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## Assessment of dechlorane plus and related compounds in foodstuffs and estimates of daily intake from lebanese population

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24 **Abstract**

25 Dechlorane Related Compounds (DRCs), including Dechlorane Plus (*syn/anti*-DP or *syn/anti*-DDC-CO)  
26 and related compounds (Dec-601 or DDC-ID, Dec-602 or DDC-DBF, Dec-603 or DDC-Ant and  
27 Chlordene Plus or DDC-PDD), are a group of polychlorinated flame retardants of concern since they  
28 were first reported in various environmental and biota matrices about one decade ago. In this work,  
29 we investigated the dietary intake of the Lebanese population to these lipophilic environmental  
30 contaminants upon the evaluation of selected foodstuff contamination. Collected food samples  
31 ( $n=58$ ) were selected to be representative of the lipid fraction of the whole diet of the Beirut  
32 population. The samples were analysed using pressurized liquid extraction, silica multilayer column  
33 followed by gel permeation chromatography for purification and GC-EI-HRMS for separation and  
34 detection. Detection frequency of at least one compound among Dechlorane Plus (*syn*-DP and *anti*-  
35 DP), Dechlorane 602, 603 and Chlordene Plus) was 91%. The mean concentrations of  $\sum_6$ DRCs, by food  
36 group, ranged from 4.7 to 29.5  $\text{pg g}^{-1}$  wet weight in lowerbound (LB) and from 6.7 to 76.9  $\text{pg g}^{-1}$  wet  
37 weight in upperbound (UB). Based on food habits, the dietary intake of Beirut adults was estimated  
38 to be between 3.71 (LB) and 5.61 (UB)  $\text{ng day}^{-1}$ . Dechlorane Plus and Dechlorane 602 were the  
39 dominant compounds, contributing to 70 and 24% of the total intake (LB value), respectively. This  
40 study reports for the first time the occurrence of Dechloranes in Lebanese foods and proposes  
41 corresponding deterministic dietary exposure scenario.

42

43 **1. Introduction**

44 Flame retardants (FRs) are chemical substances that are incorporated in flammable products like  
45 electronics, textiles and plastics, to inhibit flammability and increase fire safety. Dechlorane Plus (CAS  
46 13560-89-9,  $C_{18}H_{12}Cl_{12}$ ), or DDC-CO according to the accepted practical abbreviation (PRAB) (Bergman  
47 et al., 2012) (Table S1), a highly chlorinated FR additive, has been manufactured for more than 40  
48 years and used in electrical wire and cable coatings, computer connectors, and plastic roofing  
49 materials (Wang et al., 2016). DDC-CO has been used as a substitute to Mirex (also known as  
50 Dechlorane) (Fang et al., 2014) and has been suggested as a potential substitute to  
51 decabromodiphenyl ether in some applications (Zhou et al., 2017). The United States Environmental  
52 Protection Agency has classified DDC-CO as a high-production volume chemical (US EPA, 2019).  
53 OxyChem (Niagara Falls, USA) and Anpo Electrochemical Co. (Jiangsu, China) are the only two known  
54 DDC-CO manufacturers with an annual production of about 450–4500 tons in USA and 300–1000 tons  
55 in China (Von Eyken et al., 2016; Wang et al., 2016). The commercial DDC-CO mixtures are composed  
56 of two isomers, *syn*- and *anti*-DDC-CO, with an approximate ratio of 1:3 (Giulivo et al., 2017). DDC-CO  
57 shares similar properties to Persistent Organic Pollutants (POPs), including high lipophilicity,  
58 bioaccumulation potential, and potential for long- range transport, and has become ubiquitous in the  
59 environment (Na et al., 2017; Wang et al., 2017). The environmental occurrence of DDC-CO was first  
60 reported in air, fish and sediment samples from the Great Lakes area in 2006 (Hoh et al., 2006). Some  
61 studies showed that DDC-CO accumulates in wildlife and human tissue (Sühring et al., 2015; Wu et  
62 al., 2016). Tomy et al. (2007) demonstrated the biomagnification of DDC-CO along trophic levels via  
63 food chain.

64 In the wake of DDC-CO, other alternative Dechlorane Related Compounds (DRCs), based on  
65 hexachlorocyclopentadiene as well, such as Dechlorane 601 (CAS 13560-90-2,  $C_{20}H_{12}Cl_{12}$ , DDC-ID),  
66 Dechlorane 602 (CAS 31107-44-5,  $C_{14}H_4Cl_{12}O$ , DDC-DBF), Dechlorane 603 (CAS 13560-92-4,  $C_{17}H_8Cl_{12}$ ,  
67 DDC-Ant), and Chlordene Plus (CAS 13560-91-3,  $C_{15}H_6Cl_{12}$ , DDC-PDD), with PRAB according to  
68 Bergman et al. (2012) and Bergman and Rydén (2019) (Table S1), were commercialized to replace of

69 Mirex. These DRCs have subsequently been detected in various environmental and food samples as  
70 they are released into air, soil and water due to manufacturing and improper handling and disposal  
71 of DRC-containing products and materials (Sales et al., 2017; Abdel Malak et al., 2018).

72 Recent human biomonitoring studies reporting the presence of DRCs in plasma and human serum  
73 (Yan et al., 2012; Cequier et al., 2013; Brasseur et al., 2014; Chen et al., 2015; Fromme et al., 2015,  
74 Qiao et al., 2018), hair (Chen et al., 2015; Qiao et al., 2018), cord blood (Sales et al., 2017) and human  
75 milk (Siddique et al., 2012). Alike other chemical contaminants, human exposure to DRCs can be  
76 related to inhalation of indoor air, ingestion of dust and dietary intake (Kim et al., 2014).

77 Several studies have reported the occurrence of DRCs in various food and seafood matrices in Japan  
78 (Kakimoto et al., 2012; Yasutake et al., 2018), Korea (Kim et al., 2014), Belgium (L'Homme et al.,  
79 2015 ; Poma et al., 2018) and European markets (Aznar-Alemany et al., 2016). Estimated mean daily  
80 intakes were reported in the order of magnitude of  $\text{ng day}^{-1} \text{ person}^{-1}$  for Asian countries, considering  
81 at least most fatty matrices of animal origin and oils, where POP-like substances are expected.  
82 Reported exposure was much lower in Belgium, at the hundreds of  $\text{pg day}^{-1} \text{ person}^{-1}$  order of  
83 magnitude, but not all fatty food groups were considered. As such, dietary exposure data is missing  
84 worldwide to enable proper risk assessment.

85 In the present study, we investigated food consumption as a possible route of human exposure to  
86 DRCs in Lebanon. Various foods, including vegetable oil, meat, fish, egg, and dairy products, were  
87 analysed in order (i) to investigate the occurrence of DRCs in Lebanese foodstuffs and (ii) to estimate  
88 the dietary intake of the Lebanese adult population in a risk assessment context.

89

## 90 **2. Materials and methods**

### 91 *2.1. Sample collection and storage*

92 A total of 58 food samples, considered as representative of the average consumption of fatty food  
93 groups of animal and vegetable origins for the Lebanese population (Nasreddine et al., 2006), were  
94 collected in June 2017 from supermarkets located in Beirut (Lebanon). Nutritional categories

95 included meat ( $n=12$ ), fish ( $n=21$ ), egg ( $n=5$ ), milk and dairy product ( $n=13$ ), fat and oils added at  
96 table ( $n=7$ ). More precisely, collected samples were: chicken meat ( $n=6$ ), bovine meat ( $n=3$ ), bovine  
97 liver ( $n=3$ ), boops ( $n=3$ ), gilt head bream ( $n=2$ ), mackerel ( $n=2$ ), whiting ( $n=2$ ), anchovy ( $n=2$ ), grouper  
98 ( $n=5$ ), blackspot seabream ( $n=2$ ), European bass ( $n=2$ ), other fish ( $n=1$ ), egg ( $n=5$ ), milk ( $n=5$ ), labne  
99 ( $n=4$ ), yogurt ( $n=4$ ), olive oil ( $n=4$ ) and sesame oil ( $n=3$ ). The food samples were weighed,  
100 homogenized, lyophilized (except for oil), homogenized again and stored at  $-20\text{ }^{\circ}\text{C}$  until analysis.

101

## 102 2.2. Quantification of DRCs in samples

103 Trace analysis of targeted DRCs (*anti*-DDC-CO, *syn*-DDC-CO, DDC-ID, DDC-DBF, DDC-Ant and DDC-  
104 PDD) was performed according to previously published work (Abdel Malak et al., 2018), pending  
105 minor modifications in sample preparation. Briefly, lyophilized samples (approximately 3 g for fish or  
106 4 g for meat, egg and dairy products) were extracted by Pressurized Liquid Extraction (Büchi,  
107 SpeedExtractor, E-914), except for oil samples (1 g). The lipid content was determined  
108 gravimetrically. Purification of the extracts involved a multilayer silica gel column (acidic, neutral and  
109 basic layers) followed by Gel Permeation Chromatography. Purified extracts were analysed by gas  
110 chromatography (6890, HP, Palo Alto, CA, USA) on a HT8-PCB column (30 m  $\times$  0.25 mm, SGE  
111 Analytical Science, Ringwood, Australia) coupled to high resolution mass spectrometry (JMS 700D,  
112 Jeol, Tokyo, Japan), operating at a resolution of 10,000 according to 10% valley definition, in the  
113 electron impact ionisation mode and in a single sequence. Identification was based on two ion traces.  
114 Determination of DDC-ID, known to be challenging, was made possible by the successful  
115 chromatographic separation from *syn*-DDC-CO monitored with the same ion ( $[\text{C}_5\text{Cl}_6]^+$ ) in electron  
116 impact (Abdel Malak et al., 2018). Quantification was performed through isotopic dilution using  
117  $^{13}\text{C}_{10}$ -*syn*-DDC-CO,  $^{13}\text{C}_{10}$ -*anti*-DDC-CO and  $^{13}\text{C}_{10}$ -DDC-DBF as internal standards, and  $^{13}\text{C}_{12}$ -PCB-194 as  
118 recovery standard. Diagnostic ions are described Table S2.

119

## 120 2.3. QA/QC and reporting

121 Samples were analysed through four successive series of 10 to 20 samples. Recovery rates,  
122 determined for each sample, reached averages of  $74 \pm 14\%$ ,  $59 \pm 15\%$  and  $92 \pm 10\%$  for  
123  $^{13}\text{C}_{10}$ -*syn*-DDC-CO,  $^{13}\text{C}_{10}$ -*anti*-DDC-CO and  $^{13}\text{C}_{10}$ -DDC-DBF, respectively. Six procedural blanks (started  
124 from the extraction step) were introduced within the series to monitor procedural contamination. If  
125 procedural contamination was observed, a limit of reporting (LoR) was set at mean plus 3 times the  
126 standard deviation of the procedural blank. Otherwise, a limit of quantification (LOQ) was set for  
127 each sample based on a signal to noise ratio of 3:1 for the less intense signal. Trace amounts of  
128 *syn*-DDC-CO, *anti*-DDC-CO and DDC-DBF were always detected in procedural blanks, at values in the  
129 4.0-15.5, 2.0-6.8 and 0.9-6.4 pg ranges, leading to LoRs of 23.2, 9.4 and 9.1 pg per sample,  
130 respectively. For the other target compounds, LOQs were in the 1.2-14.5, 1.3-18.8 and 0.3-1.4 pg  
131 ranges per sample, respectively for DDC-ID, DDC-Ant and DDC-PDD.

132 An in-house fish oil (1 g aliquots) routinely used as quality control for ISO 17025 methods dedicated  
133 to a range of POPs (Jondreville et al., 2017) was fortified (4 ng of each native compound) and  
134 analysed as well ( $n=5$ ). Corresponding results regarding accuracy (intermediate precision and  
135 trueness uncertainty) were previously published and considered as satisfying (Abdel Malak et al.,  
136 2018).

137

#### 138 **2.4. Data processing**

139 Due to rather low DRC levels observed in this study, left-censored (lowerbound, LB) and right-  
140 censored (upperbound, UB) values were both calculated so that true values were under- and over-  
141 estimated (WHO, 2013). LB values corresponded to zero when lower than LOQ/LoR, observed value  
142 when higher than LOQ and observed value minus LoR when higher than LoR. UB values corresponded  
143 to LOQ/LoR when lower and to observed values when higher (Figure S1). LoRs being higher than  
144 LOQs, discrepancies between LB and UB values were more pronounced for *syn*-DDC-CO, *anti*-DDC-CO  
145 and DDC-DBF. The relationship between selected DRCs was statistically evaluated by Pearson  
146 coefficients of determination using Excel 2016 spreadsheets.

147

### 148 3. Results and discussion

149 The concentrations of *syn*-DDC-CO, *anti*-DDC-CO, DDC-ID, DDC-DBF, DDC-Ant and DDC-PDD in food  
150 groups based on wet weight (ww) and according to LB and UB scenarios are shown in Table 1.  
151 Detailed results, along with lipids content (lw), are also provided in Table S3 to facilitate the  
152 comparison with the literature.

153 Along the discussion, the results were compared to available values reported from Korea (Kim et al.,  
154 2014), Japan (Yasutake et al., 2018) and Belgium (L'Homme et al., 2015), bearing in mind that food  
155 groups do not necessary include the same food items.

156

#### 157 3.1. Concentrations of $\Sigma$ DDC-CO

158 Detection frequencies of *syn*- and *anti*-DDC-CO for all investigated foodstuffs were 40% and 86%,  
159 respectively. Depending on the food group considered, detection frequencies of the sum of *syn*-DDC-  
160 CO and *anti*-DDC-CO ( $\Sigma$ DDC-CO) ranged from 57 to 100%. The highest mean concentration of  $\Sigma$ DDC-  
161 CO was obtained in a meat sample, (161 pg g<sup>-1</sup> ww, UB), followed by an olive oil sample (136 pg g<sup>-1</sup>  
162 <sup>1</sup> ww, UB). However, the mean concentration of  $\Sigma$ DDC-CO obtained in the meat group was lower  
163 than in the vegetable oil group (Table 1).

164 Regarding fish, the mean concentration of  $\Sigma$ DDC-CO (5.0 to 8.0 pg g<sup>-1</sup> ww, [LB-UB]) was in the same  
165 order of magnitude to levels reported for fish samples collected in supermarkets in Osaka, Japan in  
166 June 2011 ( $n=20$ , average of 2.35 pg g<sup>-1</sup> ww) (Kakimoto et al., 2012) or in rivers in Latvia in October  
167 2012 ( $n=25$ , average of 11.34 pg g<sup>-1</sup> ww) (Rjabova et al., 2016). It was also lower than those obtained  
168 from fish and shellfish collected in March 2011 in Korea ( $n=70$ , average of 36.34 pg g<sup>-1</sup> ww) (Kim et  
169 al., 2014). No significant correlation between concentrations and lipid contents was observed.

170 Concerning vegetable oils, the mean concentration of  $\Sigma$ DDC-CO (21.1 to 52.8 pg g<sup>-1</sup> lw, [LB-UB]) was  
171 in the same order of magnitude as those reported in vegetable oils ( $n=2$ ) from Spain (average of



172 49.15 pg g<sup>-1</sup> lw) (Von Eyken et al., 2016) and in oils and fats ( $n=5$ ) from Japan (average of 18.5 pg g<sup>-1</sup> ww) (Yasutake et al., 2018).

174 In meat, the mean concentration of  $\Sigma$ DDC-CO (17.4 to 19.9 pg g<sup>-1</sup> ww, [LB-UB]) was lower than values  
175 reported from Korea ( $n=35$ , average values ranging from 9.8 to 169.8 pg g<sup>-1</sup> ww, according to meat  
176 type) (Kim et al., 2014) but higher than those obtained from Japan ( $n=5$ , average of 3.34 pg g<sup>-1</sup> ww)  
177 (Yasutake et al., 2018).

178 As regards eggs, the mean concentration of  $\Sigma$ DDC-CO (6.9 to 8.9 pg g<sup>-1</sup> ww, [LB-UB]) was in the same  
179 order of magnitude as those reported from Korea ( $n=5$ , average of 15.31 pg g<sup>-1</sup> ww) (Kim et al.,  
180 2014), but higher than those obtained from Belgium ( $n=8$ , average of 26.27 pg g<sup>-1</sup> lw) (L'Homme et  
181 al., 2015). Our values were also lower than those reported in herring gull eggs from North America  
182 (up to 15 ng g<sup>-1</sup> ww) (Gauthier et al., 2009). Authors suggested that the presence of DDC-CO in eggs  
183 could result from the accumulation of FRs in maternal specimen via their diet and be transferred  
184 during ovogenesis.

185 Finally, for milk and dairy products, the mean concentration of  $\Sigma$ DDC-CO (1.7 to 3.1 pg g<sup>-1</sup> ww,  
186 [LB-UB]) was lower than those reported from Korea (28.1 to 39.3 pg g<sup>-1</sup> ww) (Kim et al., 2014), but  
187 higher than those reported from Belgium (average of 17.6 pg g<sup>-1</sup> lw) (L'Homme et al., 2015).

188 These results tend to indicate that the DDC-CO contamination of foods from Lebanon is lower than  
189 those from Korea (Kim et al., 2014), in the same order of magnitude than those from Japan (Yasutake  
190 et al., 2018), and higher than those from Belgium (L'Homme et al., 2015).

191

### 192 3.2. Profiles of DDC-CO isomers

193 The commercial mixture of DDC-CO exhibits two stereoisomers, *anti*- and *syn*-DDC-CO, in a ratio of  
194 about 3:1 (Feo et al., 2012). The corresponding enantiomeric fraction of *anti*-DDC-CO ( $f_{anti}$ ), defined  
195 as the levels ratio of *anti*-DDC-CO over  $\Sigma$ DDC-CO, reaches 0.75. In biota, various authors reported  
196 values of  $f_{anti}$  at  $0.6 \pm 0.2$ , depending on the origin of the sample (Kang et al., 2010; Wang et al., 2011;  
197 Von Eyken et al., 2016), due to specific enrichment in *syn*-DDC-CO in the environment. In our study,

198 depending on the food item considered, the  $f_{anti}$  values, when quantified, ranged from 0.26 to 0.76  
199 (Figure 1), which were close to, or lower than, the values in the technical product, and similar to  
200 those reported in the literature (Kim et al., 2014; L'Homme et al., 2015; Von Eyken et al., 2016; Wang  
201 et al., 2016). The highest value of  $f_{anti}$  (0.76) was obtained from an olive oil sample, suggesting that, in  
202 this unique case of a food item of vegetable origin, the contamination could have occurred by direct  
203 contact with the technical product, with no biological enrichment observed. Inversely, the lowest  
204 values of  $f_{anti}$ , close to 25%, were found in dairy product samples, suggesting a significant enrichment  
205 in *syn*-DDC-CO through the producing mammal organisms, possibly involving lactation, and/or  
206 fermentation. For the fish samples, the  $f_{anti}$  ranged between 0.38 and 0.62 with a mean value equal  
207 to  $0.50 \pm 0.08$ , which appeared similar to that of the commercial product and those reported in fish  
208 samples in the literature (Kakimoto et al., 2012; Wang et al., 2015; Sühling et al. (2015)).

209

### 210 3.3. Concentration of other DRCs

211 DDC-DBF was detected in 41% of the sample set, at concentrations reaching up to  $97 \text{ pg g}^{-1} \text{ ww}$  (UB)  
212 (Table S3). Thus, DDC-DBF appeared as the second most important DRC after  $\Sigma$ DDC-CO, in terms of  
213 concentration, average concentrations being even higher than  $\Sigma$ DDC-CO in fish and milk and dairy  
214 products. The concentrations of DDC-DBF were in the same order of magnitude as those reported  
215 from Korea, lower than those obtained from Japan but higher than those reported from Belgium  
216 (Table S4).

217 DDC-Ant was detected in 28% of the sample set, at relatively low concentrations (similar to the  
218 results reported in foods from Korea, Japan and Belgium, Table S4). The highest values (7.7 and  
219  $8.1 \text{ pg g}^{-1} \text{ ww}$ , UB) were observed in sesame and olive oil samples.

220 Similarly, DDC-PDD was detected in 28% of all investigated foods at rather low concentrations  
221 ( $<2.9 \text{ pg g}^{-1} \text{ ww}$ ), except for one sesame oil and an egg samples at 22.5 and  $6.8 \text{ pg g}^{-1} \text{ ww}$  (UB),  
222 respectively. These concentrations were globally higher than those reported in food from Belgium

223 and Japan, but lower compared to those reported from China, which were in the  $\text{ng g}^{-1}$  lw range  
224 (Wang et al., 2015).

225 Finally, DDC-ID was not detected in any sample.

226

### 227 3.4. Correlation among Dechloranes

228 In order to investigate co-contaminations in contamination phenomenon, linear regression  
229 correlation analysis was performed between DP isomers and DDC-DBF only, due to the low detection  
230 rate of the other DRCs. Data analysis was achieved for  $\text{pg g}^{-1}$  ww (LB) concentrations, while  
231 accounting only for quantified ( $>\text{LoR}$ ) values. The correlation between *syn*-DDC-CO and *anti*-DDC-CO  
232 was moderate but significant ( $R^2=0.66$ ,  $P=0.0008$ ) whereas no correlation was observed between  
233 DDC-DBF and *syn*-DDC-CO ( $R^2=0.09$ ,  $P=0.76$ ) or *anti*-DDC-CO ( $R^2=0.05$ ,  $P=0.84$ ). Thus, although the  
234 contamination sources are considered the same, differential bioaccumulation and biotransformation  
235 properties of DDC-CO isomers among species and individuals appear as the main reason for  
236 moderate correlation. In addition, as expected, contaminations sources of DDC-CO and DDC-DBF  
237 appear to be independent.

238

### 239 3.5. Estimation of DRCs dietary intake

240 The Lebanese adult population (25-54 years old) was considered in the objective of proposing a first  
241 reference point for further studies in the geographical area. The occurrence levels of DRCs  
242 determined in the selected set of foodstuffs were used to perform a deterministic dietary exposure  
243 assessment, according to recognised guidelines (FAO-WHO, 2009; EFSA-FAO-WHO, 2011). The  
244 estimations of DRCs daily intake were calculated by the addition of quantities determined for each  
245 food item by multiplying the average concentration of DRCs ( $\text{pg g}^{-1}$  ww) in the considered food item  
246 (Table 1) with the daily dose of the considered food item ( $\text{g day}^{-1}$ ). Based on the study of food habits  
247 described by Nasreddine et al. (2006), the average total food intake per person and per day in  
248 Lebanon is 1881.6 g, including beverages except water. The collected food samples made up 387 g

249 per person per day in Lebanon (21% of the total food intake), covering all food from animal origin  
250 and vegetable oils, the expected major dietary intake contributors for such lipophilic environmental  
251 contaminants.

252 The mean dietary daily intake of DRCs per adult via selected Lebanese foods was estimated between  
253 3.71 and 5.62 ng day<sup>-1</sup>, according to LB and UB scenarios respectively, as shown in Table 2. It  
254 appeared lower than those reported by Kim et al. (2014) in Korea (11.7 ng day<sup>-1</sup>) and by Yasutake et  
255 al. (2018) in Japan (7.6 ng day<sup>-1</sup>), both based on representative fatty food groups, but higher than  
256 those reported by L'Homme et al. (2015) in Belgium (136 pg day<sup>-1</sup>), based on limited fatty food group.  
257 Considering substances profile (Figure S2), DDC-CO accounted for about 70% of  $\sum_6$ DRC in both  
258 scenarios. This contribution was lower than in Korea (92%, LB) and Belgium (88%, UB) but higher  
259 than in Japan (23%, LB). Indeed, DDC-DBF was the main DRC in Japan (75%, LB). In Lebanon, DDC-DBF  
260 accounted for about 24% of the profile in both scenarios, appearing as the second most important  
261 DRC like in Korea. With less than 7.2% of cumulated contributions, other DRCs (DDC-ID, DDC-Ant and  
262 DDC-PDD) were minor compounds.

263 As regards food groups (Figure S3), meat and poultry appeared as the main contributor of  $\sum_6$ DRC (43  
264 to 34%, [LB-UB]) as well as  $\sum$ DDC-CO (61 to 46%, [LB-UB]). They were followed by milk and dairy  
265 products as well as fat and oils added at table, each one contributing between 16 and 31% to the  
266 total, depending on the scenario. The high contribution of milk and dairy products to the exposure to  
267 DRCs is rather explained by the high consumption of these products rather than the relatively low  
268 occurrence levels. Conversely, relatively high occurrence levels in fat and oils added at table  
269 appeared as the main factor explaining the contribution to the exposure rather than the relatively  
270 low consumption level. Fish and egg represented the lowest contributors to the dietary daily intake,  
271 with less than 10% cumulated contribution.

272 In their food consumption survey, Nasreddine et al. (2006) also provided stratification data according  
273 to gender and age (3 groups of 25-34, 35-44 and 45-54 years old). Comparing genders, the average  
274 daily intake of  $\sum_6$ DRC in this study was higher for men (between 4.220 (LB) and 6.382 (UB) ng day<sup>-1</sup>)

275 than for women (between 3.257 (LB) and 4.911 (UB) ng day<sup>-1</sup>) (Table 3, Figure S3). The main reason is  
276 that men ingest more food than women (403.4 versus 371.9 g day<sup>-1</sup>) for all considered food groups  
277 except for milk and dairy products group (227.4 versus 257.1 g day<sup>-1</sup>, respectively). For similar  
278 reasons of decreasing food consumption with age, the estimated daily intake of  $\Sigma_6$ DRC has a  
279 downward trend as age increases.

280

#### 281 **4. Conclusions and perspectives**

282 A robust analytical method has been applied to a series of various Lebanese fatty foods of animal and  
283 vegetable origins ( $n=58$ ) in order to investigate the occurrence of DRCs and estimate associated daily  
284 intake. The detection frequency of at least one DRC in food items was 91%, supporting the fact that  
285 this compound family is ubiquitous. The mean concentrations of DRCs in each food item were rather  
286 low, in the pg g<sup>-1</sup> ww order of magnitude. However, the daily intake reached several ng per day and  
287 per person, with slight variations depending on gender and age. DDC-CO appeared as the main  
288 contributor, followed by DDC-DBF. To the best of our knowledge, this study is the first to analyse and  
289 detect emerging halogenated FRs in Lebanese foods and quantify the extent of human dietary  
290 exposure to them. Future work should focus on (i) total basket, including non-fatty food categories  
291 and on (ii) the indoor environment (dust, air) in order to be more accurate in the estimation of  
292 human exposure to these compounds. Toxicological reference values are also required to make  
293 conclusions as regards resulting risk to human health.

294

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300

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443

444 **Figure captions**

445 **Figure 1.** Quantity (raw data minus mean blank, pg per sample) of *anti*-DDC-CO versus *syn*-DDC-CO

446 when both values were above LoR, for all food categories. Brackets: number of plots out of

447 number of assayed samples.

448

449 **Table 1.** Concentration of DRCs obtained for various food groups from Lebanese market, reported according to lowerbound (LB) and upperbound (UB)  
 450 approaches, in  $\text{pg g}^{-1}$  ww. DF: detection frequency.

Food group		<i>syn</i> -DDC-CO	<i>anti</i> -DDC-CO	DDC-ID	DDC-DBF	DDC-Ant	DDC-PDD	$\Sigma$ DDC-CO	$\Sigma_6$ DRC
Meat and Poultry ( <i>n</i> =12)	Mean [LB-UB]	[10.1 - 11.8]	[7.4 - 8.1]	[0.0 - 0.2]	[0.0 - 0.7]	[0.1 - 0.3]	[0.0 - 0.05]	[17.4 - 19.9]	[17.5 - 21.1]
	DF (%)	25	83	0	0	17	0	83	92
Fish ( <i>n</i> =21)	Mean [LB-UB]	[2.0 - 4.2]	[3.0 - 3.9]	[0.0 - 0.3]	[7.0 - 7.8]	[0.2 - 0.3]	[0.4 - 0.5]	[5.0 - 8.0]	[12.6 - 16.8]
	DF (%)	57	95	0	62	33	62	100	100
Egg ( <i>n</i> =5)	Mean [LB-UB]	[1.7 - 3.1]	[5.2 - 5.8]	[0.0 - 0.3]	[1.2 - 1.7]	[0.2 - 0.4]	[1.36 - 1.41]	[6.9 - 8.9]	[9.6 - 12.7]
	DF (%)	40	80	0	20	20	20	100	100
Milk and Dairy Products ( <i>n</i> =13)	Mean [LB-UB]	[1.0 - 2.0]	[0.7 - 1.1]	[0.0 - 0.1]	[2.9 - 3.3]	[0.05 - 0.1]	[0.01 - 0.03]	[1.7 - 3.1]	[4.7 - 6.7]
	DF (%)	31	69	0	54	31	8	77	92
Fat and oils added at table ( <i>n</i> =7)	Mean [LB-UB]	[2.4 - 25.0]	[18.7 - 27.9]	[0.0 - 4.6]	[3.0 - 11.8]	[2.3 - 3.9]	[3.2 - 3.7]	[21.1 - 52.8]	[29.5 - 76.9]
	DF (%)	14	57	0	29	29	14	57	57

451

452

453 **Table 2.** Estimated mean daily intakes of  $\Sigma$ DDC-CO and  $\Sigma_6$ DRC [LB – UB] for the adult population in Beirut, in  $\mu\text{g day}^{-1}$ .

Food group	Mean of dietary intake, $\text{g day}^{-1}$	$\Sigma$ DDC-CO	$\Sigma_6$ DRC	Contribution to $\Sigma$ DDC-CO (%)	Contribution to $\Sigma_6$ DRC (%)	
Meat and Poultry (n=12)	91.7	[1597 - 1818]	[1607 - 1933]	[61% - 46%]	[43% - 34%]	
Fish (n=21)	19.7	[99.5 - 158]	[248 - 332]	[4% - 4%]	[7% - 6%]	
Egg (n=5)	12.1	[84 - 108]	[117 - 154]	[3% - 3%]	[3% - 3%]	
Milk and Dairy Products (n=13)	243.1	[418 - 760]	[1140 - 1622]	[16% - 19%]	[31% - 29%]	
Fat and oils added at table (n=7)	20.4	[430 - 1078]	[602 - 1569]	[16% - 27%]	[16% - 28%]	
Total	387.0	[2629 - 3922]	[3713 - 5609]	100%	100%	
Contribution (%)		[71% - 70%]	100%	100%	100%	
Food group	<i>syn</i> -DDC-CO	<i>anti</i> -DDC-CO	DDC-ID	DDC-DBF	DDC-Ant	DDC-PDD
Meat and Poultry (n=12)	[922 - 1079]	[675 - 739]	[0 - 22.7]	[0 - 61.4]	[9.6 - 26.3]	[0 - 3.9]
Fish (n=21)	[40.3 - 81.9]	[59.2 - 76.1]	[0 - 5.1]	[137 - 154]	[3.2 - 6.6]	[7.9 - 8.3]
Egg (n=5)	[20.9 - 38]	[63.1 - 70.0]	[0 - 3.4]	[14.2 - 20.9]	[1.9 - 4.4]	[16.5 - 17]
Milk and Dairy Products (n=13)	[253 - 496]	[165 - 264]	[0 - 27.9]	[709 - 804]	[11.1 - 23.8]	[1.6 - 6.7]
Fat and oils added at table (n=7)	[48.2 - 509]	[382 - 569]	[0 - 94.3]	[60.9 - 241]	[46 - 79.8]	[65.7 - 76]
Total	[1285 - 2204]	[1344 - 1718]	[0 - 153]	[921 - 1281]	[71.8 - 141]	[91.7 - 112]
Contribution (%)	[34% - 39%]	[36% - 31%]	[0% - 3%]	[25% - 23%]	[2% - 3%]	[2% - 2%]

454

455

456 **Table 3.** Estimated daily intakes of  $\Sigma_6\text{DRC}$  [LB – UB] for the adult population in Beirut according to  
457 gender and age, in  $\text{pg day}^{-1}$ .

Gender	Age (year)			
	25-34	35-44	45-54	25-54
Men	[4511 – 6749]	[4221 – 6420]	[3813 – 5829]	[4220 – 6382]
Women	[3381 – 5048]	[3278 – 4928]	[3066 – 4710]	[3257 – 4911]

458

- Eggs (n=2/5)
- × Fat and oils (n=1/7)
- ◆ Fish (12/21)
- Meat and poultry (n=3/12)
- ▲ Milk and dairy products (n=4/13)





