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► To cite this version:

Roseline Bonnard, Thomas E. Mckone. Integration of the predictions of two models with dose measurements in a case study of children exposed to the emissions from a lead smelter. *Human and Ecological Risk Assessment*, 2009, 15 (6), pp.1203-1226. ineris-00961945

HAL Id: ineris-00961945

<https://ineris.hal.science/ineris-00961945>

Submitted on 4 Apr 2014

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1 **Integration of the predictions of two models with dose measurements in a** 2 **case study of children exposed to the emissions of a lead smelter**

3 **Abstract**

4 The predictions of two source-to-dose models are systematically evaluated with observed
5 data collected in a village polluted by a currently operating secondary lead smelter. Both
6 models were built up from several sub-models linked together and run using Monte-Carlo
7 simulation, to calculate the distribution children's blood lead levels attributable to the
8 emissions from the facility. The first model system is composed of the CalTOX model linked
9 to a recoded version of the IEUBK model. This system provides the distribution of the
10 media-specific lead concentrations (air, soil, fruit, vegetables and blood) in the whole area
11 investigated. The second model consists of a statistical model to estimate the lead deposition
12 on the ground, a modified version of the model HHRAP and the same recoded version of the
13 IEUBK model. This system provides an estimate of the concentration of exposure of specific
14 individuals living in the study area. The predictions of the first model system were improved
15 in terms of accuracy and precision by performing a sensitivity analysis and using field data to
16 correct the default value provided for the leaf wet density. However, in this case study, the
17 first model system tends to overestimate the exposure due to exposed vegetables. The second
18 model was tested for nine children with contrasting exposure conditions. It managed to
19 capture the blood levels for eight of them. In the last case, the exposure of the child by
20 pathways not considered in the model may explain the failure of the model. The interest of
21 this integrated model is to provide outputs with lower variance than the first model system,
22 but at the moment further tests are necessary to conclude about its accuracy.

23

24 **Keywords :** Exposure assessment, multimedia models, probabilistic risk assessment, lead,
25 IEUBK.

26

27 INTRODUCTION

28 Exposure models for chemicals are used around the world to assess the human health risks
29 and to support regulatory decision making. For example, they are used to support policies for
30 pollution control (Bonnard 2006) such as the Hot Spots Program of California Environmental
31 Protection Agency (OEHHA 2003), in which the risks linked to atmospheric emissions from
32 existing facilities are estimated. They are also used in France for Registered Installations for
33 Protection of the Environment (INERIS 2003), which requires a human health risk
34 assessment study to be performed and included in all the environmental impact analyses for
35 new or modified facilities. Exposure models are also employed to estimate the risks linked to
36 manufactured substances, as required by the European regulation for new and existing
37 substances (EC 2003, 2004). Many countries define safety limits with models for various
38 exposure media such as food, water and air (WHO 1993, 2000 and national regulations), as
39 well for soils, for which human health risk assessment is used to identify, prioritise and assess
40 the need for remedial actions (US EPA, Ferguson 1999).

41 Because of the critical role played by models, scientists are required to show that their
42 predictions are reliable. Even though a “model can never be truly validated, (but only
43 invalidated)”(Oreskes 1998, NCR 2007), it is still necessary to evaluate its fitness for use.
44 Part of this process is checking if models yield results matching the observations from field
45 studies, and for the right reasons (that is by providing the correct cause-effect relationships
46 between input and output) and capture this link with a sufficient level of precision.

47 In the context of industrial facilities and contaminated soils, health risk assessment studies
48 employ multimedia exposure models to predict contaminations at a local scale and sometimes
49 employ physiologically based pharmacokinetic (PBPK) models, when the exposure levels
50 need to be expressed in terms of internal doses, as for lead, whose risk management threshold
51 value is usually fixed at 100 µg/l in blood for children.

52 A number of different multimedia models have undergone empirical evaluation in the last
53 decade, but these exercises were carried out at regional or continental scales (Schwartz 2000)
54 and most of the time, they were limited to environmental concentrations (Kawamoto and Park
55 2006, Armitage et al. 2007, Luo and Yang 2007). In the case of the PBPK models, results
56 have been evaluated with subjects in controlled conditions where there were either
57 administrated doses or measured environmental concentrations (Cohen et al. 1998, Hogan et

58 al. 1998, Griffin et al. 1999, Biesiada et al. 1999, Glorennec et al. 2007a), but none of these
59 studies have tracked multimedia transport from a contamination source. Because of the
60 difficulties in collecting for the same period and the same location sufficient data for the
61 source emissions, environmental concentrations, activity patterns, biomonitoring data, and
62 reconstructing past exposures, predictions of multimedia models have seldom been compared
63 with biomonitoring data from the source of contamination. Such an analysis was conducted in
64 a case of contaminated soils by polycyclic aromatic hydrocarbons (Dor et al. 2003), where
65 urinary 1-hydroxypyren data collected from workers were compared to the levels predicted
66 by several multimedia models. That study showed discrepancies among the models tested, in
67 terms of magnitude of the output and of predominant exposure pathways. However, the
68 authors carried out comparisons using a deterministic approach with a point estimate obtained
69 from the different models. Because they did not know the precision of those point estimates,
70 they could not determine to what extent these point estimates were truly different or even
71 comparable.

72 The goal of this paper is to evaluate the performance (accuracy and precision) of two source-
73 to-dose modeling approaches, for a contaminated site by a secondary-lead smelter. Lead
74 contamination was selected because lead is a contaminant often considered in health risk
75 assessments for industrial sites. At such sites, lead can be found in various exposure media, a
76 situation that requires consideration of multiple exposure pathways and provides the
77 opportunity to assess exposure levels both by modeling and measuring blood lead. The
78 comparison of lead measurements and lead modeling results can be carried out at several
79 levels: first in environmental media (such as air or soil), second in exposure media (such as
80 vegetables) and lastly in the human body. The intermediate points of comparison help to
81 determine at which level discrepancies may appear between models and observations and
82 whether the final results are correct for the right reasons. With this approach, one strives not
83 only for good agreement between models and observations (exposure model predictions
84 versus biomarker data) but uses the intermediate steps to confirm consistent hypotheses
85 regarding the magnitude and variation among competing exposure pathways.

86 **METHODS**

87 The lead smelter is located in a French village. The local authorities have ordered many
88 investigations in this village and have implemented a control of the environment media
89 concentrations for several years. The data available were collected in order to carry out the

90 model evaluations and several campaigns of measurements were performed to complete the
91 dataset.

92 Two source-to-dose models, built-up from existing sub-models, were used for this study and
93 run simultaneously by Monte-Carlo sampling from the ranges of parameter input values.

94 The data related to the site and the models used are presented in the following subsections.
95 The data not shown (measurement data and input parameters of the models) are detailed in a
96 report available upon request (Bonnard 2008).

97 **Study Village and Lead Emissions**

98 The village has about 800 inhabitants. The facility producing lead began operations in 1970.
99 It recycles lead from engine-batteries. In relation to the facility, houses are located in a sector
100 between the north-north-east axis to the south-west axis, with the major portion of them been
101 located between the north-east and the south-east axes. The distance from the facility stacks
102 ranges from 100 meters to 1 km. Currently, this smelter is the only facility in the village
103 emitting lead in the atmosphere. From 2000 to the end of 2001, several measures were
104 implemented to reduce the contamination linked to its activity. These measures resulted in a
105 significant reduction of the atmospheric emissions of lead.

106 **Child Biomonitoring Data and Exposure Survey**

107 A biomonitoring campaigns for children's blood lead was carried out in this village in June
108 2002 by the local health administration. A questionnaire was filled out for each child at that
109 sampling time to collect information on his or her exposure conditions. The questionnaire
110 documented age, residence time in the village, places frequented, age of the home, proportion
111 of fruit and vegetables consumed from the family garden, type of water (tap or bottled)
112 consumed, and parents' place of work.

113 To protect the confidentiality of the children, we were not able to obtain the exact location of
114 the house where each child dwelled. Only the distance to the facility was provided, with an
115 indicator, going from 1 to 3, expressing the propensity of the wind blowing from the facility
116 towards the child's house. In addition, a map was provided to us indicating three different
117 sectors assigned a wind propensity value equal to 1, three others a value of 2, and two sectors
118 a value of 3.

119 Among all the children likely to be exposed, 67% took part into the study, namely 97
120 children. Among them, 19 had parents working in the facility. Because the local sanitary
121 authorities have demonstrated a positive link between the children's blood lead and the
122 parents working in the facility for this sample of data (Schmitt et al 2002) and since no
123 multimedia exposure model is currently able to take into account such a relationship, we only
124 kept the lead blood data from the 78 children whose parents did not work in the facility.

125 **Environmental Media Measurements**

126 **Air concentrations**

127 Daily air concentration data are collected from ATMO Champagne (2002), a public
128 association in charge of monitoring the air quality. An air sampler, measuring PM10, was
129 placed in the village at 120 meters from the facility stacks. Air concentrations were measured
130 by atomic absorption spectrometry (AFNOR, 1990).

131 **Plant concentrations**

132 A monitoring program has been conducted by a private company. Each year, it consists of the
133 collection of about 20 samples of fruit or vegetables, from 6 or 7 volunteers' gardens, located
134 between 80 to 600 meters from the facility. All the samples are prepared and washed before
135 being analysed (Prost 2002-2005).

136 **Deposition to the ground surface**

137 A campaign was conducted to measure monthly lead deposition on the ground at 15 locations
138 in the village, using deposit gauges (ISO 1989). These measurements were made in October
139 and November 2005. One duplicate device was used to check the reproducibility and 7 of the
140 samples collected were distributed on the North-East axis relating to the facility, which
141 corresponds to the preferential wind direction. The results of this measurement campaign, as
142 reported in Bonnard (2008), showed a clear decreasing level of deposition with the distance.

143 **Soil concentrations**

144 Several campaigns of soil lead concentration measurements have been performed in the
145 village since 1998. However, because of the location and the depth of the samples, few data
146 could be considered as representative of the population exposure. Thus, there were two

147 additional measurement campaigns in the summer of 2006. The first consisted of 48 surface
148 soil samples and 12 samples taken from the first 20 centimeters. All were composite samples
149 assembled by mixing five sub-samples from the same area. The samples were not distributed
150 according to a regular grid but taken in the village from locations close to dwellings and
151 selected according to accessibility. The surface samples were collected in open areas and in
152 the inhabitants' gardens, where the soil had not been disturbed. All of the the first-twenty-
153 centimeter samples came from the inhabitants' kitchen gardens. The soils were digested by
154 aqua regia and analysed by inductively coupled plasma optical emission spectrometry
155 (ICP/OES) according to the European standard analytical method (ISO 1996, CEN 2000).
156 The second campaign consisted of in-situ measurements made with a portable X-ray
157 fluorescence device. Measurements were performed at 133 locations. In order to evaluate
158 correlation of results between the two methods, some of the fluorescence measurements were
159 made at the location where soil surface analytical samples were collected and 24 additional
160 measurements by X-ray fluorescence were performed on soil samples prepared at the
161 laboratory for extraction before ICP/OES analysis. During the program that monitored lead in
162 vegetables cultivated by inhabitants, measurements of lead in the first twenty centimeters of
163 soil were performed. They were added to our database. A geostatistical study, with a set of
164 156 data corresponding to the lead concentration in the surface soil and with a set of 39 data
165 corresponding to the first-twenty-centimeter soil samples, was achieved.

166 Lead is characterized by a low mobility in soils and tends to accumulate in the surface layers
167 (Adriano 1986, Kabatia-Pendias 1992, Alloway 1995). However, even if the quantities of
168 lead collected in each gauge during our two-month-measurement campaign (while the stack
169 emissions were higher than the 2002 to 2006 average ones) had been deposited on the ground
170 for four years (between 2002 and 2006) and had not moved, that deposition of lead would
171 correspond to an increase of the concentration in the first two centimeter layer of less than
172 5%. Given this low accumulation rate, we assumed that the values measured in the soils in
173 2006 are representative of the concentrations in 2002. Subject to the condition that no other
174 source was added to the soils and the surface soil was not mixed with other soil layers, this
175 assumption seems reasonable.

176 **Source to Dose Modeling**

177 Source to dose modeling was carried out with two alternative integrated models. Both of
178 them link a multimedia fate model with a pharmacokinetic model. The first integrated model

179 is process based and its purpose is to provide a regional mass balance to track lead from a
180 source to receptor using generic parameters. The second model uses an empirical relationship
181 to estimate the lead deposition on the ground and is intended to be both site and receptor
182 specific.

183 **Description of integrated model A**

184 Model A consists of the CalTOX multimedia fate and multi-pathway exposure model
185 (Mckone 1993, 2002) integrated with the exposure-uptake-biokinetic (IEUBK) model (US
186 EPA 1994, White et al 1998). We set up this model system in EXCEL© files, and ran
187 probabilistic assessments with the software Crystal Ball (Decisoneering 2005).

188 CalTOX is a fugacity-based mass balance model in which each environmental medium (air,
189 soil, water, etc) is represented by a homogeneous compartment. IEUBK is the model
190 recommended by the United States Environmental Protection Agency (US EPA) to estimate
191 children's blood lead exposure. It permits calculation of the lead intake and uptake from the
192 exposure media concentrations and then determines the blood lead levels using a biokinetic
193 sub-model. To address the inability of IEUBK to propagate uncertainty and variability from
194 lead intake, Syracuse Research Corporation developed the Integrated Stochastic Exposure
195 (ISE) model (SRC, 2003) using the same conceptual model as IEUBK but allowing for
196 stochastic simulations. We did the same, by developing our own version of IEUBK using
197 EXCEL© and Visual Basic, so that we could link it directly to the stochastic outputs of
198 CalTOX. We checked that our implementation of IEUBK gives exactly the same results as
199 the original code. With this approach our blood lead prediction model could consider
200 temporal variation of lead intake and probabilistic distributions for some inputs defined as
201 point estimates in the ISE model.

202 We accounted for intake of lead through inhalation, soil ingestion, ingestion of local fruits
203 and vegetables, and background exposure from non-local food consumption (Glorennec
204 2007b).

205 Based on atmospheric emissions and the distribution of lead concentrations measured in the
206 first twenty centimeters of soil, model A was used with Crystal Ball to characterize the
207 distributions of lead in the village air, in the surface soil, in the above-ground fruits and
208 vegetables and the below-ground vegetables grown in the village, and finally in the blood of
209 the children of the village.

210 **Description of integrated model B**

211 Model B has three components: (1) a regression model that calculates lead deposition
212 according to the location of the household in the village, (2) a multimedia exposure model
213 that calculates local lead concentrations in environmental and exposure media based on the
214 deposition rate, the surface soil concentration and the soil concentration in the first twenty
215 centimeters and (3) our version of IEUBK recoded in EXCEL©.

216 Model B was built to estimate lead exposures for precise locations, an approach often
217 required for contaminated sites to distinguish the areas requiring remediation from those
218 which do not. Unlike model A, which is aimed to capture the distribution of the
219 concentrations over the whole village, model B enables to represent the exposure of
220 individuals. Because the exact location of each child's house was not known, we had to
221 identify it basing on the concordance among three sources of information: (1) the child's
222 distance from the facility, (2) the sector where the house was located and (3) a bird's-eye
223 view zonal map of the village. In some cases, only one location of household corresponded to
224 information derived from these three sources, in other there were several possibilities.
225 Because of this difficulty, blood lead level was modeled for a subset of nine children for
226 which we had identified only one or two possibilities of housing location. If there were two
227 possible locations for one child's house, two predictions were made for that child's blood
228 lead based on a different soil concentration and a different lead deposition on the ground. The
229 children of this subset had contrasting exposure conditions (high, low or no consumption of
230 home-grown vegetables, close or far from the facility, low or high blood lead level) (Table 1).

231 We use the regression model in place of a classical gaussian air dispersion model because we
232 found the accuracy of these models, which predict deposition and air concentration, at each
233 point of a grid, to be inappropriate in regards of the estimation of fugitive emissions (see
234 section called Source term definition). Instead we fitted deposition data collected during the
235 sampling campaign (see section called Deposition to the ground surface) using a statistical
236 relationship, taking into account the distance (D_i in meters) of a specific location i from the
237 facility, the frequency (W_{ij} unitless) of the wind blowing from the facility towards a specific
238 location during month j and the magnitude of stack emissions (E_j in g/month). We used
239 Statistica (Statsoft 1999) for this analysis. We obtained the higher correlation coefficient and
240 the lower residual standard deviation with the following equation by ordinary least squares:

241
$$\hat{Y}_{ij} = \log dep_{ij} = 0.35 - 1.7 \times 10^{-3} D_i + 1.31 W_{ij} + 1.4 \times 10^{-5} E_j \quad (\rho^2 = 0.93)$$

242 where dep_{ij} = total deposition of lead on the ground at location i during month j (in
243 $mg/m^2/month$).

244 We used a Student's T-test to test if a coefficient of this relationship is significantly different
245 from zero. Although the equation above is based on data collected for only two months, all
246 coefficients were found to be statistically significant. In a good regression model, residuals
247 (difference between the observed values and the predicted values) are normal and
248 homoscedastic (residuals have the same variance). The residuals of the above equation
249 appeared normal and homoscedastic.

250 We characterized the uncertainty of the predicted deposition (\hat{Y}) using a Student's
251 distribution, with 26 degrees of freedom, multiplied by the estimated deviation of \hat{Y} (Foucart
252 1997). We have 26 degrees of freedom because we have 30 data to define a relationship that
253 has 4 coefficients. The estimated deviation of \hat{Y} is a T distribution based on the residual
254 estimation error from the regression model. The output of this relationship along with the
255 error were used as inputs in the multimedia exposure model.

256 The media concentrations were calculated from the equations described in the Human Health
257 Risk Assessment Protocol (HHRAP) document (US EPA 2005). However, a supplemental
258 soil layer (between 0 to 10 cm from the surface) for predicting the forage contamination from
259 soil and additional classes of plants (fruit, leafy vegetables and fruit vegetables) were
260 introduced, as well as equations for plant contamination by irrigation and deposition of
261 resuspended soil particles. The equations used for these pathways are those given in the
262 model ERMYN (US DOe 2003a). The concentrations in air, surface soil and plants yielded
263 by the multimedia exposure model were used as input of the new version of IEUBK to obtain
264 the child blood lead distributions.

265 **Values used for model parameters and inputs**

266 Here we describe how we obtained model inputs. First we consider the period for which we
267 had to model the doses adsorbed by children to estimate their blood lead level at the time of
268 the biomonitoring campaign. Next we consider the case of source terms, lead chemical
269 properties, environmental parameters, intake and uptake parameters. We developed
270 probability distributions for each input parameter of both models.

271 ***Period for reconstructing children's intake***

272 The half-life of lead in blood is estimated to be one month. Although lead is also stocked in
273 bones and slowly released in blood from there, it has been shown that a reduction of child
274 exposure brings out a rather fast reduction of the blood lead (Declercq and Ladrière 2004,
275 Khoury et al 2003). The conceptual model implemented in the IEUBK software also yields a
276 rapid reduction of the blood lead, once intake is stopped. We ran the IEUBK model for the
277 case of a child with a cumulative ingestion dose 3 times higher than the estimated
278 background intake in France for 3 years and then a dose equal to the background intake
279 (Glorennec 2007b). We found that one year after reducing the intake, the blood lead would be
280 only 2 percent higher than the one calculated with the background intake for 4 years.
281 Therefore, the efforts to reconstruct the intake doses were focused on a period starting from
282 spring 2001 to the date of the campaign of blood lead measurements. Beforehand, the
283 exposure conditions were assumed to be the same as those between January and June 2001.

284 ***Source term definition***

285 Model A

286 A lognormal distribution was fitted to the observed lead concentrations in the first twenty
287 centimeters of soil using a Z-plot chart. The resulting distribution with an arithmetic mean of
288 261 mg/kg and a standard deviation of 249 mg/kg was assigned to the root zone soil
289 concentration parameter of CalTOX.

290 We estimate smelter stack emissions from monthly sampling of particule emissions, the
291 measurement of the lead concentration in the emitted particles and the duration of operation.
292 The smelter operators try to limit the fugitive emissions by keeping the pressure inside the
293 buildings lower than the outside pressure, washing outdoor surfaces daily and stocking raw
294 materials products and by-products indoors. Nevertheless, preliminary calculations performed
295 with an air dispersion model (CERC 2000) showed that the air concentrations and particle
296 fallout recorded downwind could not be explained purely by the recorded stack emissions
297 (Bonnard 2008). Therefore, we had to include fugitive emissions. We estimated them using
298 three methods (inverse modeling, predictions based on the emission factors and the
299 production rate, and a calculation based on the indoor air concentration in the buildings of the
300 factory and the ventilation rate) and obtained consistent estimates. With inverse modeling, we
301 calculated the volumetric flux required to get the best correlation between the air dispersion
302 model and the lead deposition measured during the campaign conducted in October and

303 December 2005 and obtained a fugitive emissions estimate between 20 and 60 kg/month.
304 Emissions factors for this type of facility and information on its production rate give fugitive
305 emissions between 25 and 73 kg/month. Data on lead concentration measured in indoor air of
306 the facility building with an assumption of a ventilation rate of 10 times per hour give a
307 fugitive emissions estimate of 34 kg/month.

308 To cover the whole growing period of the local fruit and vegetables consumed till the date of
309 the campaign of blood lead measurements, stack lead emissions were estimated from the data
310 recorded from April 2001 to June 2002. As information on fugitive emissions is poor, a
311 triangular distribution was assigned to the total atmospheric emissions from the estimates
312 obtained for the stack and the fugitive emissions during this period. The mode is equal to
313 1,200 g/d, the minimum to 750 g/d and the maximum to 2,800 g/d.

314 For comparing the predicted vegetation concentrations with the observed ones, the emissions
315 were estimated over the vegetation exposure period to fallout from the facility (from April to
316 July, in 2002 to 2005). The total atmospheric emissions were defined by a triangular
317 distribution with a mode of 780 g/d, a minimum of 450 g/d and a maximum of 1,450 g/d.

318 Model B

319 Spatial soil mean concentration in the land associated to each child's house was estimated by
320 kriging. Cumulative distributions were defined for the surface soil and the root-zone soil
321 owing to the turning band method (Chilès 1999, Bonnard 2008).

322 The atmospheric emissions were estimated from the statistical model described above
323 (Equation 1) for each location and period used to represent the atmospheric deposition on the
324 different kinds of home-grown plants.

325 *Chemical properties for lead*

326 Model A

327 With the exception of the particle-water partition coefficients, we used the default
328 distributions provided by CalTOX for chemical properties—water solubility, partition
329 coefficients, and bioconcentration factors. We found that the particle-water partition
330 coefficient (K_d) in CalTOX was high relative to ranges found in other papers. Since K_d for
331 lead is known to depend on the soil pH, we used the recommendations from the Office of Air
332 and Radiation (US EPA 1999) and the pH values collected in the garden soils of the village to
333 define a triangular distribution. The measured pH going from 5.3 to 8.3, the minimum and

334 maximum values for the closest ranges of soil pH, given by the Office of Air and Radiation,
335 were used as minimum and maximum of the distribution (150 L/kg and 23,279 L/kg
336 respectively). The mode was assigned a value of 1731 l/kg, which was calculated from the
337 modal value of measured pH and the Office of Air and Radiation's relationship between lead
338 Kd and pH.

339 Model B

340 The parameters required for model B are the soil-plant transfer coefficients. To obtain values,
341 we made a review of the primary literature and fitted the resulting dataset of selected values
342 to a lognormal distribution (Table 2, Bonnard 2008).

343 *Environmental parameters*

344 Model A

345 For model A we used the default distributions provided by CalTOX for all environmental
346 parameters except area, rainfall, wind speed, and temperature. We replaced distributions for
347 these parameters in CalTOX by local data. In addition we replace the CalTOX root zone soil
348 depth with a mean value equal to 0.2 m to be consistent with the measured data for the
349 village.

350 Model B

351 Most of environmental parameters required in model B are different from those of CalTOX.
352 They mainly concern the description of the various categories of plants. Distributions were
353 defined from the literature (Table 2, Bonnard 2008).

354 *Intake parameters*

355 From birth to the age of 7, we defined plant consumption and lead exposure from local food
356 consumption for seven age classes based on French surveys (Boggio et al. 1999, Volatier
357 2000).

358 For soil ingestion, we used the cumulative distribution provided by ISE with a median equal
359 to 135 mg/d (US EPA 1994, 1999). We note that other distributions with lower median values
360 were available from empirical data published by Thompson et al. (1991) and Stanek et al.
361 (2000, 2001). However, the IEUBK model was calibrated with a point estimate equal to 135
362 mg/day. On the other hand, the lognormal distribution given in the ISE's model for soil
363 ingestion, has a standard deviation that appears too low relative to what is available in the
364 literature. For example, the ratio of the 95th percentile to the 5th percentile in the ISE

365 distribution is less than 2, indicating almost no inter-individual variability. We did use the
366 ISE age-specific weighting factors for soil ingestion in our recoded version.

367 For model B, we use observed individual data for each child from the study population to
368 obtain the age, the seasonal variation in the percentage of home-grown fruits and vegetables
369 consumed, and the date of moving into the village.

370 *Uptake parameters*

371 The bioavailability of lead in food and the relative bioavailability of lead in soil were revised
372 to better reflect data documented in IEUBK for these factors.

373 The range reported by the USEPA in the IEUBK guidance (US EPA 1994) for absorption of
374 lead from diet is 42 to 53%. These values correspond to the 40th and 63th percentiles of the
375 lognormal distribution defined in the ISE model for this parameter (mean = 0.5, standard
376 deviation = 0.2). A lower value was used for the standard deviation (0.05 instead of 0.2) to
377 get a distribution more in accordance with the range of data given above (in these conditions,
378 the low and high values given by the US EPA correspond to the 5th and 75th percentiles of the
379 new distribution defined for absorption of lead from diet).

380 For the absolute bioavailability of soil lead, ISE gives a point estimate of 0.3, but no
381 distribution is defined. We used the values collected by Ruby et al. (1999) for lead uptake
382 from soils (data from wastes and mines were eliminated), to define a normal distribution with
383 a mean equal to 0.6 and a standard deviation equal to 0.2 for the relative bioavailability of soil
384 lead. The mean of the soil lead absolute bioavailability obtained is then close to the point
385 estimate used in the IEUBK and ISE models.

386 **Probabilistic computation**

387 All of our calculations were based on Monte Carlo simulations using 5000 runs with latin-
388 hypercube sampling. We used the Monte Carlo results to develop cumulative probability
389 distributions that can be compared to cumulative distributions of measured concentrations in
390 the media.

391 RESULTS AND DISCUSSION

392 Model A

393 Because no significant statistical relationship between the blood level and the age could be
394 identified (Schmitt et al. 2002), we analysed the model's output as the mean of the blood lead
395 from 6 months to 7 years old.

396 Figure 1 shows the probability plot charts for the (base 10) logarithms of the observed and
397 predicted concentrations in the different exposure media. On these charts, concentrations in
398 the y-axis are represented against the cumulative probability expressed as the number of
399 standard deviation from the geometric mean. For blood lead, figure 1e shows the
400 concentrations calculated by the integrated model and those predicted with the new version of
401 IEUBK from the measurements performed in the samples of surface soil, fruit and vegetables
402 collected in the village.

403 The observed values in air, ground soil, exposed produce and blood lead tend to follow
404 lognormal distributions. However, concentrations above 500 mg/kg in the ground soil deviate
405 from a lognormal distribution (these values correspond to a small area close to the facility
406 and without dwelling). The distributions of the calculated concentrations in air and ground
407 soil also follow lognormal distributions, whereas the distributions of the predicted
408 concentrations for the protected produce, the exposed produce and in particular for the blood
409 lead are characterized by two different slopes, indicating bimodal distributions.

410 Except for the exposed produce, the median values predicted by the integrated model are in
411 good agreement with the observed medians. The predicted median-to-observed median ratios
412 are between 0.5 and 1.3 for these media. But for the exposed produce, the ratio is equal to 18.

413 Regarding distributions, the observed data for ground soil are well captured until 500 mg/kg.
414 The ranges of the observed and predicted values for the air concentrations are close and the
415 two distributions are quite similar. But for protected produce, the distribution of the observed
416 data is underestimated and above all the distribution of the observed concentrations for
417 exposed produce is overestimated. For blood lead, the variability of the calculated
418 concentrations is higher than that of the observed concentrations. In the case of
419 concentrations predicted from the measurements performed in the exposure media, blood lead
420 concentrations are underestimated up to the 90th percentile, then they are overestimated. The

421 values predicted from the integrated model are higher and the difference between the two
422 distributions of calculated concentrations increased with percentiles.

423 According to figure 2, even though emissions are primarily to air, inhalation of smelter
424 emissions is not a major exposure pathway, because most of the lead emitted to air deposits
425 to soil and from there is transferred to the children by various ingestion pathways.

426 When blood lead level is predicted from the exposure media concentrations, soil ingestion
427 appears as the predominant pathway in the upper percentiles of the distribution of the
428 predicted blood lead concentrations, as in former studies conducted near smelters (Roels
429 1980, Gulson 1994, Landrigan 1996). However, when blood lead level is calculated by the
430 integrated model, the ingestion of exposed produce appears as the main pathway in the last
431 quarter of the blood lead distribution. Therefore, the overestimation of the exposure produce
432 concentrations by the integrated model contributes to the overestimation of the upper part of
433 the distribution of blood lead concentrations calculated by this model.

434 Analysis of the contributions to exposed produce contamination showed that in the first part
435 of the distribution, transfer from air and from surface soil to plants are higher than transfer
436 from root-soil, and then the situation is reversed. In order to understand the reasons for the
437 overestimation of the exposed produce concentrations, we carried out a sensitivity analysis
438 with Crystal Ball. The parameters with the highest contributions to the variance are listed in
439 Table 3 in descending order.

440 The distributions of the first two parameters with the highest sensitivity (particle-water
441 partition coefficient and root-soil concentration) were defined by taking into account specific
442 data from the studied site. The third parameter is the leaf wet density (ρ_{leaf}). The default
443 value, which was used in the computations, is equal to 820 kg/m^3 . With a volume fraction of
444 water in leaf (β_{leaf}) equal to 0.5, as provided in CalTOX, we get a value of 0.61 for the
445 mass water fraction ($0.5 \times 1000 / 820$), whereas the data for the exposed produce (leafy
446 vegetables) collected in the gardens ranged from 0.77 to 0.98. So, we determined a new value
447 for the leaf wet density on the basis of the mean of the leaf mass water fraction measured in
448 the collected samples of exposed produce (0.91). We found a value equal to 550 kg/m^3 . It
449 remains in accordance with the typical values used in models for leaf wet density, which are
450 comprised between 500 and 900 kg/m^3 (Riederer 1990, Paterson et al. 1991, Bacci et al.
451 1992, Trapp and McFarlane 1994, Maccrady and Maggard 1995).

452 As a result, the distribution obtained for the exposed produce concentrations (figure 3a, curve
453 3) is closer to that of the observed concentrations. The predicted concentrations were divided
454 nearly by a factor 2, except at the lower tail of the distribution. The air, soil and protected
455 concentration distributions were not altered by this change, but the variance in the predicted
456 blood-lead concentrations (calculated by the integrated model from the predicted
457 concentrations in produce) was reduced (figure 3b, curve 3).

458 For the other parameters pointed out in the sensitivity analysis, we found no new data
459 justifying any modifications of the default values and significantly improving the prediction
460 of the exposed produce concentrations.

461 But, we needed to consider that fruits and vegetables had been washed before the analyses
462 and recognized that the impact of washing is not considered in the model. Unfortunately, the
463 extent to which such a treatment reduces the contribution of air deposition and rain-splash to
464 fruit and vegetables contamination is not well known. In our model, if the contributions of
465 both these pathways are divided arbitrarily by ten, it appears clearly that the lower part of the
466 predicted distribution (figure 3a, curve 4) is closer to that of the observed concentrations data.
467 That means that the variability of the measurements is then captured better by the model. The
468 model over-predicts the concentration in exposed fruits and vegetables by a factor 4 at the
469 median, under these assumptions. The distribution of the blood lead concentrations predicted
470 from the integrated model is then very close to that predicted by the new version of IEUBK
471 from the exposure media measurements (figure 3b, curves 4 and 5).

472 Nevertheless, these two distributions still deviate from the distribution of the observed blood
473 lead at the upper percentiles and according to the integrated model, the ingestion of
474 homegrown plants remains the main contributor to the blood lead level above the 90th
475 percentile. This is not in accordance with the result of the univariate analysis of observed data
476 showing a higher mean of blood lead concentration for no consumers of homegrown plants
477 ($m=65 \mu\text{g/l}$, $n=46$) than for consumers ($m=45 \mu\text{g/l}$, $n=32$), even if for the group of
478 homegrown plant consumers, the blood lead level increases with the fraction of homegrown
479 plants in the diet. The discrepancy between observed and predicted data may be due to the
480 fact that in the sample of children tested, those with the highest consumption of kitchen
481 garden foods were those living the farthest away from the facility, thereby with the lowest
482 lead concentrations in plants. In other words, the actual consumption of home-grown produce
483 in the village and its contribution to blood lead level at the upper percentiles is overestimated

484 by the model, which assumes a homogeneous distribution of the home-grown produce
485 consumptions within the village.

486 **Model B**

487 The results of model B are provided in Figure 4 for nine children with one or two possible
488 housing locations. We can see that the measured blood lead levels are between the 5th and the
489 95th percentiles of the model results, with the exception of child 43. The measured blood-lead
490 levels correspond to various percentiles of the distribution given by the model, going from
491 the 25th to the 88th percentile according to the different children. Measured values of blood
492 lead above the predicted median ratios are between 0.7 and 1.7 (excluding the results
493 corresponding to child 43).

494 Model B was able to capture the blood-lead-level ranges for the various conditions tested,
495 except for child 43. The discrepancy between the model output and the blood lead level
496 measured for this child may be explained by the fact that this child lived in an old house built
497 before the ban of lead for interior paint. The univariate analysis of the data of the child
498 biomonitoring and of the exposure survey showed a higher mean of blood lead concentration
499 for children living in houses built before 1948 than for children living in more recent houses
500 (DDASS 2002). But, this exposure pathway is not taken into account by the model.

501 It has to be noted that using either the Thompson et al. (1991) or Stanek et al. (2000, 2001)
502 distributions for soil ingestion, instead of a distribution centered on the value used to calibrate
503 IEUBK for soil ingestion, results in lower estimates of blood lead concentrations, especially
504 when the soil ingestion is a significant contributor to overall exposure. For example soil
505 ingestion dominates when the children consume few or no homegrown foods, and when the
506 soil lead concentration is high. In the sub-sample tested here, the geometric mean of the
507 measured median above the calculated value ratio would increase from 1.1 to 1.3, if the
508 distribution used for the quantity of soil ingested was replaced by Thompson et al.'s
509 distribution and it would reach a higher value with Stanek et al.'s distribution.

510 Many studies showed a correlation between blood lead level and soil lead (Xintaras 1992,
511 Landrigan 1996, Mielke et al 1998). However, for instance, for child 71, who did not
512 consume kitchen garden plants and who lived close to the facility, model B indicates that
513 exposure due to ingestion of no local food is slightly higher than exposure due to soil
514 ingestion. This is certainly due to the fact that soil lead concentration at the location of his

515 house is not high (200-230 mg/kg) compared to values measured in these studies. For child
516 28, who ate a large amount of home-grown plants and lived relatively far from the facility,
517 the ingestion of homegrown produce appears as the largest contributor to the blood lead level.
518 Wilhem et al. (2005), who studied the dietary intake of lead by two groups of children living
519 in an industrial region, found no difference between those eating a large part of produce from
520 the families own vegetable garden and those consuming exclusively food from the
521 supermarket. But in this case, comparison was made at a region scale without an identified
522 local source of lead. Here, measures of concentrations in plants from child 28's garden would
523 be necessary to confirm the role played by the ingestion of homegrown plants.

524 The uncertainties in the modeled blood lead predictions for model B are reflected by the
525 ratios of the 95th percentile to the 5th percentile, which are between 2.6 and 4.9. The widest
526 90% interval is given for child 28. In this case, the root soil concentration is the highest
527 contributor to the variance. This high contribution is due to a lack of precision in the
528 estimation of the root soil concentration in the child's living area. Increasing the sampling
529 efforts in soils of the farthest areas of the village could permit the reduction of the overall
530 uncertainty. Nevertheless, model B is able to predict blood lead distributions with lower
531 variance than model A (the ratio of the 95th percentile to the 5th percentile is equal to 10.2
532 with model A). Indeed, model A has to integrate inter-individual variability in the input
533 distributions, while model B took it into account by a better description of the influence of
534 the distribution of lead deposition and individual characteristics.

535 **Limitations of the comparison between predicted data and observed data**

536 Finally, we acknowledge that the exercise of comparison made with both models may be
537 limited by the quality of data used to perform it.

538 Soil lead concentration appeared as the most sensitive parameter for the blood lead
539 predictions performed. However, it was measured in bulk soil and not in the finest particle
540 fraction, more likely representative of lead concentration in soil ingested by children.
541 Campaigns for soil lead and blood lead concentrations were not conducted at the same time.
542 (so, there is a possibility that soil was moved in some areas of the village between the two
543 campaigns) and soil lead was not measured in children's garden soil, but estimated by a
544 geostatistical study. Air emissions used in model A were also estimated from indirect
545 methods.

546 On the other hand, the lack of measurement data in the village for tapwater and lead
547 bioavailability, which is usually recognized as a determining factor for estimating blood lead
548 level (Mushak 1998), should not represent a major fault. For drinking water, the analysis of
549 observed data showed that there is no significant difference between blood lead level of
550 children drinking tapwater and those drinking bottled water. Lead bioavailability is not either
551 a very sensitive parameter in the previous calculations. The absolute lead bioavailability and
552 the relative bioavailability of soil lead together contribute to 3% of variance in model A and
553 to 7% in model B at the maximum.

554 **CONCLUSIONS**

555 Both models, built for estimating human health risks linked to future emissions of facilities,
556 may provide useful insight regarding source-to-dose relationships for the exposure
557 population. Model A may be used to address general trends and model B is more focused on
558 capturing individual exposures. In considering the results presented above, there is an issue
559 that cuts across our evaluation of the individual models and data. In this process, we
560 recognized that not only did we gain insight into the two models, but by making a systematic
561 comparison of two models with a specific case study, we gained important insights about lead
562 exposure in these communities that could not be obtained from either model or the
563 environmental/biomonitoring data used in isolation. Thus we conclude that using the two
564 models together with population and site-specific measurements of blood lead and
565 environmental conditions provides for this population's lead exposure patterns a level of
566 understanding that could not be achieved with either model alone or with just the blood
567 surveillance measurements.

568 Model A provides an ability to capture the distributions of lead in the media and the blood
569 lead concentrations at a local scale. It offers the opportunity to identify the potential pathways
570 of concern and to characterize also the distribution of the exposure level. From the initial
571 results of the model, mainly used with default values, the information from the sensitivity
572 analysis and the pathway contributions evaluation help to identify the parameter
573 improvements needed to calibrate the model to this case. However, predicted concentrations
574 in exposed vegetables may be a potential source of overestimation of blood lead level.

575 Model B provides outputs that can be targeted to specific areas and exposure conditions. It
576 was set up to answer some questions, such as "in which zones or from which distance may
577 the consumption of home-grown produce raise a health concern ?" Because of this feature,

578 model B can predict blood lead distributions with lower variance than model A. Precision of
579 assessment is important for discriminating those situations that require management measures
580 from those which do not, especially if the risk management threshold is low. Although, the
581 results provided by model B, for contrasting exposure conditions, are in accordance with the
582 measurements (apart from a case that may involve an exposure pathway that is not considered
583 in the model), the low number of cases tested here does not enable to conclude on the
584 performances of model B at the moment.

585 **ACKNOWLEDGEMENTS**

586 The authors thank the local administration (Direction Régionale de l'Industrie, de la
587 Recherche et de l'Équipement de Champagne-Ardennes and Direction Départementale des
588 Affaires Sanitaires et Sociales des Ardennes), the director's facility, ATMO Champagne and
589 the company Gestion des sites et des sols contaminés (GSC) for communicating their data, as
590 well as the studied village inhabitants who permitted us to take samples from their garden and
591 the mayor for his help. We also thank F. Bois of Institute of Industrial Environment and
592 Risks for his review of the paper and M. Sohn of Lawrence Berkeley National Laboratory for
593 his guidance and input on this work. R. Bonnard's work was supported by the French
594 Ministry of Ecology and Sustainable Development (Budget Civil de la Recherche et du
595 Développement, grant BRCD-03-DRC-06). T. McKone was supported in part by the US
596 Environmental Protection Agency through Interagency Agreement DW-988-38190-01-0
597 carried out through the US Department of Energy contract Grant No. DE-AC02-05CH11231.
598 McKone was also supported by Cooperative Agreement Number U19/EH000097-03 from the
599 US Centers for Disease Control and Prevention (CDC).

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794
795

796 Table 1: Cases investigated with the model B (because of uncertainty in the location of some
 797 children's houses, there are two entries for some children based on two different assumptions
 798 for soil concentrations and lead deposition around their houses)

child number identification	age (month)	blood lead ($\mu\text{g/l}$)	distance to the stacks (m)	% home- grown fruit	% of home- grown vegetables	surface soil lead concentration in the location of the child's home (mg/kg)
48	33	67.1	250	0	50	173
28	47	83.1	900	75	75	96
28	47	83.1	900	75	75	84
27	74	41.5	900	62.5	75	96
27	74	41.5	900	62.5	75	84
41	52	31.9	450	10	20	112
41	52	31.9	450	10	20	103
71	65	44.7	150	0	0	203
71	65	44.7	150	0	0	227
90	37	44.7	300	0	0	261
90	37	44.7	300	0	0	174
97	81	22.3	1000	0	0	66
43	21	108.7	450	0	0	112
43	21	108.7	450	0	0	103
50	71	99.1	200	0	0	493

799
800

801 Table 2: Distributions assigned to environmental and chemical parameters of model B

Parameters	Distributions	References
Fraction of wet deposition that adheres to plant surfaces (-)	U (0.15, 1) ^a	GRNC 2002, US EPA 2005
Deposition velocity of particles (m/s)	CUM ^b perc. ^c 0: 3 E-4 perc. 84: 3 E-2 perc. 16: 1 E-3 perc. 100: 3 E-1 perc. 50: 8 E-3	US DOe 2003b, c
Atmospheric mass loading of particles (kg/m ³)	T (2.5 E-8, 1.2 E-7, 2.0 E-7)	ECETOC 1992, US DOe 2003b, c
Yield of crops (kg/m ²)		
Leafy vegetables	U (0.1, 0.5)	GRNC 2002, US DOe 2004
Fruiting vegetables	U (0.1, 0.3)	US DOe 2004
Fruits	U (0.2, 0.5)	GCNC 2002, US DOe 2004
Plant surface loss coefficient (year ⁻¹)		
Leafy vegetables	T (11, 26, 51) ^d	GRNC 2002
Fruiting vegetables and fruits	T (8.4, 18, 51)	US DOe 2004
Length of plant exposure to deposition (year)		
Leafy vegetables	T (0.12, 0.21, 0.58)	GCNC 2002, GRNC 2002, US EPA 2005
Fruiting vegetables and fruits	T (0.16, 0.25, 0.41)	
Interception fraction of the edible portion of crops (-)		
Leafy vegetables	T (0.10, 0.20, 0.50)	Baes 1984, GCNC 2002, GRNC 2002. US EPA 2005
Fruiting vegetables	T (0.03, 0.15, 0.15)	
Fruits	T (0.03, 0.10, 0.10)	
Dry matter of plants (%)		
Leafy vegetables	T (0.05, 0.07, 0.12)	Baes 1984. US DOe 2004. APRIFEL 2008
Fruiting vegetables	T (0.04, 0.07, 0.10)	
Fruits	T (0.10, 0.15, 0.18)	
Root-vegetables	T (0.12, 0.20, 0.22)	
Soil to plant transfer coefficients (kg dry matter / kg dry matter)		
Leafy vegetables	LN (3.0 E-2, 4.6 E-2, 9.6 E-4, 2.8 E-1) ^e	Tremel-Schaub and Feix 2005, Zupan et al. 1995
Fruiting vegetables and fruits	LN (4.2 E-3, 3.1 E-3, 6.1 E-4, 1.9 E-2)	
Root-vegetables	LN (5.7 E-3, 7.3 E-3, 2.7 E-4, 4.5 E-2)	

802 ^a U (x1, x2) specifies a uniform distribution with minimum x1 and maximum x2.

803 ^b CUM stands for cumulative distribution.

804 ^c perc. stands for percentile.

805 ^d T (x1, x2, x3) specifies a triangular distribution with minimum x1, mode x2 and maximum x3.

806 ^e LN (x1, x2, x3, x4) specifies a lognormal distribution with mean x1, standard deviation x2, lower truncation limit x3 and upper truncation limit x4.

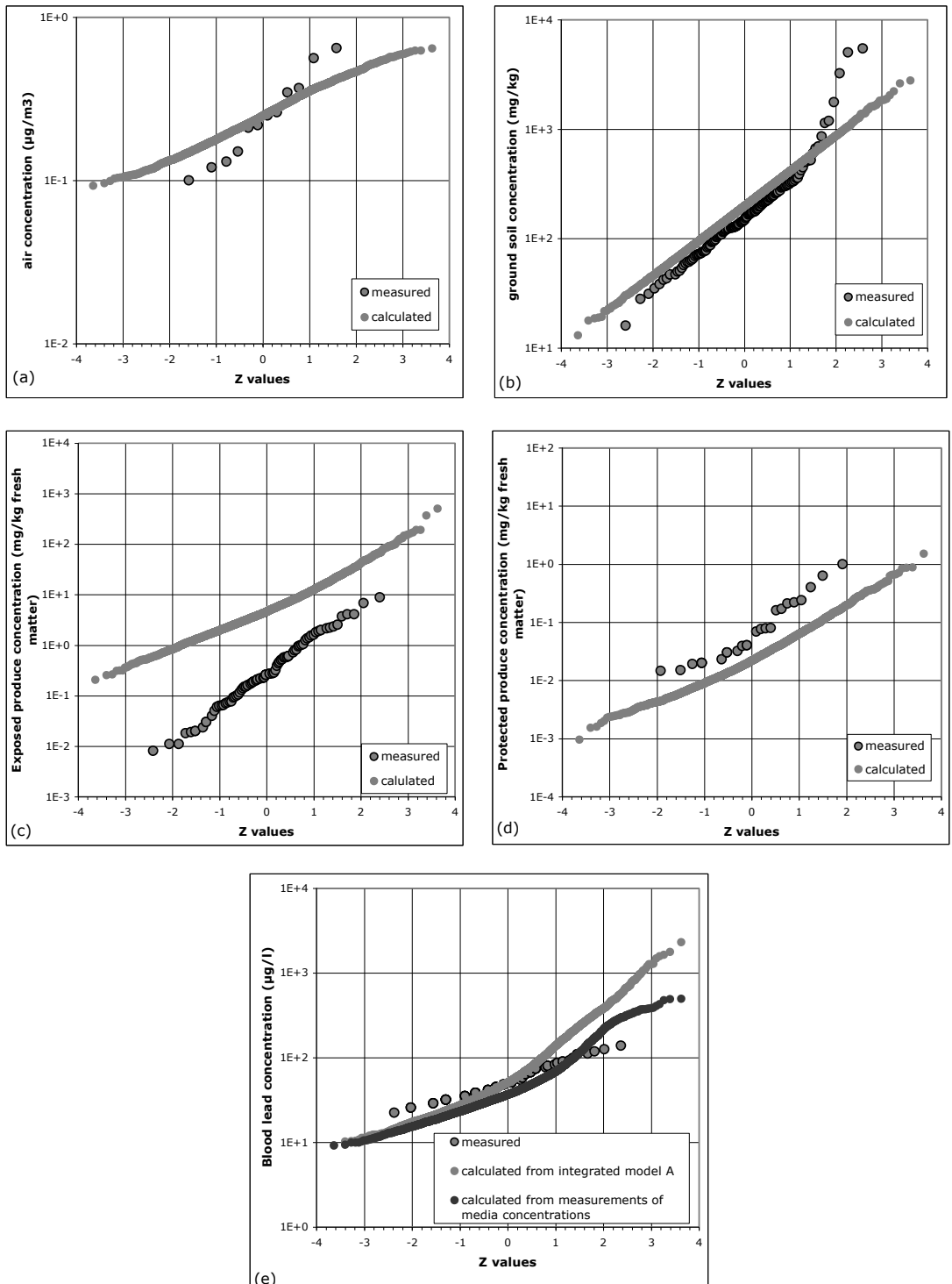
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809 Table 3: Input parameters with the highest sensitivity on exposure produce concentrations in
 810 model A

Parameters	Symbols	Rank correlation coefficients between the input parameter and exposure produce concentration
Particle-water partition coefficient	Kd	-0.44
Concentration in root-zone soil	Cs	0.41
Leaf wet density	rho_leaf	0.37
Primary production dry vegetation	veg_prod	0.31
Rainsplash	rainsplash	0.21
Leaf surface erosion half-life	Thalf_le	0.14
Stem wet density	rho_stem	-0.12
Annual average precipitation	rain	0.11
Ambient environmental temperature	Temp	-0.09
Land surface runoff	runoff	0.09

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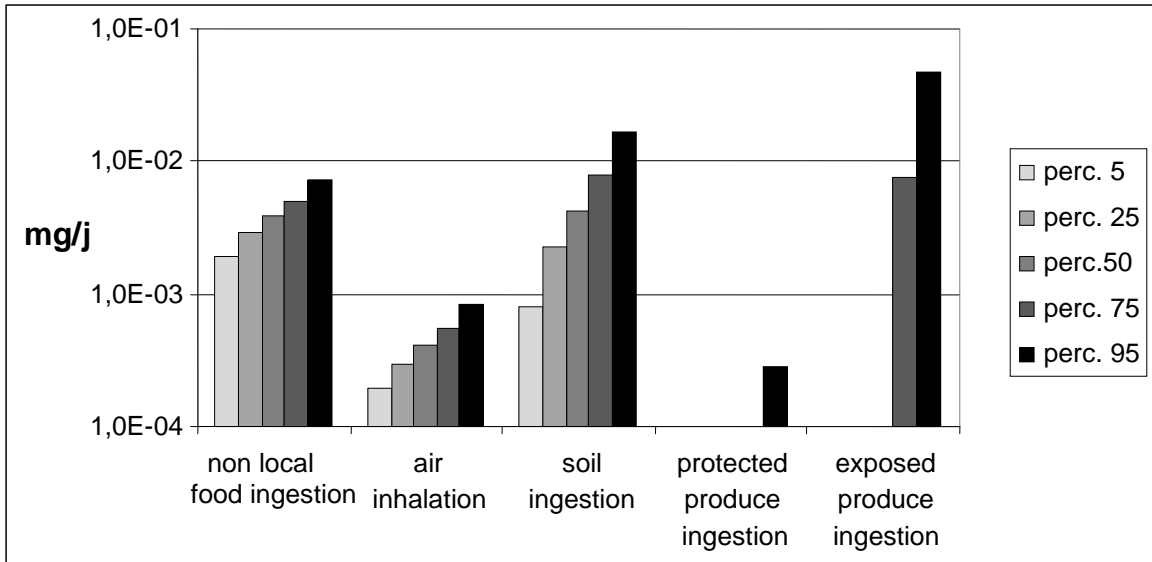
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814 Figure 1 : Distributions of the measured concentrations and of the concentrations calculated
 815 with model A in air (a), ground soil (b), exposed produce (c), protected produce (d) and blood
 816 lead (e) (for blood lead, concentrations were also predicted by IEUBK from measurements in
 817 the other media)

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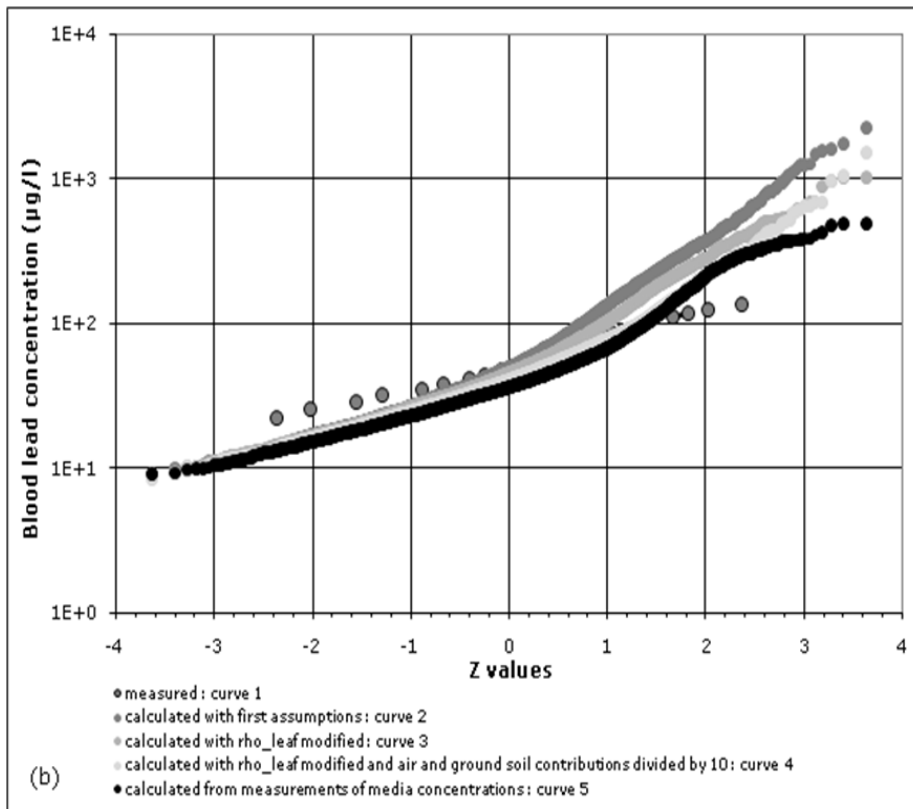
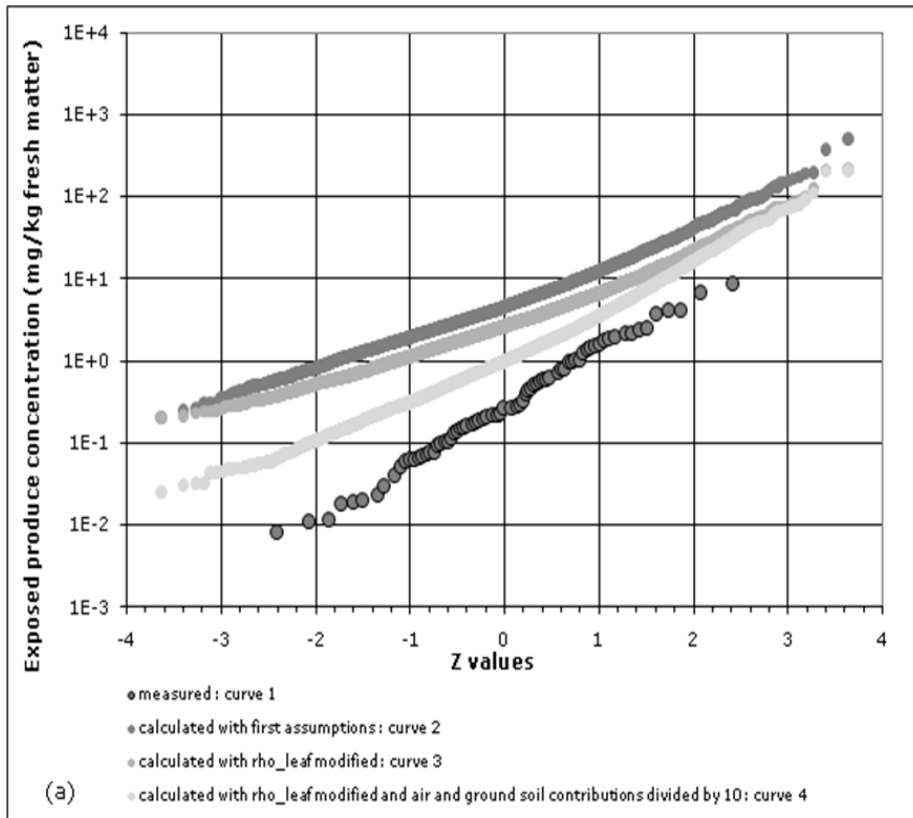


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821 Figure 2: Contributions of the exposure routes to the lead uptake according to model A

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Figure 3: Distributions of the measured concentrations and of the concentrations calculated in exposed produce (a) and blood lead (b) before and after correcting the value of leaf wet density in model A

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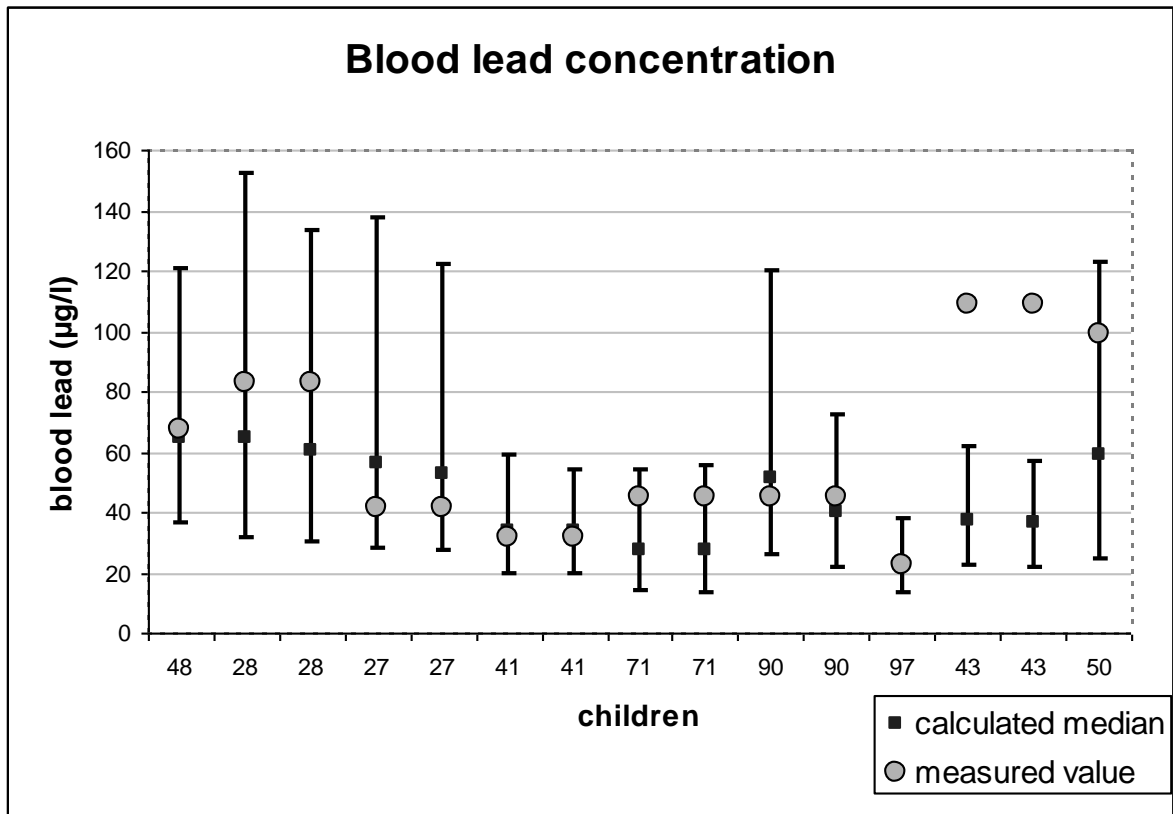


Figure 4: Comparison of the calculated and observed blood lead concentrations for the subsample of tested children (because of uncertainty in the location of some children's houses, there are two entries for some children based on two different assumptions for soil concentrations and atmospheric deposition around their houses) – the error bars show the range (from the 5th to the 95th percentile) of calculated concentrations with model B