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Gwenaëlle Lavison-Bompard, Julien Parinet, Karelle Huby, Thierry Guérin, Chanthadary Inthavong, et al.. Correlation between endemic chlordecone concentrations in three bovine tissues determined by isotopic dilution liquid chromatography–tandem mass spectrometry. *Science of the Total Environment*, 2021, 788, pp.147833. 10.1016/j.scitotenv.2021.147833 . anses-03722009

HAL Id: anses-03722009

<https://anses.hal.science/anses-03722009v1>

Submitted on 13 Jul 2022

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1 **Correlation between endemic chlordecone concentrations in three bovine tissues**
2 **determined by isotopic dilution liquid chromatography–tandem mass spectrometry**

3

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14

15 **Highlights**

- 16 • Occurrence and concentrations of chlordecone in fat, muscle and liver in 200 bovines.
- 17 • Chlordecone concentrations are correlated in fat, muscle and liver.
- 18 • Correlation factors: 0.54 muscle/fat, 3.75 liver/fat, and 0.14 muscle/liver.

19

20 **Abstract**

21 Chlordecone (CLD) is an organochlorine pesticide widely used from the 1970s to the 1990s in
22 the French West Indies that induced long-term pollution of the ecosystem. Due to involuntary
23 soil ingestion, some species bred in open-air areas can be contaminated. As CLD is distributed
24 in various tissues depending on the breeding species, this study focuses on the distribution of
25 CLD in bovines. For this purpose, three tissues, i.e. fat, muscle, and liver, from 200 bovines
26 originating from Martinique and Guadeloupe were sampled in 2016 to determine their endemic
27 contamination levels. Analyses were performed with the official method for veterinary controls,
28 isotopic dilution liquid chromatography–tandem mass spectrometry, which has been fully
29 validated and which reaches a limit of quantification of 3 $\mu\text{g}\cdot\text{kg}^{-1}$ fresh weight (fw).
30 Irrespective of the matrices, CLD was detected in 68% of samples (404 samples above the
31 LOD) and quantified in 59% of samples (332 samples above the LOQ). Regarding
32 contamination levels, the liver had a broader range of concentrations (LOQ up to 420.6 $\mu\text{g}\cdot\text{kg}^{-1}$
33 fw) than fat (LOQ up to 124.6 $\mu\text{g}\cdot\text{kg}^{-1}$ fw) and muscle (LOQ up to 67.6 $\mu\text{g}\cdot\text{kg}^{-1}$ fw). This
34 confirms the atypical behaviour of CLD compared to other persistent organochlorine pollutants.
35 Statistical processing demonstrated a correlation between CLD concentrations among the three
36 studied tissues. The CLD concentration ratios were 0.54 for muscle/fat, 3.75 for liver/fat, and
37 0.14 for muscle/liver.

38

39 **Keywords:** Chlordecone, ID-LC-MS/MS, bovine tissues, correlation factors

40 1. Introduction

41 Chlordecone (CLD) is an organochlorine pesticide that was widely used in the French West
42 Indies (Martinique and Guadeloupe) to protect banana plantations from black weevil
43 (*cosmopolites sordidus*) between 1972 and 1993. CLD is a bi-cyclic chlorinated molecule. As
44 its octanol/water coefficient exceeds 3, CLD is classified as a lipophilic pollutant. CLD also
45 exhibits a ketone function (C=O) and the presence of a non-saturated carbon-carbon bond that
46 can lead to keto-enol equilibrium.

47 CLD was classified in 2009 by the Stockholm Convention as a persistent pollutant. It is known
48 to be carcinogenic, mutagenic and reprotoxic, and is suspected of being an endocrine disrupter
49 (Multigner et al., 2010). In a cohort of a study carried out in Guadeloupe (Timoun cohort), an
50 effect of CLD exposure was found on the duration of pregnancy (Hervé et al., 2016), and on
51 the thyroid hormone system (Cordier et al., 2015), as well as impairment of fine motor function
52 in boys (Boucher et al., 2013; Dallaire et al., 2012), and visual contrast sensitivity (Saint-
53 Amour et al., 2020). For male adults, an increase in the risk of prostate cancer has also been
54 documented (Multigner et al., 2010).

55 Due to the former use of this pesticide, certain Martinican and Guadeloupean environmental
56 compartments are widely contaminated with CLD. Soil contamination, linked to former use of
57 CLD, depends on soil composition (Cabidoche et al., 2009; Woignier et al., 2013). Studies on
58 the fate of CLD have demonstrated low and poor degradation in soils with decontamination
59 projected to take decades to centuries (Cabidoche et al., 2009). More recent studies have also
60 indicated that transformation of CLD occurs in soils through highly complex pathways (Benoit
61 et al., 2017; Chevallier et al., 2019; Clostre et al., 2015). Food crops (including vegetables)
62 through root uptake (Clostre et al., 2015; Florence et al., 2015; Létondor et al., 2015), and
63 ruminants, through involuntary soil ingestion, can also be contaminated by CLD (Clostre et al.,
64 2014; Collas et al., 2019; Fournier et al., 2017).

65 Regarding food safety, in 2007, the French Food Safety Agency re-examined toxicological
66 reference values based on previous studies in rats dating back to 2003. No changes were made:
67 the chronic health-based guidance value was established at 0.5 µg/kg body weight/day, and the
68 acute value at 10 µg/kg body weight/day. The impact of food consumption, including local food
69 supply habits, was also examined based on consumption data (AFSSA, 2007): it was
70 determined that setting maximum concentration limits in foodstuffs would be of limited
71 effectiveness considering subsistence production (Dubuisson et al., 2007). Moreover, it was
72 concluded that the maximum residue levels (MRLs) established by the European Regulation on
73 MRLs of pesticides in and on food and feed of plant and animal origin (Commission Regulation
74 (EU) No 212/2013, 212AD ; Commission Regulation (EC) No 839/2008) , provided sufficient
75 protection of the population regarding CLD exposure via food of animal origin (ANSES, 2017).
76 Concerning livestock, the target tissue for official controls is perirenal fat, since it is the tissue
77 in which non-polar organochlorine pesticides are supposed to be mostly distributed. However,
78 some studies have shown that CLD has atypical behaviour regarding its tissue distribution, and
79 found that CLD is not only distributed in fat but also in muscle in certain species, such as poultry
80 (Jondreville et al., 2014a), ducks (Jondreville et al., 2014b), growing male Alpine kids (Lastel
81 et al., 2018), ewes (Saint-Hilaire et al., 2018) and pigs (Fourcot et al., 2020). This raises the
82 question of distribution in other species such as bovines, which are largely consumed and are
83 one of the most targeted species for official food controls.

84 CLD concentrations can be determined either by gas or liquid chromatography; methods are
85 mostly adapted from Bordet et al. (Bordet et al., 2007). For gas chromatography, detection can
86 be performed with an electron capture detector (GC/ECD) or mass spectrometry, with low or
87 high resolution (Clostre et al., 2014; Florence et al., 2015; Fournier et al., 2017; Lastel et al.,
88 2018; Méndez-Fernandez et al., 2018). This second type of detection is recommended in the
89 case of positive results (i.e. > the limit of quantification - LOQ) for confirmation purposes.

90 Although gas chromatography is still used, most of the current methods, including the official
91 method for controls in France, are based on isotopic dilution followed by reversed-phase liquid
92 chromatography analysis and tandem mass spectrometry detection with negative electrospray
93 ionization (Clostre et al., 2015; Lastel et al., 2018; Saint-Hilaire et al., 2018). Isotopic dilution
94 is recommended to ensure accurate identification and quantification. Recent publications also
95 report the use of high-resolution mass spectrometry with LC separation for the identification of
96 CLD transformation products in the environment through biodegradation or food processing
97 (Martin et al., 2020; Ollivier et al., 2020).

98 The present study aims at determining CLD levels in fat, muscle and liver tissues of 200
99 endemically contaminated bovines originating primarily from the most contaminated areas of
100 Martinique and Guadeloupe. The analytical method used was the official method for monitoring
101 controls in France, and is based on isotopic dilution liquid chromatography hyphenated with
102 mass spectrometry (ID-LC-MS/MS). These data were then used to assess the correlations
103 between CLD concentrations in fat, muscle and liver.

104

105 **2. Material and methods**

106 2.1. Chemicals and reagents

107 All solutions were prepared with analytical reagent grade chemicals and ultrapure water (18.2
108 M Ω .cm) obtained by purifying distilled water with a Milli-Q system (Merck Millipore, Saint-
109 Quentin-en-Yvelines, France). All solvents (acetone, acetonitrile, dichloromethane, hexane,
110 and methanol) were HPLC grade and were purchased from Fisher Scientific (Illkirch, France).
111 Sodium hydroxide and formic acid (98%) were also purchased from Fisher Scientific (Illkirch,
112 France). CLD (98%) and CLD ¹³C₁₀ (98%) standards were purchased from Cluzeau Info Labo
113 (Sainte-Foy-La-Grande, France). Working solutions were prepared by dilution of the
114 commercial standards in methanol before use, to prepare calibration standards and fortify the
115 samples for quality controls and method validation. All solutions were stored at 5 ± 3°C.

116

117 2.2. Equipment

118 LC-MS/MS measurements were performed using a 1200 series HPLC binary pump system
119 (Agilent Technologies, Courtaboeuf, France) coupled with an API 5500 QTRAP hybrid tandem
120 mass spectrometer (AB Sciex, Les Ulis, France). An Aqua C18 column (150 mm x 2.0 mm i.d.,
121 3 µm particle size) equipped with an Aqua C18 guard column (4.0 mm x 3.0 mm i.d., 3 µm
122 particle size) was used for chromatographic separation. An Eppendorf centrifuge 5810 (Sigma
123 Aldrich, Saint-Quentin-Fallavier, France), a refrigerated Hettich 32 R centrifuge (Sigma
124 Aldrich, Saint-Quentin-Fallavier, France), and a Genie 2 vortex (Scientific Industries, Bohemia,
125 NY, United States) were used for the extraction. A Grindomix GM 200 (Retsch Industries,
126 Haan, Germany) and a Polytron PT 3100 (Fisher Scientific, Illkirch, France) were used for
127 sample preparation. An IKA Ultraturrax T25 digital Disperser (Retsch Industries) was used for
128 both sample preparation and extraction. An XS 204 balance (Mettler-Toledo, Viroflay, France)
129 was used to weigh the samples. A water bath heater from Fisher (Fisher Scientific, Illkirch,
130 France) was used for fat melting. A Reacti-Therm Pierce heating module (ThermoFisher, Les
131 Ulis France) was used for solvent evaporation.

132

133 2.3. Sampling

134 From January to October 2016, an oriented exploratory survey plan was implemented by the
135 French authorities in charge of food control (Directorate for food, agriculture and forestry of
136 Guadeloupe and Martinique) toward the most probable contaminated bovines in the French
137 West Indies. In all, 200 bovines were selected, mostly in Martinique (120 bovines). In
138 Guadeloupe, open-air farms located in the most contaminated areas were chosen, whereas in
139 Martinique, animals were selected as a priority when the livestock was found to have positive
140 results in previous official control plans. Livestock number, age, and sex of the animals were

141 reported, as well as slaughterhouse locations (18 locations for Martinique and a single location
142 for Guadeloupe). For each bovine, the three tissues – perirenal fat, skirt for muscle, and liver –
143 were sampled and clearly identified for traceability.

144 For quality controls and method validation, liver and muscle samples were purchased from a
145 local supermarket (Choisy-le-Roi, France) and perirenal fat was collected from a local butcher
146 (Saint-Maur-des-Fossés, France). These matrices were determined to be CLD-free.

147

148 2.4. Sample preparation and extraction

149 The analytical procedure employed in this study was adapted from (Bordet et al., 2007), with
150 minor changes regarding sample preparation and major changes regarding separation and
151 detection: GC-ECD was switched to LC-MS/MS². This is the official method to be
152 implemented in France for monitoring and control plans for food of animal origin by approved
153 laboratories.

154 2.4.1. Preparation and extraction procedure for fat

155 Frozen fat samples were cut into pieces, blended with a Grindomix apparatus, heated to a
156 maximum of 60 °C and filtered through a strainer to clean samples of residues. A total of 0.5 g
157 was precisely weighed in a test tube. Then, 75 µL of internal standard CLD-¹³C₁₀ at a
158 concentration of 0.5 ng·µL⁻¹ was added to melted fat. After 1 min vortex-stirring, the internal
159 standard was kept in contact for 1 h with the matrix. CLD was extracted from the melted fat
160 sample 3 times with 3 mL of a mix of acetonitrile/dichloromethane 75/25 (v/v). For each
161 extraction, the test tube was briefly shaken with a vortex homogenizer and then centrifuged for
162 20 min at 1,200 g at a temperature of -20 °C. The supernatants were collected into another test
163 tube and solvent was evaporated to dryness at a temperature not exceeding 40 °C under a gentle
164 stream of nitrogen. The dry extract was dissolved in 15 mL of a mixture of hexane/acetone
165 85/15 (v/v) and then transferred to a 50 mL centrifuge tube. 5 mL of 0.5 M aqueous sodium

166 hydroxide solution was added, and the tube was vortexed for 15 s. The lower aqueous phase
167 was collected in another centrifuge tube. This extraction step was repeated twice, and the three
168 aqueous phases were combined in the same tube, and then washed with 5 mL of hexane by
169 gentle manual shaking. The hexane phase was discarded after a 3 min centrifugation step at
170 750 g. Then, 5 mL of aqueous sulfuric acid solution at 60% were added to the aqueous extract,
171 and the tube was shaken for 15 s with a vortex homogenizer and gently left to cool at room
172 temperature. CLD was extracted 3 times with 5 mL of a mixture of hexane/acetone 85/15 (v/v)
173 by shaking for 15 s with a vortex homogenizer, then centrifuged for 3 min at 750 g, and the
174 upper organic phases were collected in a 15 mL centrifuge tube and washed with 2 mL of
175 ultrapure water. After final centrifugation for 3 min at 750 g, the organic phase was evaporated
176 to dryness at a temperature not exceeding 40 °C under a gentle stream of nitrogen. The dry
177 extract was dissolved in 1 mL of methanol and transferred to a vial for chromatographic
178 analysis.

179 *2.4.2. Preparation and extraction procedure for muscle and liver*

180 Due to the lower fat content of liver and muscle compared to perirenal fat, a different sample
181 preparation method was used. Samples were cut into pieces and homogenized first with a
182 Grindomix apparatus, then a Polytron and a T25 Ultra-Turrax apparatus. Then, 2 g of
183 homogenized sample were precisely weighed in a 50 mL centrifuge tube before the addition of
184 50 µL of internal standard CLD-¹³C₁₀ at a concentration of 0.6 ng.µL⁻¹. After 1 min vortex-
185 stirring, the internal standard was kept in contact for 1 h with the matrix. CLD was extracted
186 by 10 mL of a mixture of hexane/acetone 85/15 (v/v) by using a T25 Ultra-Turrax for 1 min at
187 10,000 rpm. The organic phase was collected in another 50 mL centrifuge tube. The extracts
188 then followed the same basic and acidic transformations as for the fat extracts.

189

190 2.5. LC-MS/MS analysis

191 A total amount of 5 μL of the individual final extract was injected into the chromatographic
192 system at room temperature (Phenomenex, Le Pecq, France). Ultrapure water containing 0.1%
193 (v/v) formic acid (mobile phase A) and methanol containing 0.1% (v/v) formic acid (mobile
194 phase B) were used for the gradient elution at a constant flow rate ($200 \mu\text{L min}^{-1}$). The gradient
195 started with 40% of phase B and increased to 100% within 6 min. Phase B was kept constant
196 for the next 8 min and was then set back to 60% in 2 min before a 5 min equilibrium time,
197 before the next injection.

198 Electrospray ionization was run in negative mode. The spray voltage was set at -4,500 V. The
199 curtain gas was set at 25 psi of nitrogen. Spray gas and auxiliary gas were set at 40 and 60 psi
200 of air, respectively. The collision gas was set at “Medium” value. The source temperature was
201 set at $600 \text{ }^\circ\text{C}$. The instrument was operated in multiple reaction-monitoring mode (MRM). CLD
202 and its isotopic standard were detected under the deprotonated hydrated form (Bichon et al.,
203 2015). The quantifying and qualifying transitions for CLD were $506.7 > 426.7$ and $506.7 >$
204 428.7 , respectively. For these two transitions, the decluttering potential was -160 V, entrance
205 potential -10 V, collision cell exit potential -15 V, and collision energy -25 eV. The transition
206 for CLD- ^{13}C was $516.7 > 435.7$ with the same parameters, except the collision energy of -28
207 eV. The transition for CLD- ^{13}C was $516.7 > 435.7$ with the same parameters, except the
208 collision cell exit potential -10 V, and the collision energy -30 eV. The dwell time for each
209 transition was set at 100 ms. Analyst 1.51 Software (AB Sciex) was used for system control
210 and data acquisition and processing.

211

212 2.6. Method validation and quality assurance/quality control

213 The method was validated as per standard NF V03-110: 2010 (AFNOR, 2010), based on
214 tolerance intervals and overall determination of the performance of the method through the
215 accuracy profile (Mermet and Granier, 2012).

216 The accuracy profile provides a graphical representation of the risk of error for each
217 concentration level. The validated range is defined as the domain where β -expectation limits lie
218 within the acceptance limits (λ). This method also evaluates the performance of the calibration
219 model, the accuracy, and the uncertainty. The acceptance limits (λ) were set to 70-120% in
220 accordance with European Union guidelines (SANTE/11945/ 2015, 2015). Probability β was
221 set to 80%, meaning that the risk of results lying outside the limits is below 20% on average.
222 Initially, the method was validated and implemented to cover two commodity groups mentioned
223 in the SANTE document: “Fat from food of animal origin” and “Meat (muscle) and seafood”.
224 For the second commodity group, as fishes were targeted for official controls, it was decided to
225 select barracuda, a fish consumed in the French West Indies. Since (i) sample preparation is
226 identical for fish and meat samples, and (ii) the analysis is based on isotopic dilution, the method
227 was considered to be validated for the whole commodity group. This was confirmed by the
228 conformity of routine quality control, implemented for the present study, including recoveries
229 on meat as detailed below. Later, the method was extended to liver tissues. Therefore, three
230 separate profiles were built: one for the “meat and seafood” commodity, one for liver, and one
231 covering fat regarding its specific preparation. Five series spiked at 4 levels (3 levels for liver)
232 from 3 to 500 $\mu\text{g.kg}^{-1}$ fw were analyzed, each on different days in duplicate. The performances
233 of the method are presented in [Table 1](#).
234 An overall limit of quantification (LOQ) (3 $\mu\text{g.kg}^{-1}$ fw) was experimentally tested and the range
235 of application was tested up to 500 $\mu\text{g.kg}^{-1}$ fw, with success. Compliance of the test
236 concentrations with performance requirements laid down in the SANTE document
237 ([SANTE/11945/ 2015, 2015](#)) was checked, and the accuracy profiles were validated. The LOQ
238 is consistent with previous studies (Lastel et al., 2018; Saint-Hilaire et al., 2018) and the
239 European regulation on MRLs of pesticides ([Commission Regulation \(EU\) No 212/2013](#),

240 [212AD ; Commission Regulation \(EC\) No 839/2008](#)), which sets the MRL at 100 $\mu\text{g.kg}^{-1}$ fw
241 for fat, muscle and liver.

242 The limit of detection (LOD) was set according to the calibration model used for quantification
243 with standard solutions and to the enrichment factor between the matrix and the extracts: it was
244 set at 2 $\mu\text{g.kg}^{-1}$ fw for fat, and 0.5 for $\mu\text{g.kg}^{-1}$ fw for muscle. For liver, the LOD was set at one
245 third of the LOQ, i.e. 1 $\mu\text{g.kg}^{-1}$ fw.

246 The repeatability coefficients of variation for all matrices were within the range 1.2–8.3%, all
247 concentration levels considered. The intermediate precision coefficients of variation ranged
248 from 2.5 to 9.9%. Both criteria met the requirements of the SANTE Guidelines ([SANTE/11945/](#)
249 [2015, 2015](#)), which recommend a maximum value of 20%.

250 The expanded uncertainty of the method was estimated taking into account the standard
251 deviation of the tolerance interval and an estimation of the bias. It was set at 20 %,
252 corresponding to the maximum estimated uncertainty for each matrix and tested concentrations.
253 For routine analysis, quality controls were included in each sample batch, with criteria
254 recommended by SANTE guidelines ([SANTE/11945/ 2015, 2015](#)). Reagent blanks were
255 injected after each sample to check the absence of cross-contamination, with a tolerance
256 corresponding to 30% of the LOQ. Bracketing calibration (6 levels) was performed, and
257 linearity was controlled by residual deviation below 20% from the calibration curve. In every
258 sample batch, recovery at a level corresponding to 1.5 x LOQ was checked to be in the 70–
259 120% range. Retention times (± 0.1 min between CLD and CLD ^{13}C retention times) and ion
260 ratio deviations ($\pm 30\%$) were systematically examined to ensure correct analyte identification
261 in case of a result above the LOQ. Results were validated only if all criteria were within the
262 acceptable limits.

263 Some of the analyses (73 out of 200 whole triplets) were subcontracted to the laboratory
264 Inovalys, Le Mans, France. This laboratory, belonging to the network approved for French

265 official controls, applied the same official method described above after in-house validation
266 and demonstration of equivalent performance. In both laboratories, analyses were performed
267 under the international standard ISO/IEC 17025:2005 (ISO/IEC 17025, 2005) by qualified
268 operators.

269

270 2.7. Statistical analysis

271 The correlation study of the CLD concentrations in the three tissues was performed in multiple
272 steps. In order not to skew the determination of the correlation factors, results below the LOQ
273 were not included in the statistical analysis. For dataset description, in the lower bound scenario,
274 zero and the LOD values replaced results below LOD and LOQ, respectively whereas in the
275 upper bound scenario, these results were replaced by LOD and LOQ, respectively. A Shapiro–
276 Wilk test was used to determine the normality of the concentration distributions for each of the
277 three studied tissues. Pearson tests (SPSS software, v 23.0 (IBM, New York, NY, United States)
278 were used to assess the correlations of CLD concentrations in the triplets (fat, muscle and liver).
279 The statistical significance was set to $p < 0.05$. Linear regressions, prediction models, and
280 graphical representations of the correlations were performed with Studio software, v 1.2 (R
281 Development Core Team, 2016).

282

283 3. Results and discussion

284 3.1. Occurrence of CLD content in bovine perirenal fat, muscle, and liver

285 The main statistical descriptors are presented in [Table 2](#), and [Figure 1](#) illustrates the
286 distribution of the determined concentrations. The whole dataset is available in the
287 Supplementary Information. The most contaminated triplets exhibit atypically high values
288 compared to the whole dataset: CLD concentrations in liver and muscle ($812 \mu\text{g.kg}^{-1}$ fw and
289 $152 \mu\text{g.kg}^{-1}$ fw) are almost two fold higher than for the penultimate triplet ($420.6 \mu\text{g.kg}^{-1}$ fw and

290 67.6 $\mu\text{g}\cdot\text{kg}^{-1}$ fw). This triplet was excluded from the dataset for description and statistical
291 analysis. Irrespective of the type of matrix, CLD was detected in 68% of samples (404 samples
292 above LOD) and quantified in 59% of samples (332 samples above LOQ). Considering the
293 three tissues, CLD was mostly quantified in liver (65%, 130 out of 199 samples), followed by
294 fat (53%, 106 out of 199 samples), and muscle (48%, 96 out of 199 samples). Regarding
295 contamination levels, liver covers a broader range of concentrations (LOQ up to 420.6 $\mu\text{g}\cdot\text{kg}^{-1}$
296 fw) than fat (LOQ up to 124.6 $\mu\text{g}\cdot\text{kg}^{-1}$ fw), and muscle (LOQ up to 67.6 $\mu\text{g}\cdot\text{kg}^{-1}$ fw), as
297 mentioned in Table 2.

298 For muscle and liver, a previous study regarding food exposure of Martinicans and
299 Guadeloupeans reported that CLD was detected in 19% of beef meat samples ($n=38$)
300 ([Dubuisson et al., 2007](#)). The samples were collected during a sampling plan that was not only
301 focussed on contaminated areas (without precision) and was organised between 2002 and 2004.
302 The type of tissue collected was not detailed. The observed concentration range started at 0.83
303 $\mu\text{g}\cdot\text{kg}^{-1}$ fw for the average lower-bound residue level, and at 1.16 $\mu\text{g}\cdot\text{kg}^{-1}$ fw for the average
304 upper-bound, and reached up to 2.88 $\mu\text{g}\cdot\text{kg}^{-1}$ fw for the highest values. The higher detection
305 occurrence (59% for muscle and 85% for liver) and contamination ranges in the present study
306 are difficult to compare since they might be linked to a bias in sample collection, since it is
307 oriented toward most-probable contaminated bovines in the present study.

308

309 Considering muscle tissue, the most consumed part, the results can be compared with those
310 reported in the Kannari study ([ANSES, 2017](#)), where meat samples were collected between
311 2009 and 2015 from different supply chains, including slaughterhouses, local markets,
312 subsistence production, and medium and large wholesalers. The Kannari study differentiates
313 contaminated and non-contaminated areas of Martinique and Guadeloupe. In slaughterhouses,
314 64% ($n=293$) of the samples were above the LOQ of 1–10 $\mu\text{g}\cdot\text{kg}^{-1}$ fw in contaminated areas.

315 These figures are consistent with those found in the present study oriented toward contaminated
316 areas: 48%, 96 samples out of 199 above the LOQ for muscle, and 65%, 130 samples out of
317 199 for liver. The median level calculated in the Kannari study ($11 \mu\text{g.kg}^{-1}$ fw in contaminated
318 areas) was higher than the median level of $4.1 \mu\text{g.kg}^{-1}$ fw found in the present study. Moreover,
319 considering the lower and upper bound hypotheses, the same trend is observed (mean
320 concentrations of 5.8 and $6.2 \mu\text{g.kg}^{-1}$ fw versus 0.6 and $4 \mu\text{g.kg}^{-1}$ fw in the Kannari study). The
321 differences regarding these figures may be due to the type of animals sampled (not detailed in
322 the Kannari study) and to the sampling strategy that included imported commodities for the
323 purpose of the Kannari plan, whereas only local production was considered in the present work.
324 Regarding other species, our results in endemically contaminated bovines are similar to those
325 reported in endemically contaminated pigs originating from Guadeloupe ([ANSES 2019](#)). In pig
326 skirt sample ($n=77$), CLD concentrations ranged from 10 to $56 \mu\text{g.kg}^{-1}$ fw with a median at 23
327 $\mu\text{g.kg}^{-1}$ fw. In three other muscles (longissimus dorsal muscle, semi-membranous muscle and
328 flank) collected on another set of 15 pigs, the concentration range was also similar (11 – 34
329 $\mu\text{g.kg}^{-1}$ fw and median within 18 – $27 \mu\text{g.kg}^{-1}$ fw). These figures document the potential endemic
330 background contamination range of non-artificially fed bovines and pigs bred in Martinique
331 and Guadeloupe. Another study measured CLD concentrations in fat, legs and liver of free-
332 range ducks bred in endemically contaminated areas of Martinique ([Jondreville et al., 2014b](#)).
333 Again, concentration ranges are consistent with those reported in the present study: 47 – $1,215$
334 $\mu\text{g.kg}^{-1}$ fw for liver, 9 – $278 \mu\text{g.kg}^{-1}$ fw for fat, and 6 – $145 \mu\text{g.kg}^{-1}$ fw for legs (without skin). In
335 duck tissues, livers were also more highly contaminated than fat and muscle.

336 The distribution of CLD in fat, muscle and liver has also been reported to document the
337 contamination and/or decontamination kinetics for male Alpine kids ([Lastel et al., 2018](#)),
338 Muscovy ducks ([Jondreville et al., 2014b](#)) and pigs ([ANSES 2019](#)). It was also documented for
339 poultry, where liver and fat were investigated ([Jondreville et al., 2014a](#)). The same structure of

340 contamination was observed: liver followed by fat and muscle. Since the above-mentioned
341 studies concerned artificially contaminated animals, the comparison of both CLD levels and
342 occurrence is not relevant.

343 The contamination of all studied tissues, both occurrence and concentration levels, documents
344 the atypical behaviour of CLD in different species, including bovines. It is distributed in
345 different tissues and does not accumulate mainly in fat. This raises the question of a potential
346 correlation factor between CLD concentrations in tissues.

347

348 3.2. Statistical correlation between tissues

349 For the purposes of the correlation study, only quantified results in the three tissues were taken
350 into account, thus limiting the dataset to 96 animals. As mentioned above, the most
351 contaminated triplet (concentrations in fat/muscle/liver: 172/152/812 $\mu\text{g.kg}^{-1}$ fw) was excluded
352 from data processing.

353 As mentioned in [Table 3](#), Pearson correlation coefficients were above 0.884, indicating a strong
354 correlation. Moreover, a significance test was performed: values were below 0.01, which
355 supports the strong correlation.

356 A linear relationship of the observed CLD concentrations was found for muscle/fat,
357 muscle/liver, and liver/fat. The intercept was forced to zero to avoid negative values at low
358 concentrations. The slope of the curves represents the CLD correlation factors between the
359 tissues ([Figure 2](#)). The CLD concentration ratios were estimated: 0.54 for muscle/fat, 3.75 for
360 liver/fat, and 0.14 for muscle/liver. The coefficients of correlation (R^2) of the model were 0.973,
361 0.901, and 0.906, respectively. To go further, prediction intervals were also determined, based
362 on the dispersion of the measurements ([ANSES 2018.](#)). Depending on a two standard deviations
363 confidence level, it was possible to determine a prediction range of CLD concentration in fat
364 (tissue considered for controls) from a target concentration in skirt or liver (as consumed parts).

365 The value targeted in skirt was set at $20 \mu\text{g.kg}^{-1}$ fw on the basis of national authorities'
366 recommendations. For the two standard deviation interval, the CLD concentration in fat ranged
367 from 27 to $47 \mu\text{g.kg}^{-1}$ fw: a CLD concentration of $27 \mu\text{g.kg}^{-1}$ fw in fat was associated at 97.5%
368 to a CLD concentration below the target value of $20 \mu\text{g.kg}^{-1}$ fw in skirt. For the three standard
369 deviation interval, the range was 32 to $42 \mu\text{g.kg}^{-1}$ fw: a CLD concentration of $32 \mu\text{g.kg}^{-1}$ fw in
370 fat was associated at 84% to a concentration of CLD below the target value of $20 \mu\text{g.kg}^{-1}$ fw in
371 skirt. For liver, it was not possible to determine a range of concentrations of CLD in fat
372 corresponding to $20 \mu\text{g.kg}^{-1}$ fw in liver for any of the two confidence levels (95 nor 68%). It
373 was only possible to conclude that a concentration of $20 \mu\text{g.kg}^{-1}$ fw in liver, corresponded to a
374 value of $5 \mu\text{g.kg}^{-1}$ fw in fat in average. These prediction interval were used to define a CLD
375 mitigation concentration of $27 \mu\text{g.kg}^{-1}$ fw in fat for official control plans ([Ministerial decree of](#)
376 [Mai the 23rd, 2019](#)), corresponding to a MRL of $20 \mu\text{g.kg}^{-1}$ fw in muscle and liver.

377 As no data on CLD distribution in fat/muscle/liver are reported in the literature for endemically
378 contaminated bovines, these results were compared to those obtained for other species, i.e.
379 ducks and pigs. The observed ratios of CLD concentrations in liver/fat and muscle/liver for
380 bovines match those reported by Jondreville et al. ([Jondreville et al., 2014b](#)) on fresh matter for
381 Muscovy ducks: 3.9 for liver/abdominal fat, 0.6 for muscle with skin/fat, and 0.5 for muscle
382 without skin/fat. The muscle/fat ratio was also modelled with an intercept not forced to zero for
383 endemically contaminated pigs from Guadeloupe (n=77 animals), where skirt muscle was
384 investigated, and another set of pigs (n=15 animals) where skirt muscle but also three other
385 types of muscles were considered (longissimus dorsi muscle, semi-membranous muscle, and
386 flank) ([ANSES 2019](#)). A muscle/fat ratio of 0.64 was determined, considering the two groups
387 of animals, and thus including muscle type variability. This ratio is similar to that modelled in
388 the present study, even though animals and muscle types are different.

389 Fourcot et al ([Fourcot et al., 2020](#)) modelled the CLD distribution in two breeds of growing
390 pigs (Large White and Creole pigs) contaminated with intravenous administration of CLD (n=7
391 for each breed). To allow comparison with our data, we calculated a muscle/fat ratio from the
392 published figures in Fourcot *et al.*: total amount of CLD estimated in muscle or fat (Muscle:
393 8.9 mg for Creole pigs and 12.5 mg for Large White pigs- Fat: 13.2 mg for Creole pigs and 10.0
394 for Large White pigs) is divided by muscle or fat percentage of the animals respectively
395 (Muscle: 43.3 % for Creole pigs and 54.8% for Large White pigs. Fat: 34.7% for Creole pigs
396 and 22.4% for Large White pigs). Muscle/fat ratios are similar between bovine (0.54), Creole
397 pigs (0.54) and Large White pigs (0.51).

398 The CLD tissue distribution in artificially fed growing kids was also assessed ([Lastel et al.,](#)
399 [2018](#)). The observed ratios (liver/fat: 31 during the contamination period, 28 during the
400 decontamination period – muscle (skirt) /fat: 0.5 during the contamination period and 0.6 during
401 the decontamination period) are very different from those estimated in the present bovine study:
402 the liver/fat ratio is 55 times higher and the skirt muscle/fat ratio is 7 times lower in goats. These
403 differences could be linked to the general tissue composition of the different studied species,
404 including the differences in lipid classes, the overall fat content of the whole animal, as
405 mentioned by Lastel *et.al.*, and the metabolic pathways of CLD, as mentioned above.

406

407 **4. Conclusion**

408 An in-house validated ID-LC-MS/MS method was used to analyze CLD in 200 samples of
409 bovine fat, liver and muscle (skirt) originating from contaminated areas of Martinique and
410 Guadeloupe. CLD was mostly quantified in liver (65%), followed by fat (53%), and muscle
411 (48%). These results confirmed that CLD can be distributed in the studied bovine tissues, as
412 previously reported for goats, poultry and ducks. Strong CLD concentration correlations were
413 found between these three tissues, with concentration ratios estimated at 0.54 for muscle/fat,

414 3.75 for liver/fat, and 0.14 for muscle/liver. These correlations were used to define a CLD
415 mitigation concentration of 27 $\mu\text{g}\cdot\text{kg}^{-1}$ fw in fat for official control plans ([Ministerial decree of](#)
416 [Mai the 23rd, 2019](#)), corresponding to an MRL of 20 $\mu\text{g}\cdot\text{kg}^{-1}$ fw in muscle and offal. Moreover,
417 those data might be the support for MRL revision at European level.
418 It would be interesting to include other types of muscle tissue in a similar study, to take into
419 account their different composition, including fat content. This could reinforce the correlation
420 factors to further support consumer protection through dietary exposure.

421

422

423 **Acknowledgments**

424 The authors wish to acknowledge the French Directorate General for Food of the French
425 Ministry for Food, Agriculture and Fisheries for their financial support and for organizing the
426 oriented exploratory survey plan. The authors also acknowledge the ANSES Risk Assessment
427 Department- Methodology and Survey Study Unit for its support for the statistical analysis. The
428 authors are also grateful to Inovalys (Le Mans, France) for its participation in the analyses

429

430 **List of figures and tables**

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432 **Figure 1:** CLD concentrations ($\mu\text{g}\cdot\text{kg}^{-1}$ fw) of positive results (\geq LOQ) in muscle (n=96), fat
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434 Boxes represent the concentrations between percentiles 25 and 75. The line dividing the box
435 represents the median concentration.

436 The whiskers below and above the box represent percentiles 10 and 90.

437 **Figure 2:** Linear model and confidence intervals on means and observances for muscle/fat and
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439

440 **Table 1:** Analytical performances of the method

441 **Table 2:** Main descriptors of the dataset (n=199 animals)

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443

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445 **Funding sources**

446 This work is funded by the French Directorate for food of the French Agriculture ministry

447 (DGAL)

448

449 **Conflicts of interest**

450 The authors declare no conflict of interest.

451

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Table 1: Analytical performances of the method

Matrix	CLD concentration levels ($\mu\text{g}\cdot\text{kg}^{-1}$ fw)	Accuracy				
		Mean recoveries (%)	Trueness (relative bias %)	Precision		
				Repeatability (%)	Intermediate precision (%)	
Perirenal fat	3	109	8.5	2.1	2.5	
	10	108	7.5	1.2	2.8	
	100	104	4.1	2.6	5.8	
	500	99	-1.4	2.5	8.1	
Meat and seafood	Liver	3	99	-1.1	4.6	6.3
		20	102	2.5	4.3	5.1
		500	106	5.9	3.5	4.2
Other commodities	3	100	0.4	4.3	9.1	
	10	101	1.1	1.7	8.3	
	20	99	-0.7	3.3	9.9	
	500	92	-7.9	3.6	7.7	

Table 2: Main descriptors of the dataset (n=199 animals)

	Number of detections ^a	Number of quantifications ^b	Concentration ($\mu\text{g}\cdot\text{kg}^{-1}$ fw)				
			Maximum	Lower bound hypothesis		Upper bound hypothesis	
				Mean	Median	Mean	Median
Fat	117	106	124.6	11.4	4.2	12.2	4.2
Muscle	117	96	67.6	5.8	0.5	6.2	3.0
Liver	170	130	420.6	40.0	19.0	40.5	19.0

a:>LOD

b: >LOQ

Table 3: Pearson correlation coefficients

	Fat	Liver
Fat	-	0.884
Muscle	0.974	0.871

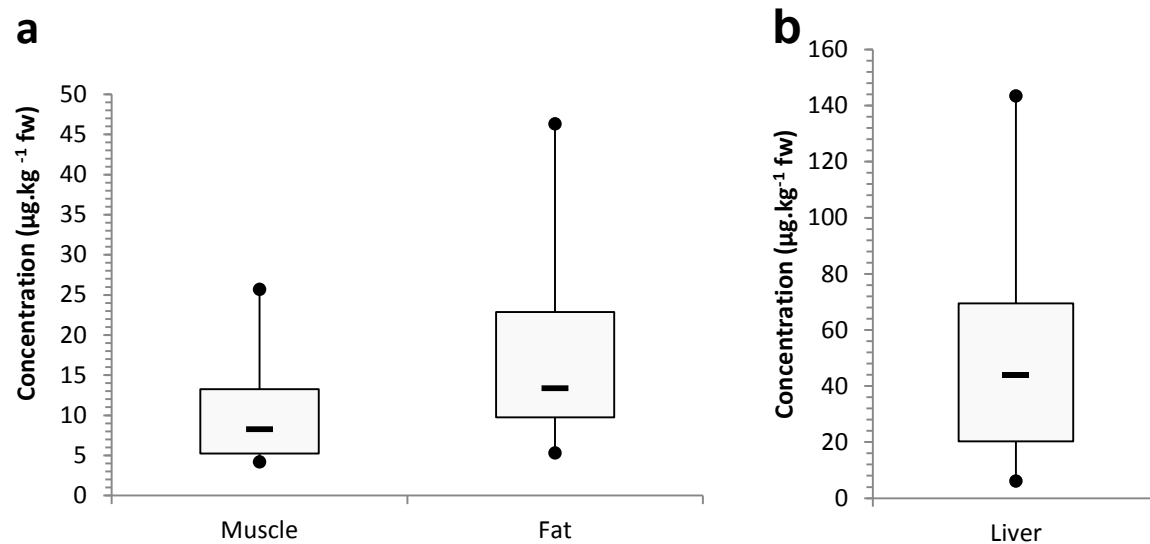


Figure 1: CLD concentrations ($\mu\text{g.kg}^{-1}\text{fw}$) of positive results ($\geq \text{LOQ}$) in (a) muscle ($n=96$) and fat ($n=106$), and (b) in liver ($n=130$) for the studied bovines. Boxes represent the concentrations between percentiles 25 and 75. The line dividing the box represents the median concentration. The whiskers below and above the box represent percentiles 10 and 90.

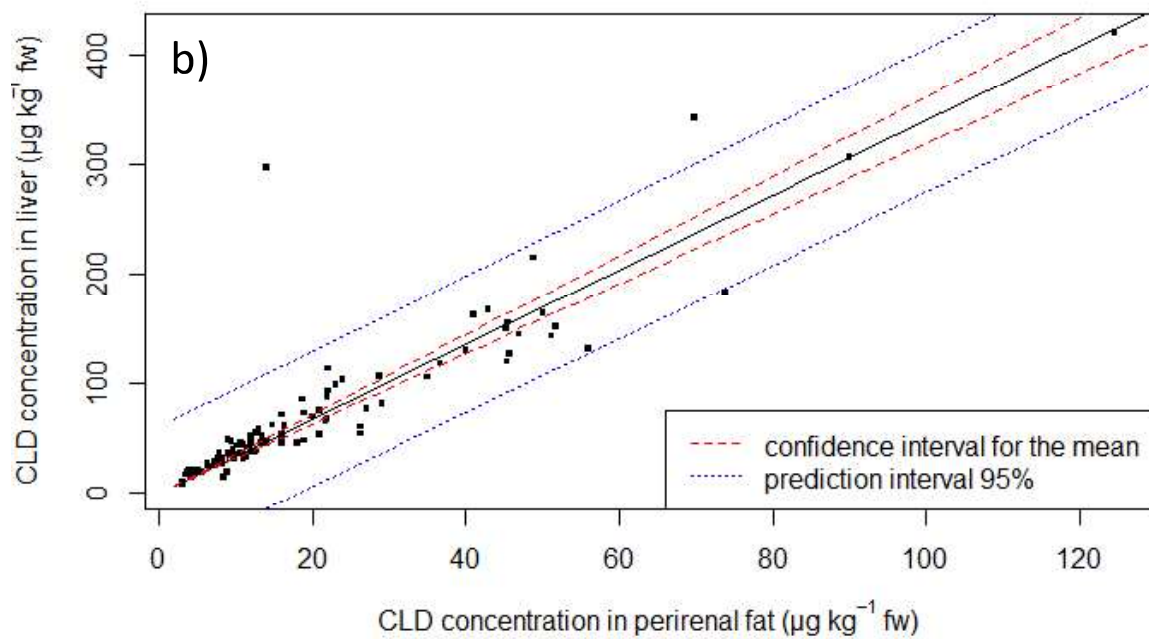
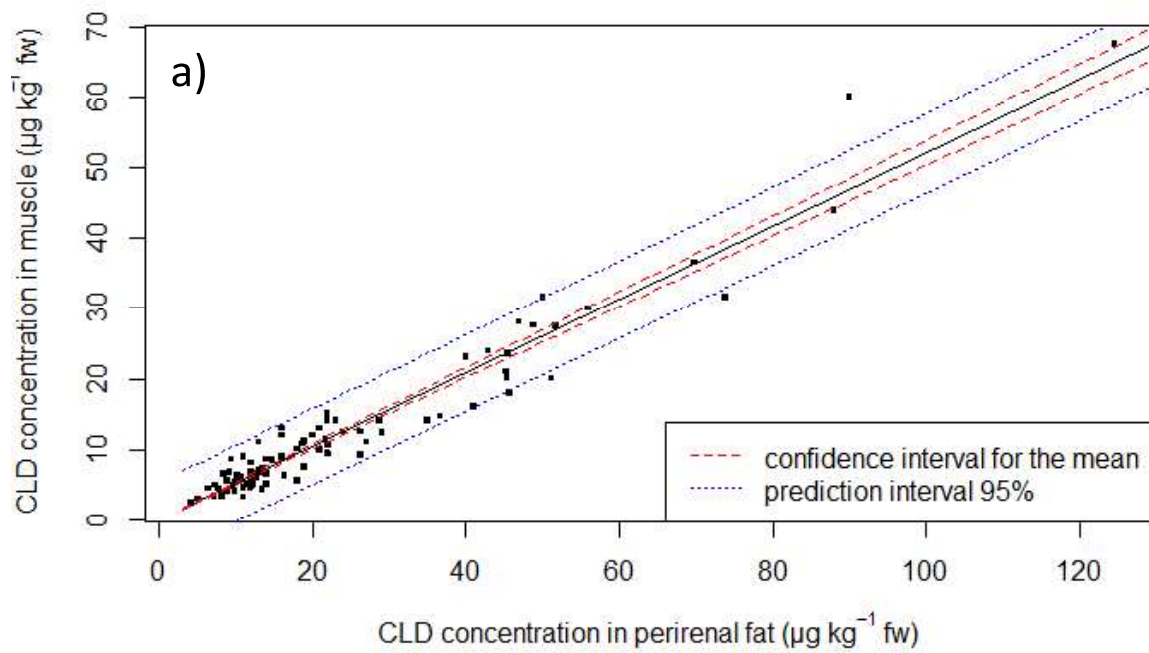
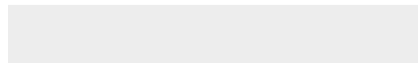
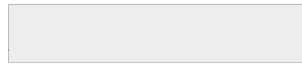


Figure 2: Linear model and confidence intervals on means and observances for (a) muscle/fat and (b) liver/fat correlations of CLD concentrations. Concentrations are expressed on fresh weight (fw).



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All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding (Internal public funding: French Agency for Food, Environmental and Occupational Health & Safety (ANSES), grant number CRD-2018-174).

Acknowledgments: We would like to express our gratitude to the Directorate General for Food (DGAI - French Ministry of Agriculture and Food) for its facilitating role and without whom this work would have not been possible, and especially to Pauline Favre and Mathilde Palussière. The authors sincerely acknowledge all the Resources and Environment Laboratories (LER) at IFREMER and the coordination of the French programme for monitoring emerging toxins (EMERGTOX scheme) for the collection of samples and for organizing the sampling campaigns. We also would to express our gratitude to the REPHY-REPHYTOX scheme, and in particular, Maud Lemoine and Nadine Neaud Masson, for the information on hydrological and phytoplankton data discussed in this report. We extend special thanks to the Metal and Mineral Trace Elements (ET2M) unit at ANSES for providing gastropods and for preparing them. Finally, the authors would like to thank all the staff at the Marine Biotoxins and Pesticides unit (PBM) at ANSES for their contribution to this work, and especially for preparing the bivalve mollusc samples.