
30 strategies. We aimed to estimate the contribution of environmental sources of lead to different  
31 BLLs in children in France.

32 **Methods:** We enrolled 484 children aged from 6 months to 6 years, in a nationwide cross-  
33 sectional survey in 2008-2009. We measured lead concentrations in blood and environmental  
34 samples (water, soils, household settled dusts, paints, cosmetics and traditional cookware).  
35 We performed two models: a multivariate generalized additive model on the geometric mean  
36 (GM), and a quantile regression model on the 10<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup> and 90<sup>th</sup> quantile of BLLs.

37 **Results:** The GM of BLLs was 13.8 µg/L (=1.38 µg/dL) (95% Confidence Intervals (CI):  
38 12.7-14.9) and the 90<sup>th</sup> quantile was 25.7 µg/L (CI: 24.2-29.5). Household and common area  
39 dust, tap water, interior paint, ceramic cookware, traditional cosmetics, playground soil and  
40 dust, and environmental tobacco smoke were associated with the GM of BLLs. Household  
41 dust and tap water made the largest contributions to both the GM and the 90<sup>th</sup> quantile of  
42 BLLs. The concentration of lead in dust was positively correlated with all quantiles of BLLs  
43 even at low concentrations. Lead concentrations in tap water above 5 µg/L were also  
44 positively correlated with the GM, 75<sup>th</sup> and 90<sup>th</sup> quantiles of BLLs in children drinking tap  
45 water.

46 **Conclusions:** Preventative actions must target household settled dust and tap water to reduce  
47 the BLLs of children in France. The use of traditional cosmetics should be avoided whereas  
48 ceramic cookware should be limited to decorative purposes.

49 **Keywords:** blood lead, lead exposure, dust, water, soil.

## 1. Introduction

Blood lead levels (BLLs) in young children have considerably declined in developed countries over the past 15 years. The geometric mean of BLLs in children decreased from 36 to 15  $\mu\text{g/L}$  between 1996 and 2009 in France (Etchevers et al. 2013). Similar BLLs have been reported in Germany (Becker et al. 2013), the USA (CDC 2013), Croatia, the Czech Republic, Poland, Slovakia, Slovenia (Hruba et al. 2012) and Sweden (Stromberg et al. 2008) in recent years.

Many scientific publications have shown adverse health effects associated with BLLs below 50  $\mu\text{g/L}$  ( $=5 \mu\text{g/dL}$ ) (National Toxicology Program 2012) and there is currently no defined toxicity threshold (Canfield et al. 2003;Jusko et al. 2008;Lanphear et al. 2005). As a consequence, the German Federal Environmental Agency and the Centers for Disease Control and Prevention (CDC) have recently revised the blood lead ‘levels of concern’ of 100  $\mu\text{g/L}$ . It was lowered from 100  $\mu\text{g/L}$  to 35  $\mu\text{g/L}$  (Wilhelm et al. 2010) in Germany and from 100  $\mu\text{g/L}$  to 50  $\mu\text{g/L}$  (CDC 2012) in the USA. Similarly, in France, a reduction of the current level of 100  $\mu\text{g/L}$  to an as-yet undetermined threshold is under revision.

Since the removal of lead from gasoline, residential sources have become the biggest sources of lead exposure for children in developed countries (Lanphear et al. 2003). Levallois *et al.* (Levallois et al. 2013) and Oulhote *et al.* (Oulhote et al. 2011;Oulhote et al. 2013) recently demonstrated that exposure to several sources of lead including tap water, home and exterior dust and soil is associated with BLLs in children.

It is essential to evaluate the contribution of each individual source to BLLs in children both to design prevention strategies and to limit environmental exposure. Such prevention strategies must target both children with low BLLs (the most frequent) and children with

74 elevated BLLs. The identification of environmental factors that contribute to elevated BLLs  
75 will facilitate both the design of effective screening programs and strategies to remove these  
76 sources. Identification of the contributors to low BLLs is important because they contribute to  
77 the main burden of IQ loss (Lanphear et al. 2005) and have the largest economic impact  
78 (Pichery et al. 2011).

79 The first objective of the study was to identify the contribution of lead sources to BLLs in  
80 young children living in France 1) for the whole population (corresponding to the geometric  
81 mean) and 2) for the most exposed children (corresponding to the 90<sup>th</sup> quantile of BLLs). The  
82 second objective was to compare the contribution of dust and tap water lead levels at different  
83 points of the BLL distribution (geometric mean, 10<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup> and 90<sup>th</sup> quantiles of  
84 BLLs).

85

## 86 **2. Methods**

87

### 88 **2.1. Population and sampling**

89 Between 2008 and 2009, the French Institute for Public Health Surveillance (InVS)  
90 implemented a nationwide (n=3831) cross-sectional survey to estimate BLLs in children (6  
91 months to 6 years of age) in France (Etchevers et al. 2013). Between November 2008 and  
92 August 2009, the Scientific and Technical Building Institute (CSTB) carried out a nested  
93 environmental survey in homes of a random subsample of 484 children in mainland France  
94 (Etchevers et al. 2013;Lucas et al. 2012). Hereafter, only the main features will be recalled.  
95 We used a two-stage probability sample design: in the first stage, the primary sampling units  
96 were hospitals and in the second stage, we included hospitalized children. The hospitals were  
97 stratified by administrative regions and the risk of lead poisoning, the extent of old and poor  
98 housing in the catchment area, and industrial activity related to lead. Hospitals in high risk

99 areas were intentionally oversampled. The sampling weights were then adjusted by post-  
100 stratification based on auxiliary variables (region, sex, age and eligibility for complementary  
101 free health insurance (CMUc)) to increase the precision of estimates and to make the sample  
102 more representative of the population (Lumley T 2010). The participation was 83% for  
103 hospitals. The participation for parents was 97% at hospitals and 62% at home.

104

105

## **2.2 Data and sample collection**

106 Children gave venous blood samples (1.5 mL) during the hospital stay. At each child's home,  
107 we interviewed one adult who was living with the child, about demographic, housing and  
108 behavioral characteristics and we sampled residential sources of lead. In each dwelling, we  
109 collected wipe samples of floor dust from a 0.1 m<sup>2</sup> surface area of the floor where the child  
110 was reported to play, in up to five rooms (U.S.HUD 2002). In addition, we collected one or  
111 two dust samples in the entrance hall and in the landing for apartments. If the child was  
112 reported to play outside, in a garden or playground in close vicinity to the home, the ground  
113 was sampled by coring (2 cm deep) for soil surfaces or by dust wiping (0.1 m<sup>2</sup>) for hard  
114 surfaces. We collected a 2 L tap water sample after a 30 min stagnation time of water in the  
115 pipework. The water samples were homogenized and were poured into a 0.25 L flask  
116 acidified with 1% of HNO<sub>3</sub> to ensure a pH < 2. We performed paint measurements with  
117 portable X-ray fluorescence (XRF) lead-based paint analyzers (Niton) on each part of the  
118 room (wall, door, and window) accessible to the child, except on new parts of the room.  
119 Finally, if the family agreed, we also collected traditional cosmetics (kohl) or dishes known to  
120 be potential sources of lead.

121

## **2.3 Chemical analyses**

122

### ***2.3.1 Blood lead levels***

123 Blood lead levels were analyzed by inductively coupled plasma mass spectrometry (ICP-MS).

124 The blood samples were diluted (1:10) with an aqueous matrix modifier solution (0.2%

125 butanol, 0.1% Triton and 1% nitric acid). The limit of quantification (LOQ) was 0.037 µg/L.  
126 In all cases, BLLs were above the LOQ. All blood samples with a BLL greater than 80 µg/L  
127 were analyzed a second time to confirm the result. Quality control procedures were  
128 performed: blanks and internal quality controls from reference materials (Utak blood samples  
129 of 27.91 µg/L and 394.92 µg/L) were analyzed for every 10 samples. External quality control  
130 procedures included participation in the AFSSAPS (French Agency for medical care safety)  
131 interlaboratory control (2007 and 2009) and the use of external samples from the INSPQ  
132 (National Institute of Public Health of Quebec). The external control test was considered  
133 successful if there was less than 10% difference between the expected and observed values.

### 134 *2.3.2 Lead measurements in environmental samples, kohl and traditional* 135 *cookware*

136 We analyzed environmental samples for lead content using an ICP-MS (Agilent Technology)  
137 7500ce equipped with a quadrupole mass filter and an octopole reaction cell. We analyzed all  
138 environmental samples (except for water) for leachable (regulatory method in France) and  
139 total lead content (Le Bot et al. 2010;Le Bot et al. 2011). The LOQ were 1 µg/L for water, 1  
140 µg/m<sup>2</sup> (0.09 mg/ft<sup>2</sup>) for leachable lead in dust and 2 µg/m<sup>2</sup> (0.19 mg/ft<sup>2</sup>) for total lead in dust.  
141 For soil, the LOQ were 0.5 µg/g and 1.3 µg/g, for leachable and total lead respectively. For  
142 traditional cosmetic (kohl), we used the same leachable digestion method as for soil. The  
143 LOQ was 1.3 µg/g. For traditional cookware, leachable lead was measured by contact with  
144 acetic acid (4%) for 24 hours at room temperature (ISO 7086-1 2000). The LOQ was 1 µg/L.  
145 Quality control was performed with analytical blanks and standard reference materials SRM  
146 2583 and SRM 2584 for dust, certified reference material CRM 013-050 for paint, CRM SS2  
147 for soil and kohl, and the National Institute of Standards and Technology NIST 1643 for  
148 water and traditional cookware. Control samples were included in all digestion series or  
149 analyses series (for water) to determine lead concentration in a manner identical to that of the

150 real samples. The lab has French accreditation (Comité Français d'accréditation (COFRAC))  
151 for the analysis of lead in water and dust. The intra-laboratory relative standard deviation for  
152 lead in all types of sample was lower than 10%.

153

## 154 **2.4 Statistical analyses**

155 We used two different modeling approaches: 1) a generalized additive model (GAM) of  
156 expected geometric mean of BLLs to quantify the risk factors for the whole population and 2)  
157 quantile regressions for expected 10<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup> and 90<sup>th</sup> quantiles of BLLs to study risk  
158 factors of specific areas of the BLLs distribution. In the GAM model, we included the  
159 following variables: lead levels in interior dust, dust from common areas of the building, tap  
160 water, soil, playground dust, paints, cookware ceramics, traditional cosmetics (kohl, surma,  
161 tiro), along with children's sex, children's age, environmental tobacco smoke (ETS) exposure,  
162 tap water consumption and parents' occupational exposure to lead. In the quantile regression  
163 models, we removed some covariates (i.e. lead in paints, ceramics, traditional cosmetics,  
164 ETS) that were collinear with other risk factors at the 90<sup>th</sup> quantile of BLLs due to the absence  
165 or quasi-absence of these risks factors in children.

166 The construction of the variables is presented in Appendix A.

167 BLLs were natural log-transformed to ensure proper distribution of the residuals. We  
168 transformed environmental lead concentrations to their cubic root to address the high degree  
169 of skewness. In the GAM model, the variables age, tap water, interior dust, dust from indoor  
170 common areas concentrations were included as penalized smoothing splines to capture  
171 possible non-linearity with BLLs; the other variables were introduced as linear terms. In the  
172 quantile regression models, the variables age, interior dust and tap water concentrations were  
173 introduced as natural spline functions with three degrees of freedom to potentially adjust for



174 non-linearity, whereas the other variables were introduced linearly. We performed analysis on  
175 the complete data set (434 children), with no missing data for the included variables.

176 For the sake of simplicity, we presented the increase in BLLs associated with a change of the  
177 environmental lead concentration from its 25<sup>th</sup> or 50<sup>th</sup> percentile to its 95<sup>th</sup> percentile and from  
178 its 25<sup>th</sup> or 50<sup>th</sup> percentile to its 99<sup>th</sup> percentile to assess the contribution of each environmental  
179 component to BLLs. The estimated increases are presented for the geometric mean (GM) for  
180 GAM analyses and for the 90<sup>th</sup> quantile of BLLs for quantile regressions. A variable was  
181 considered as a risk factor if its estimated increase was positive. Evidence of the association  
182 was assessed both in terms of 95% confidence interval and consistency with published results.  
183 Using the GAM and quantile regression models, we predicted the GM, 10<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup>  
184 and 90<sup>th</sup> quantiles of BLLs for lead concentrations in dust and tap water to construct dose-  
185 response relationship curves (Figure 1).

186 We used the survey package (Lumley T 2011) in the R software (R : a Language and  
187 Environment for Statistical Computing 2012) to account for the complex sampling design and  
188 the sampling weight of each participant. We carried out the quantile regressions with the  
189 quantreg package (Koenker R 2012).

190 The results in this manuscript are for total lead. The results for leachable lead are available in  
191 Appendix B.

192

193

### 194 **3. Results**

#### 195 **3.1 Characteristics of the study population and environmental factors**

196 The estimated blood and environmental lead concentrations are described in Table 1. They  
197 were determined by Lucas *et al.* (Lucas 2012) for paints, dust and soil. The population-  
198 weighted geometric mean BLL was 13.8 µg/l and the 95<sup>th</sup> percentile was 32.8 µg/l.

199 **Insert Table 1**

200 About 21% of children were exposed to indoor passive smoking (Table 2), and 10% were  
201 exposed for more than 2 hours/day. Thirteen percent of parents used traditional cookware and  
202 1.5% used ceramics releasing lead. Approximately 3% of families declared the use of  
203 traditional cosmetics from their native countries; 0.8% used cosmetics containing lead.  
204 Twenty one percent of parents were potentially exposed to lead during their work or hobbies.  
205 The distribution of children's age, sex and passive smoking in this sub-sample was similar to  
206 those estimated in the original sample selected at hospital (Etchevers et al. 2014).

207 **Insert Table 2**

208 **3.2 Associations between environmental exposure and BLLs**

209 ***3.2.1 Associations between environmental sources and geometric mean of BLLs***

210 The geometric mean of BLLs was associated with all potential sources included in the model.  
211 However, the precision of the estimate was low (the confidence interval contained 0) for dust  
212 from indoor common areas, exterior playground soil, ETS and the use of cosmetics releasing  
213 lead. An association between BLLs and dust from common areas and paints in good condition  
214 was only detected for a change from their 25<sup>th</sup> percentile to their 99<sup>th</sup> percentile, although the  
215 precision of this estimate was low. The geometric mean of BLLs was associated with  
216 traditional ceramic cookware (+56% in GM), traditional cosmetics releasing lead (+43% in  
217 GM), floor dust from inside homes (+33% in GM with a range of 3 [0.3 µg/ft<sup>2</sup>] to 62 µg/m<sup>2</sup> [6  
218 µg/ft<sup>2</sup>]), tap water (+44% in GM with a range of 1 to 14 µg/L), and outdoor playground dust  
219 (+33% in GM with a range of 0 to 187.5 µg/m<sup>2</sup> [17 µg/ft<sup>2</sup>]).

220 **Insert Table 3**

221

222 **3.2.2 Associations between environmental sources and the quantile 90<sup>th</sup> of BLLs**

223 The risk factors of the 90<sup>th</sup> quantile (25.7 µg/L) of BLLs are displayed in Table 4. BLLs ≥ the  
224 90<sup>th</sup> quantile were associated with dust from the household interior and indoor common areas  
225 and with tap water, for both children drinking tap or bottled water. Precision was lower for tap  
226 water and dust from indoor common areas than for household dust. High BLLs were  
227 estimated for high levels of exposure to these factors (99<sup>th</sup> quantile).

228 **Insert Table 4**

229

230 **3.2.3 Predicted BLLs according to exposure to household dust and tap water**

231 For the two main risk factors (household dust and tap water) we plotted separately the  
232 concentration-response relationship for the geometric mean and the 10<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup>, 90<sup>th</sup>  
233 quantiles of BLLs (Figure 1). For each prediction, the contribution of other factors was set  
234 arbitrarily to zero. . The zone shaded in grey shows the 95% confidence interval of the  
235 estimated geometric mean.

236 **Insert Figure 1**

237 Lead loadings in interior floor dust were positively correlated with all quantiles of BLLs. The  
238 higher the BLLs were, the higher the contribution of lead loadings from interior floor dust  
239 was. Low and median BLLs (10<sup>th</sup> to 50<sup>th</sup> quantile) increased with increasing dust lead  
240 loadings between 1 and 60 µg/m<sup>2</sup> (5.6 µg/ft<sup>2</sup>). The 75<sup>th</sup> and 90<sup>th</sup> quantiles increased until 200  
241 µg/m<sup>2</sup> (18.6 µg/ft<sup>2</sup>) and tended to reach a plateau or to decrease above 200 µg/m<sup>2</sup>. This  
242 decrease of BLLs may be explained by the high uncertainty for the high dust lead loadings.  
243 The geometric mean did not follow this pattern and BLLs were still positively correlated with  
244 dust lead loadings above 200 µg/m<sup>2</sup>. The highest BLL predicted by exclusively interior floor  
245 dust did not exceed 21 µg/L.

246 The presence of lead in tap water was positively correlated with all quantiles of BLLs in  
247 children drinking tap water. Tap water lead concentrations higher than 5  $\mu\text{g/L}$  affected the  
248 geometric mean and elevated BLLs (75<sup>th</sup> and 90<sup>th</sup> quantile) and concentrations higher than 10  
249  $\mu\text{g/L}$  affected the lower quantiles (10<sup>th</sup> and 25<sup>th</sup>). For children drinking tap water with a lead  
250 concentration of 10  $\mu\text{g/L}$ , the predicted GM BLL was 15  $\mu\text{g/L}$  and the predicted 90th quantile  
251 of BLLs was 19  $\mu\text{g/L}$ . The highest levels of water lead exposure (between 30 and 50  $\mu\text{g/L}$ )  
252 contributed to predict BLLs between 36 to 55  $\mu\text{g/L}$ , if we consider no contribution from other  
253 sources. Additionally, BLLs of the 90<sup>th</sup> quantile were positively correlated with tap water lead  
254 concentrations in children drinking bottled water, most probably due to the use of leaded tap  
255 water for food preparation and cooking (Triantafyllidou and Edwards M. 2011). BLLs of the  
256 geometric mean and the 75<sup>th</sup> quantile tended to increase for water lead concentrations higher  
257 than 20  $\mu\text{g/L}$  in children drinking bottled water. A water lead concentration of 10  $\mu\text{g/L}$  was  
258 associated with a GM BLL of 11  $\mu\text{g/L}$  and a BLL of 17  $\mu\text{g/L}$  for the more exposed (90th  
259 quantile of BLLs).

260

#### 261 **4. Discussion**

262

263 The aim of this study was to estimate the contribution of environmental sources of lead on the  
264 geometric mean and 90th percentile of BLLs and to compare, for tap water and dust, the  
265 relationship between its concentration and BLLs at different points of the BLLs distribution.  
266 Household dust and tap water were the main predictors of both the geometric mean and 90th  
267 quantile of BLLs. Playground soil and dust affected the geometric mean of BLLs but not the  
268 90th quantile of BLLs. We also found a strong association between ceramic cookware,  
269 traditional cosmetics and the geometric mean of BLLs in children.

## 270 **4.1 Environmental determinants of BLLs**

### 271 *4.1.1 Interior floor dust*

272 The concentration of lead in household dust was positively correlated with all quantiles of  
273 BLLs and this was the main risk factor for all quantiles of BLLs studied in our study. We  
274 demonstrated that lead loadings much lower than the current American standard of 40  $\mu\text{g}/\text{ft}^2$   
275 ( $430 \mu\text{g}/\text{m}^2$ ) were positively correlated with BLLs: for example they increased the 90th  
276 quantile of BLLs by 75% for a change from 4  $\mu\text{g}/\text{m}^2$  to 62  $\mu\text{g}/\text{m}^2$  (6  $\mu\text{g}/\text{ft}^2$ ) and by 96% for a  
277 change from 4  $\mu\text{g}/\text{m}^2$  to 172  $\mu\text{g}/\text{m}^2$  (16  $\mu\text{g}/\text{ft}^2$ ). Approximately 5% of children were exposed  
278 to lead loadings in dust above 62  $\mu\text{g}/\text{m}^2$  (6  $\mu\text{g}/\text{ft}^2$ ) and 1% to levels above 172  $\mu\text{g}/\text{m}^2$  (16  
279  $\mu\text{g}/\text{ft}^2$ ). The geometric mean of BLLs estimated in the American NHANES study (1999-2004)  
280 was higher (20.3  $\mu\text{g}/\text{L}$ ) than our estimate here (Dixon et al. 2009); nonetheless, we found that  
281 interior dust lead loading had a similar contribution on BLLs at dust levels below 34.4  $\mu\text{g}/\text{m}^2$   
282 (3.2  $\mu\text{g}/\text{ft}^2$ ). Above 34.4  $\mu\text{g}/\text{m}^2$ , the contribution of dust was lower in our study than in that of  
283 the NHANES. A more recent study in Montreal (Canada) also reported a weak association  
284 between BLLs >18  $\mu\text{g}/\text{L}$  (75th quantile in their study) and floor dust >13  $\mu\text{g}/\text{m}^2$  (Levallois et  
285 al. 2013).

### 286 *4.1.2 Dust from indoor common areas*

287 Although lead concentrations in dust from indoor common areas were nearly four times  
288 higher (GM = 32.2  $\mu\text{g}/\text{m}^2$ ) than in household dust (GM=8.7  $\mu\text{g}/\text{m}^2$ ), its contribution was only  
289 observable for the 1% most exposed children in the geometric mean (Table 3), the 75<sup>th</sup>  
290 quantile (data not shown) and the 90th quantile (Table 4). The estimates for the extreme  
291 quantiles of BLLs had a high uncertainty due to the small number of children in our sample  
292 (114). As few as 8 out of 114 children declared spending time in common areas; dust from the  
293 common area may be an indirect source of exposure. Indeed, dust from landings has been  
294 demonstrated to be the major contributor to lead in interior floor dust (Lucas et al. 2014).

295

### 4.1.3 Tap water

296 Lead in drinking water was an important exposure pathway in our study with a contribution of  
297 water lead concentrations above 5  $\mu\text{g/L}$  on the geometric mean, 75<sup>th</sup> and 90<sup>th</sup> quantiles of  
298 BLLs for consumers and on the 90<sup>th</sup> quantile for non-consumers. All BLLs were also  
299 positively correlated with concentrations of lead in tap water exceeding the current 2013  
300 European standard of 10  $\mu\text{g/L}$  (Council of the European Union 1998); it concerned three  
301 percent of French children in 2009. Moreover we reported that high lead water exposure alone  
302 can be responsible for elevated BLLs: the 90<sup>th</sup> predicted quantile of BLLs reached 50  $\mu\text{g/L}$   
303 with a water lead concentration of 42  $\mu\text{g/L}$  and 35  $\mu\text{g/L}$  with a water lead concentration of 29  
304  $\mu\text{g/L}$ . However, the correlation between high levels of lead in water and BLLs has a high  
305 uncertainty (based on few children). Concerning the tap water values below LOQ (58%)  
306 included as raw data, we performed a sensitivity analysis in a non-weighted GAM using a  
307 multiple imputation on values of tap water <LOQ to assess a potential bias on the estimates.  
308 The estimated risk associated to tap water exposure with multiple imputation was similar to  
309 the one estimated with missing data or with replacement of values by raw data (data not  
310 shown). If, as observed by Triantafyllidou *et al* (Triantafyllidou *et al.* 2013) at higher lead  
311 water concentrations, water lead measurements were underestimated in our study due to  
312 samples transfer, the strength of the association between lead in water and blood would be  
313 overestimated. The effect of tap water has been studied previously (Brown and Margolis  
314 2012; Lanphear *et al.* 1998a; Levallois *et al.* 2013; Miranda *et al.* 2007; Renner 2010). These  
315 results are consistent with those of Levallois *et al.* who reported that lead concentrations >  
316 3.27  $\mu\text{g/L}$  influenced the 75<sup>th</sup> quantile of BLLs (17.8  $\mu\text{g/L}$ ) (Levallois 2013). We noticed also  
317 that elevated BLLs were influenced by indirect consumption (through food preparation and  
318 cooking) but with a lower impact than for children drinking tap water. It demonstrated that  
319 water lead exposure can be reduced by consumption of exclusively bottled water. Fertmann *et*

320 *al* have shown that consuming bottled water for drinking and cooking during 11 weeks  
321 reduced maternal median blood lead exposure from 32 to 20  $\mu\text{g/L}$  (Fertmann et al. 2004). We  
322 found a similar decline on the 90<sup>th</sup> quantile of BLLs between children drinking tap water  
323 versus bottled water.

#### 324 ***4.1.4 Exterior soil and dust***

325 The geometric mean of BLLs was 18% higher at exterior dust lead concentrations of 32  $\mu\text{g/m}^2$   
326 than at 0  $\mu\text{g/m}^2$  and was 33% higher at 187  $\mu\text{g/m}^2$ . Playground dust also affected the 10<sup>th</sup>, 25<sup>th</sup>  
327 and 75<sup>th</sup> quantile of BLLs (data not shown). However we did not have enough statistical  
328 power with only 53 dust samples to model the relationship between the quantiles of BLLs and  
329 exterior dust with high precision. Nevertheless, we can assume that the contribution of  
330 exterior dust to the GM was underestimated, given that dust in indoor common areas must  
331 have captured a part of the contribution of the exterior dust (Lucas et al. 2014).

332 Lead in soil only influenced the geometric mean and the 50th quantile of BLLs. Lead content  
333 in soil increased the GM of 13% for a change from 0 to 251 mg/kg and 16% for a change  
334 from 0 to 407 mg/kg; however, there was no statistical evidence. Even above the current U.S.  
335 federal hazard standard of 400 mg/kg (1.4% of exterior play areas are above this threshold in  
336 France), the contribution is weak, probably because the effect of soil may have also been  
337 captured by household dust.

338 Studies have demonstrated an association between soil (and sometimes exterior dust from  
339 play areas) and BLLs for high lead concentrations in soils (Lanphear et al. 1998a; Lanphear et  
340 al. 1998b; Lanphear et al. 2002). However, few studies assessed the contribution of soil at  
341 similar lead concentrations.

#### 342 ***4.1.5 Interior lead-based paints***

343 Leaded paint in good condition was associated with a 13% increase in geometric mean BLL  
344 for a variation between the 25th and the 99th quantile. Paints may be either a direct source of  
345 exposure when children make contact with the walls or an indirect source of exposure through  
346 dust. Studies have demonstrated an association between high BLLs and leaded paints (Gulson  
347 et al. 2013; Lanphear et al. 1998b; Schwartz and Levin 1991) whereas a more recent study that  
348 did not find this relationship when an adjustment was made for dust and tap water (Levallois  
349 2013).

#### 350 *4.1.6 Cosmetics and ceramics*

351 Ceramics and cosmetics were associated with an increase in the GM of BLLs (56% for  
352 ceramics and 43% for traditional cosmetics releasing lead). The association was detectable for  
353 lead levels above the LOQ, which was 1 µg/L for ceramics and 0.13 mg/g for traditional  
354 cosmetics. Therefore, we demonstrate an association for a lead content far below the 4 mg/L  
355 current European legislation for food container materials (JORF 2006). The range of lead  
356 concentrations leached from ceramics was 7 to 2 380 000 µg/L (only two dishes exceeded 4  
357 mg/L) and five out of 14 sampled ceramics were tagines. We observed a similar association  
358 (27% increase in GM) for families who declared using traditional ceramic cookware, which  
359 was not tested for lead release (13% of families overall). Lead was detectable in nine out of  
360 16 kohl samples, mainly originating from Maghreb and Egypt, even though lead is forbidden  
361 in cosmetics in France. This exposure involved 3% of families (only mothers). Previous cases  
362 of lead exposure involving lead-glazed ceramics and kohl were identified among the 690  
363 children screened in France (Inserm and InVS 2008) and in neonates in three maternity wards  
364 in the surroundings of Paris in 2004 (Yazbeck et al. 2007). Traditional cosmetics, such as  
365 surma and kohl, are also associated with high levels of lead exposure in children in the  
366 countries where they are frequently used (Al-Saleh et al. 1999; Goswami 2013; Nuwayhid et  
367 al. 2003; Rahbar et al. 2002) and in sporadic cases in the US (CDC 2004; CDC 2013b).



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#### ***4.1.7 Environmental tobacco smoke***

370 Although our results seem to confirm the influence of ETS on BLLs as already demonstrated  
371 by Mannino *et al.* (Mannino et al. 2003), Dixon *et al.* (Dixon et al. 2009) and Apostolou *et al.*  
372 (Apostolou et al. 2012), the relationship we observed was not statistically significant. We  
373 also found no trend between the duration of exposure to smoking indoors and BLLs. In  
374 addition this relationship did not remain when we added free complementary health insurance  
375 (CMUc) to the model as a proxy of poverty (data not shown).

376

#### ***4.1.8 Parent's occupational risk***

377 We did not find a relationship between parents' occupation and BLLs. However the list of  
378 occupational activities was very general and 21% of parents declared activities related to lead  
379 exposure.

380

381

## **4.2 Limitations**

382 First, a potential selection bias due to hospital recruitment could persist despite the  
383 consideration of the sampling weights and the post-stratification through the use of auxiliary  
384 information (available in the census). A residual bias may persist if some characteristics of the  
385 sampled children are highly correlated to blood lead levels and are not take into account in the  
386 sampling design or in the post-stratification. Following post-stratification, the proportions of  
387 complementary free health insurance (CMUc)-covered children, children and mothers born  
388 abroad and parents' occupational categories were essentially similar for the estimate for the  
389 population and national census data (Insee 2012). So, in fine, the potential selection bias, if  
390 any, seems minimal.

391 Second, although the quantile regression was more powerful than logistic regression, we did  
392 not have a sufficient sample size to study all the risk factors for extreme BLLs and the  
393 precision of the estimates was low.

394 And finally the design of the study did not allow us to assess past exposure of the children,  
395 food contribution, or exposure sources outside the home such as nursery, primary school or  
396 day care center.

397

## 398 **5. Conclusion**

399 This study provided insights into the many determinants of BLLs in a nationally  
400 representative sample of children with a large range of measures sources of exposure and  
401 harmonized data collection. We found that tap water, floor dust, cookware and cosmetics  
402 containing lead had a substantial contribution on the BLLs of children in France. For dust,  
403 even very low dust lead loadings had a noticeable contribution on BLLs and thus attempts to  
404 reduce dust lead loadings towards zero would be very beneficial for a large proportion of  
405 children. Furthermore, lead in dust had a greater contribution on elevated BLLs than on low  
406 BLLs; therefore, such measures would also benefit to the most exposed children. Most  
407 household tap water had a lead concentration lower than the current European standard;  
408 however, water remains a risk factor for elevated BLLs. Further reduction in lead  
409 concentration is necessary to limit the risk of lead exposure in children (Scientific Committee  
410 on Health and Environmental Risks (SCHER) 2011) and also pregnant woman as fetal death  
411 can occur at very low levels of water lead exposure (Edwards 2014). Protective measures as  
412 water lead filters use or exclusion of tap water consumption should be provided until the  
413 complete lead pipes removal. Exposure to cosmetics and dishes containing lead were also

414 prevalent and were associated with an increase in BLLs. The use of such cosmetics should be  
415 avoided and traditional dishes should be limited to decorative purposes as far as possible.

416 The prevention of lead exposure is still a major goal for public health considering the absence  
417 of a clear toxicological threshold and the risk of exposure from low levels of lead in  
418 environmental sources.

419

#### 420 **Appendix A and B: Supplemental material**

421 The Appendix A defines the construction of the variables included in the models. The Table  
422 B.5 describes the environmental risk factors of lead exposure in children in France for  
423 leachable lead. The Table B.6 displays the results of the quantile regression model for the  
424 90th quantile of Blood Lead Levels for leachable lead.

425

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431

432 **Conflict of interest:** All authors declare they have no current or potential competing financial  
433 interests.

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**Table 1: Estimated lead levels in blood and environmental sources of exposure, France 2008-2009.**

	n	Min	P25	Median	P75	P95	Max	AM	GM
<b>Blood lead levels (<math>\mu\text{g/L}</math>)</b>	484	2.6	9.8	13	20.1	32.8	307.8	21.4	13.8
<b>Interior floor dust (<math>\mu\text{g/m}^2</math>)<sup>a</sup></b>	470	<2	3.7	8.3	18.9	62.1	796	19.6	8.7
<b>Dust from indoor common areas (<math>\mu\text{g/m}^2</math>)<sup>b</sup></b>	114	<2	18	25.2	43.7	384.1	6271	128.2	32.2
<b>Tap water (<math>\mu\text{g/L}</math>)<sup>c</sup></b>	472	<1	<1	<1	2	6.1	74	1.9	<1
<b>Exterior playground soil (<math>\text{mg/kg}</math>)<sup>d</sup></b>	315	1.7	17.3	27.2	60.2	253.8	3408	73.6	33.9
<b>Exterior playground dust (<math>\mu\text{g/m}^2</math>)<sup>d</sup></b>	53	<2	17	32.2	99	393.2	3225	96	44.4
<b>Presence of lead in deteriorated household paint<sup>e</sup></b>	484	0	0	0	0	0	5.6	0.03	–
<b>Presence of lead in household paint in a good condition<sup>f</sup></b>	484	0	0	0	0	0.52	19.5	0.2	–

n: number of children in the sample with no missing value, P25: 25th percentile, P75: 75th percentile, P95: 95th percentile, AM: arithmetic mean, GM: geometric mean

<sup>a</sup> Arithmetic mean of rooms frequented by the child with dust values below LOQ replaced by LOQ/2.

<sup>b</sup> Arithmetic mean of indoor common areas

<sup>c</sup> Tap water values below LOQ were replaced by raw data.

<sup>d</sup> Where the child plays

<sup>e</sup> Sum(XRF sum of deteriorated paint/surface of each room) per dwelling

<sup>f</sup> Sum(XRF sum of paint in a good condition/surface of each room) per dwelling



**Table 2: Demographic and behavioral characteristics of children aged 6 months to 6 years, France 2008-2009.**

		<b>n</b>	<b>Estimated percentage</b>
<b>Child's age (years)</b>	0.5 - < 1	71	9.7
	1	108	17.1
	2	93	14.6
	3	84	12.7
	4	62	27.1
	5	42	12.8
	6	24	6
<b>Child's sex</b>	Female	229	52.5
	Male	255	47.5
<b>Parents smoking at home</b>	Never	367	78.8
	< 1h/day	36	6.4
	1-2h/day	30	5.1
	2-5h/day	25	6.8
	>5h/day	17	2.9
<b>Exposure to traditional ceramic cookware</b>	Not used by family or measure of lead release <LOQ	414	87.3
	Used by family, but no measure of lead release available	59	11.2
	Used by family, measure of lead release >LOQ	11	1.5
<b>Exposure to traditional cosmetics</b>	Not used by family, or measure of lead release <LOQ	459	96.6
	Used by family, but no measure of lead release available	17	2.6
	Used by family, measure of lead release >LOQ	8	0.8
<b>Parents' occupational risk</b>	No	364	79.3
	Yes	120	20.7
<b>Tap water consumption</b>	No	210	39.5
	Yes	274	60.5

**Table 3: Associations between environmental lead sources and geometric mean of blood lead levels in children in France in 2008-2009.**

	Quantitative variables											
	Percentiles				% increase (95% CI <sup>a</sup> ) of BLL for a change in lead source from p0 to p1 percentile (p0-p1)							
	25	50	95	99	p25-p95		p25-p99		p50-p95		p50-p99	
<b>Lead concentration by source of exposure</b>												
Interior floor dust ( $\mu\text{g}/\text{m}^2$ )	3.7	8.3	62.1	172.4	33	(14 ; 55)	34	(3 ; 74)	16	(0 ; 35)	17	(-10 ; 52)
Dust from indoor common areas ( $\mu\text{g}/\text{m}^2$ ) <sup>b</sup>	0	0	92.4	562.0	4	(-12 ; 22)	21	(-13 ; 70)	4	(-12 ; 22)	21	(-13 ; 70)
Tap water ( $\mu\text{g}/\text{L}$ ) children drinking tap water	0.4	0.9	5.1	13.9	8	(-9 ; 28)	36	(2 ; 82)	14	(-2 ; 33)	44	(8 ; 92)
children drinking bottled water	0.4	0.8	6.0	21.0	-13	(-30 ; 7)	-11	(-38 ; 28)	-12	(-28 ; 28)	-10	(-37 ; 29)
Exterior playground soil ( $\text{mg}/\text{kg}$ ) <sup>c</sup>	0	18.8	250.7	407.1	13	(-8 ; 35)	16	(-9 ; 42)	8	(-5 ; 21)	10	(-6 ; 27)
Exterior playground dust ( $\mu\text{g}/\text{m}^2$ ) <sup>c</sup>	0	0	32.1	187.5	18	(4 ; 32)	33	(8 ; 58)	18	(4 ; 32)	33	(8 ; 58)
Sum(XRF sum of deteriorated paint/surface of each room) per dwelling	0	0	0	0.9	0	—	0	—	0	—	5	(-7 ; 20)
Sum(XRF sum of paint in a good condition/surface of each room) per dwelling	0	0	0.5	3.3	2	(-2 ; 7)	13	(-15 ; 45)	2	(-2 ; 7)	13	(-15 ; 45)
	Categorical variables											
	% increase (95% CI <sup>a</sup> ) of BLL											
Parents smoking at home				Never	< 1h/day	1-2h/day	2-5h/day	>5h/day				
				Reference	7 (-19 ; 43)	18 (-12 ; 58)	10 (-32 ; 79)	13 (-23 ; 65)				
Exposure to traditional ceramic cookware				No <sup>d</sup>	Possible <sup>e</sup>	Yes <sup>f</sup>						
				Reference	27 (7 ; 50)	56 (4 ; 132)						
Exposure to traditional cosmetics				No <sup>d</sup>	Possible <sup>e</sup>	Yes <sup>f</sup>						
				Reference	4 (-31 ; 56)	43 (-4 ; 113)						
Parents' occupational risk				No	Yes							
				Reference	-1 (-14 ; 14)							

<sup>a</sup> 95% Confidence Interval

<sup>b</sup> Concentrations set to 0 if indoor common areas were absent

<sup>c</sup> Concentrations set to 0 if the child did not play outside. The value for the exterior playground was set to 0 for soil if a hard surface was sampled and was set to 0 for dust if soil was sampled.

<sup>d</sup> Not used by family or measure of lead release <LOQ

<sup>e</sup> Used by family but no measure of lead release available

<sup>f</sup> Used by family, measure of lead release >LOQ

**Table 4: Associations between environmental risk factors of lead exposure sources and the 90th quantile of blood lead levels in children in France in 2008-2009.**

Lead concentration by source of exposure	Percentiles				% increase (95% CI) of BLL for a change in lead source from <i>p0</i> to <i>p1</i> percentile ( <i>p0-p1</i> )								
	25	50	95	99	p25-p95		p25-p99		p50-p95		p50-p99		
Interior floor dust ( $\mu\text{g}/\text{m}^2$ )	3.7	8.3	62.1	172.4	75	(29 ; 139)	96	(30 ; 195)	50	(20;87)	67	(18 ; 137)	
Dust from indoor common areas ( $\mu\text{g}/\text{m}^2$ ) <sup>b</sup>	0	0	92.4	562.0	13	(-13 ; 45)	26	(-23 ; 104)	13	(-13;45)	26	(-23 ; 104)	
Tap water ( $\mu\text{g}/\text{L}$ )	children drinking tap water	0.4	0.9	5.1	13.9	35	(-2 ; 85)	55	(-10 ; 167)	25	(-10;73)	43	(-13 ; 135)
		children drinking bottled water	0.4	0.8	6.0	21.0	36	(-7 ; 98)	66	(-18 ; 238)	26	(-5;67)	55
Exterior playground soil ( $\text{mg}/\text{kg}$ ) <sup>c</sup>	0	18.8	250.7	407.1	-7	(-29 ; 21)	-9	(-35 ; 28)	-6	(-23;16)	-8	(-30 ; 22)	
Exterior playground dust ( $\mu\text{g}/\text{m}^2$ ) <sup>c</sup>	0	0	32.1	187.5	-5	(-24 ; 18)	-9	(-37 ; 34)	-5	(-24;18)	-9	(-37 ; 34)	

<sup>a</sup> Confidence Interval

<sup>b</sup> Concentrations set to 0 if indoor common areas were absent

<sup>c</sup> Concentrations set to 0 if the child did not play outside. The value for the exterior playground was set to 0 for soil if a hard surface was sampled and was set to 0 for dust if soil was sampled.

**Table B.5: Associations between environmental risk factors of lead exposure sources and geometric mean of blood lead levels in children in France in 2008-2009 (leachable lead).**

Quantitative variables												
Lead concentration by source of exposure	Percentiles				% increase (95% CI <sup>a</sup> ) of BLL for a change in lead source from p0 to p1 percentile (p0-p1)							
	25	50	95	99	p25-p95		p25-p99		p50-p95		p50-p99	
Interior floor dust ( $\mu\text{g}/\text{m}^2$ )	3.0	6.5	47.8	91.3	38	(17 ; 62)	44	(14 ; 82)	22	(5 ; 42)	27	(1 ; 61)
Dust from indoor common areas ( $\mu\text{g}/\text{m}^2$ ) <sup>b</sup>	0	0	56.7	412.2	0	(-14 ; 16)	16	(-17 ; 61)	0	(-14 ; 16)	16	(-17 ; 61)
Tap water ( $\mu\text{g}/\text{L}$ ) children drinking tap water	0.4	0.9	5.1	13.9	7	(-10 ; 27)	36	(1 ; 82)	13	(-3 ; 32)	44	(8 ; 92)
children drinking bottled water	0.4	0.8	6	21	-14	(-30 ; 7)	-12	(-39 ; 28)	-13	(-28 ; 28)	-11	(-38 ; 28)
Exterior playground soil ( $\text{mg}/\text{kg}$ ) <sup>c</sup>	0	12.2	190	359.6	11	(-9 ; 31)	13	(-11 ; 38)	6	(-5 ; 18)	9	(-7 ; 26)
Exterior playground dust ( $\mu\text{g}/\text{m}^2$ ) <sup>c</sup>	0	0	21	155.2	16	(4 ; 28)	30	(7 ; 54)	16	(4 ; 28)	30	(7 ; 54)
Sum(XRF sum of deteriorated paint/surface of each room) per dwelling	0	0	0	0.9	0	—	0	—	0	—	5	(-7 ; 19)
Sum(XRF sum of paint in a good condition/surface of each room) per dwelling	0	0	0.5	3.3	2	(-2 ; 7)	14	(-14 ; 46)	2	(-2 ; 7)	14	(-14 ; 46)
Categorical variables												
% increase (95% CI <sup>a</sup> ) of BLL												
Parents smoking at home	Never				< 1h/day		1-2h/day		2-5h/day		>5h/day	
	Reference				7 (-22 ; 45)		17 (-14 ; 60)		7 (-37 ; 81)		14 (-23 ; 65)	
Exposure to traditional ceramic cookware	No <sup>d</sup>				Possible <sup>e</sup>		Yes <sup>f</sup>					
	Reference				28 (8 ; 52)		54 (6 ; 124)					
Exposure to traditional cosmetics	No <sup>d</sup>				Possible <sup>e</sup>		Yes <sup>f</sup>					
	Reference				7 (-32 ; 67)		44 (-10 ; 129)					
Parents' occupational risk	No				Yes							
	Reference				0 (-14;15)							

<sup>a</sup> 95% Confidence Interval

<sup>b</sup> Concentrations set to 0 if indoor common areas were absent

<sup>c</sup> Concentrations set to 0 if the child did not play outside. The value for the exterior playground was set to 0 for soil if a hard surface was sampled and was set to 0 for dust if soil was sampled.

<sup>d</sup> Not used by family or measure of lead release <LOQ

<sup>e</sup> Used by family but no measure of lead release available

<sup>f</sup> Used by family, measure of lead release >LOQ

**Table B.6: Results of the quantile regression model for the 90th quantile of Blood Lead Levels in children, France 2008-2009 (leachable lead).**

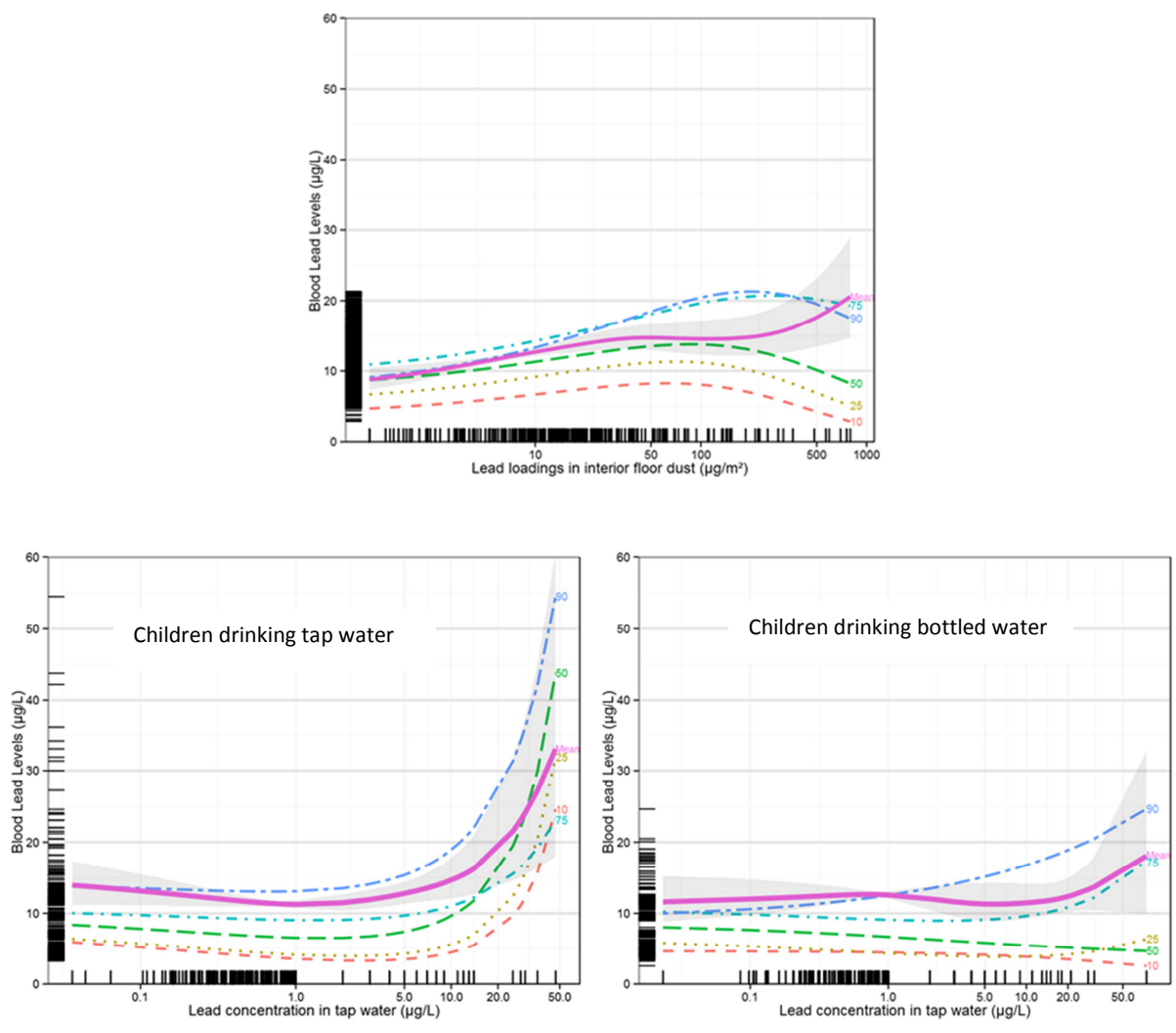
Lead concentration by source of exposure	Percentiles				% increase (95% CI) of BLL for a change in lead source from <i>p0</i> to <i>p1</i> percentile ( <i>p0-p1</i> )								
	25	50	95	99	p25-p95		p25-p99		p50-p95		p50-p99		
Interior floor dust ( $\mu\text{g}/\text{m}^2$ )	3.0	6.5	47.8	91.3	67	(23 ; 125)	104	(36 ; 204)	44	(17 ; 77)	76	(25 ; 148)	
Dust from indoor common areas ( $\mu\text{g}/\text{m}^2$ ) <sup>b</sup>	0	0	56.7	412.2	17	(-10 ; 50)	35	(-18 ; 120)	17	(-10 ; 50)	35	(-18 ; 120)	
Tap water ( $\mu\text{g}/\text{L}$ )	children drinking tap water	0.4	0.9	5.1	13.9	24	(-6 ; 65)	81	(-2 ; 237)	23	(-2 ; 55)	79	(-2 ; 227)
	children drinking bottled water	0.4	0.8	6.0	21.0	33	(-5 ; 87)	57	(-17 ; 196)	24	(-3 ; 59)	46	(-22 ; 173)
Exterior playground soil ( $\text{mg}/\text{kg}$ ) <sup>c</sup>	0	12.2	190.0	359.6	-10	(-33 ; 21)	-13	(-41 ; 29)	-8	(-26 ; 15)	-11	(-35 ; 23)	
Exterior playground dust ( $\mu\text{g}/\text{m}^2$ ) <sup>c</sup>	0	0	21.0	155.2	1	(-19 ; 27)	3	(-31 ; 53)	1	(-19 ; 27)	3	(-31 ; 53)	

<sup>a</sup> Confidence Interval

<sup>b</sup> Concentrations set to 0 if indoor common areas were absent

<sup>c</sup> Concentrations set to 0 if the child did not play outside. The value for the exterior playground was set to 0 for soil if a hard surface was sampled and was set to 0 for dust if soil was sampled.

**Figure 1: Predicted mean and 10<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup> and 90<sup>th</sup> quantiles of blood lead levels as a function of the lead levels in floor dust and tap water, France 2008-2009.**



## **Appendix A\_ Description of the selected variables introduced in the models**

We averaged lead loadings of floor dust samples that were collected in rooms frequented by the child in each home. In the absence of an indoor common area, dust lead levels were set to 0. We introduced an interaction between water lead concentrations and tap water consumption. We used two variables for the exterior playground because the main outdoor play area was either a hard surface (providing loadings expressed in  $\mu\text{g}/\text{m}^2$ ) or soil (providing concentrations in  $\text{mg}/\text{kg}$ ). When the soil was collected from a hard surface, the lead concentration in soil was set to 0  $\text{mg}/\text{kg}$ , and vice versa. The presence of lead in paints was expressed as the sum of the XRF measurements in each room divided by the room's surface. The measures were added up by dwelling depending on the condition of the paint ("deteriorated" in the presence of chalking or chipping or in "good condition" in the presence of traces of shocks or micro-cracking). For glazed ceramics or traditional cosmetics we constructed a variable with three categories: 1) no exposure (the family either does not use ceramic cookware or traditional cosmetics, or lead released from their use is below LOQ), 2) use with no available measure for lead released from these sources, 3) use with a concentration of lead release above the LOQ. The parents' occupational risk of lead exposure was assessed by a questionnaire, using a list of occupations and professional activities defined by toxicological experts.

No values below LOQ were observed for outdoor dust and soil concentrations. Values below LOQ were assigned a value of LOQ/2 for indoor dust (1.8% of values in homes, 0% in common area). For tap water values below LOQ (53%) or LOD (18%), we included raw data despite their high uncertainty. We favored this method over a replacement by a single value (e.g. LOQ/2 or LOD/2) to avoid biased variance estimates and distorted inferences (Lubin et al. 2004).