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

3

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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 10th, 25th, 50th, 75th and 90th 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 90th 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 90th 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, 75th and 90th 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 90th 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, 10th, 25th, 50th, 75th and 90th 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 m² 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 m²) 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 HNO₃ 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/m² (0.09 mg/ft²) for leachable lead in dust and 2 µg/m² (0.19 mg/ft²) 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 10th, 25th, 50th, 75th and 90th 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 90th 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 25th or 50th percentile to its 95th percentile and from
178 its 25th or 50th percentile to its 99th 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 90th 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, 10th, 25th, 50th, 75th
184 and 90th 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 95th 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 25th percentile to their 99th 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/ft²] to 62 µg/m² [6
218 µg/ft²]), 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/m² [17 µg/ft²]).

220 **Insert Table 3**

221

222 **3.2.2 Associations between environmental sources and the quantile 90th of BLLs**

223 The risk factors of the 90th quantile (25.7 µg/L) of BLLs are displayed in Table 4. BLLs ≥ the
224 90th 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 (99th 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 10th, 25th, 50th, 75th, 90th
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 (10th to 50th quantile) increased with increasing dust lead
240 loadings between 1 and 60 µg/m² (5.6 µg/ft²). The 75th and 90th quantiles increased until 200
241 µg/m² (18.6 µg/ft²) and tended to reach a plateau or to decrease above 200 µg/m². 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/m². 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 (75th and 90th quantile) and concentrations higher than 10
249 $\mu\text{g/L}$ affected the lower quantiles (10th and 25th). 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 90th 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 75th 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 75th
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, 75th and 90th quantiles of
298 BLLs for consumers and on the 90th 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 90th 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 75th 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 90th 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 10th, 25th
327 and 75th 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).

368

369

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

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

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

^b Arithmetic mean of indoor common areas

^c Tap water values below LOQ were replaced by raw data.

^d Where the child plays

^e Sum(XRF sum of deteriorated paint/surface of each room) per dwelling

^f 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.

		n	Estimated percentage
Child's age (years)	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
Child's sex	Female	229	52.5
	Male	255	47.5
Parents smoking at home	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
Exposure to traditional ceramic cookware	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
Exposure to traditional cosmetics	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
Parents' occupational risk	No	364	79.3
	Yes	120	20.7
Tap water consumption	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 ^a) 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	
Lead concentration by source of exposure												
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 ($\mu\text{g}/\text{m}^2$) ^b	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 (mg/kg) ^c	0	18.8	250.7	407.1	13	(-8 ; 35)	16	(-9 ; 42)	8	(-5 ; 21)	10	(-6 ; 27)
Exterior playground dust ($\mu\text{g}/\text{m}^2$) ^c	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 ^a) 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 ^d	Possible ^e	Yes ^f						
				Reference	27 (7 ; 50)	56 (4 ; 132)						
Exposure to traditional cosmetics				No ^d	Possible ^e	Yes ^f						
				Reference	4 (-31 ; 56)	43 (-4 ; 113)						
Parents' occupational risk				No	Yes							
				Reference	-1 (-14 ; 14)							

^a 95% Confidence Interval

^b Concentrations set to 0 if indoor common areas were absent

^c 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.

^d Not used by family or measure of lead release <LOQ

^e Used by family but no measure of lead release available

^f 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 ($\mu\text{g}/\text{m}^2$) ^b	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 (mg/kg) ^c	0	18.8	250.7	407.1	-7	(-29 ; 21)	-9	(-35 ; 28)	-6	(-23;16)	-8	(-30 ; 22)	
Exterior playground dust ($\mu\text{g}/\text{m}^2$) ^c	0	0	32.1	187.5	-5	(-24 ; 18)	-9	(-37 ; 34)	-5	(-24;18)	-9	(-37 ; 34)	

^a Confidence Interval

^b Concentrations set to 0 if indoor common areas were absent

^c 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 ^a) 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 (µg/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 (µg/m ²) ^b	0	0	56.7	412.2	0	(-14 ; 16)	16	(-17 ; 61)	0	(-14 ; 16)	16	(-17 ; 61)
Tap water (µg/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 (mg/kg) ^c	0	12.2	190	359.6	11	(-9 ; 31)	13	(-11 ; 38)	6	(-5 ; 18)	9	(-7 ; 26)
Exterior playground dust (µg/m ²) ^c	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 ^a) 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 ^d				Possible ^e		Yes ^f					
	Reference				28	(8 ; 52)	54	(6 ; 124)				
Exposure to traditional cosmetics	No ^d				Possible ^e		Yes ^f					
	Reference				7	(-32 ; 67)	44	(-10 ; 129)				
Parents' occupational risk	No				Yes							
	Reference				0	(-14 ; 15)						

^a 95% Confidence Interval

^b Concentrations set to 0 if indoor common areas were absent

^c 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.

^d Not used by family or measure of lead release <LOQ

^e Used by family but no measure of lead release available

^f 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).

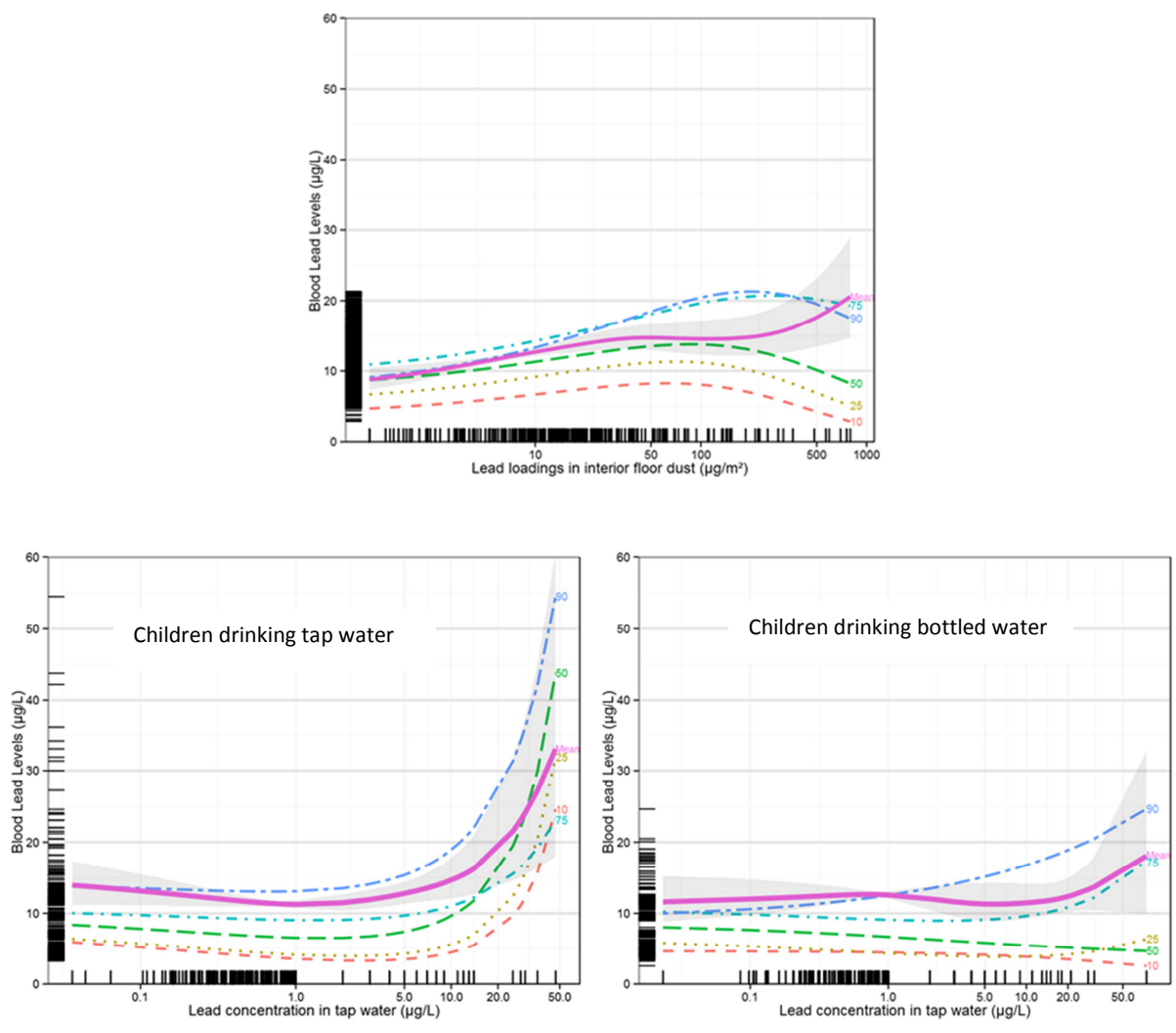
Lead concentration by source of exposure	Percentiles				% increase (95% CI) of BLL for a change in lead source from <i>p0</i> to <i>p1</i> percentile (<i>p0-p1</i>)								
	25	50	95	99	p25-p95		p25-p99		p50-p95		p50-p99		
Interior floor dust ($\mu\text{g}/\text{m}^2$)	3.0	6.5	47.8	91.3	67	(23 ; 125)	104	(36 ; 204)	44	(17 ; 77)	76	(25 ; 148)	
Dust from indoor common areas ($\mu\text{g}/\text{m}^2$) ^b	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 (mg/kg) ^c	0	12.2	190.0	359.6	-10	(-33 ; 21)	-13	(-41 ; 29)	-8	(-26 ; 15)	-11	(-35 ; 23)	
Exterior playground dust ($\mu\text{g}/\text{m}^2$) ^c	0	0	21.0	155.2	1	(-19 ; 27)	3	(-31 ; 53)	1	(-19 ; 27)	3	(-31 ; 53)	

^a Confidence Interval

^b Concentrations set to 0 if indoor common areas were absent

^c 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 10th, 25th, 50th, 75th and 90th 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 mg/kg). When the soil was collected from a hard surface, the lead concentration in soil was set to 0 mg/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).