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## A national inventory of historical dioxin air emissions sources in France

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3           **A NATIONAL INVENTORY OF HISTORICAL**  
4           **DIOXIN AIR EMISSIONS SOURCES IN FRANCE**

5

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19

20          Keywords:

21          dioxins, emission, inventory, UNEP Toolkit, exposure assessment, endocrine disruptor

22

## 23 **Abstract**

24           Dioxins, are persistent organic pollutants with endocrine disrupting properties and are  
25 classified as human carcinogens. To better characterise historical atmospheric dioxin exposure  
26 in epidemiological studies, we performed a comprehensive inventory of dioxin-emitting  
27 sources between 1990 and 2008, in France, at a fine spatial resolution, according to the  
28 methodology of the United Nations Environment Program (UNEP) Toolkit.

29           Overall, we identified 2620 distinct dioxin-emitting sources over the French territory.  
30 The majority of the sources were geolocated either at the stack (43%) or at the building (27%).  
31 The final database contains the technical characteristics and activity rates for each source over  
32 the study period. These data served to estimate total annual dioxin emissions (g-TEQ/year).

33           We compared our estimates with both measurements and emissions inventories (at  
34 departmental and national level). The discrepancies in the comparisons between the Toolkit  
35 estimates, the dioxin measurements available, and inventories remained within the uncertainty  
36 range of the UNEP Toolkit estimates. The differences observed were due to a general lack of  
37 data and to the large number of small incinerators with large dioxin emissions inventoried. Due  
38 to better access to relevant data and a decrease in emissions, the UNEP Toolkit estimates get  
39 closer to measurements and estimates of others inventories for more recent years.

40           We created the first database containing the annual atmospheric dioxin emissions for a  
41 large number of emission sources at the facility scale for an entire country. This database was  
42 used to assess exposure to dioxins in epidemiological studies.

## 43        **1. Introduction**

44        Polychlorinated dibenzodioxins (PCDD) and dibenzofurans (PCDF), commonly referred  
45 to as ‘dioxins’, are toxic chemical pollutants, with endocrine properties, produced  
46 unintentionally by the combustion of chlorinated products in industrial processes, such as waste  
47 incineration, metallurgy, etc., as well as by accidental fire events (e.g. forest fires). The 2,3,7,8-  
48 TCDD, the most toxic dioxin congener, is classified as a group 1 carcinogen by the International  
49 Agency for Research on Cancer (IARC) with a causal relationship for all cancer types combined  
50 and a positive association for lung cancer, soft tissue sarcoma and non-Hodgkin lymphoma  
51 (Baan et al., 2009). **Due to the inconsistency of the association of the atmospheric dioxin**  
52 **exposure with others cancer such as breast cancer (Dai and Oyana, 2008; Fabre et al., 2008;**  
53 **Rodgers et al., 2018; Xu et al., 2016), further studies were warranted to investigate its effect on**  
54 **human health.**

55        The multiplicity of exposure sources and the latency between exposure and cancer  
56 occurrence represent major challenges in such studies and require an accurate characterisation  
57 of the spatial-temporal variability of exposures over large areas and long time periods. In  
58 numerous studies, the lack of past residential history and historical air pollutant exposure  
59 assessment at a fine spatial and temporal scale may have resulted in non-differential exposure  
60 misclassification and, hence, is likely to have contributed to imprecise risk estimates (Basagana  
61 et al., 2013; Brody et al., 2007; Cordioli et al., 2013).

62        As confirmed by the few available long-term measurements of atmospheric concentrations  
63 of dioxin (Augusto et al., 2015; Coutinho et al., 2015; Katsoyiannis et al., 2010), a strong  
64 decrease in industrial emissions over the last two decades has been observed in Western Europe,  
65 consequent to the implementation of specific abatement technologies. However, this overall  
66 decrease in industrial emissions varies significantly across countries and, most importantly, is  
67 dependent on the sector (waste incineration, metallurgy, etc.) (Fiedler, 2015). While numerous  
68 facilities, including metal industries and cement kilns are likely to emit dioxins (Coudon et al.,  
69 2018; Lohmann and Jones, 1998; Pacyna et al., 2001; Pronk et al., 2013), the majority of

70 published studies have restricted exposure assessment to incinerators (Ashworth et al., 2013;  
71 Cordioli et al., 2013; Fabre et al., 2008b). This stresses the need to consider the exposure from  
72 multiple sources in epidemiological studies.

73 While the spatial coverage of ambient air quality monitoring networks has steadily  
74 increased in recent years, monitoring data for dioxins are extremely sparse, making it  
75 impossible to rely on the monitoring station networks to assess human exposure to atmospheric  
76 dioxin emissions.

77

78 To characterise dioxin exposure of study subjects (Cordioli et al., 2013), epidemiological  
79 studies used exposure indicators based on geographic information systems or deterministic  
80 models such as Gaussian dispersion models (Coudon et al., 2018; Liu et al., 2015; Ranzi et al.,  
81 2011). All these exposure assessment methodologies rely on an inventory of dioxin emissions,  
82 and their evolution over time, performed at a fine spatial and temporal scale and covering large  
83 periods, to take into account the latency of cancer development (Ries et al., 2006). Several  
84 historical inventories of industrial and non-industrial dioxin emissions have been performed in  
85 Europe following the Stockholm Convention (Kok et al., 2005; Pacyna et al., 2003; Quass et  
86 al., 2004; Quina et al., 2011; Saral et al., 2014; Zhang et al., 2011). The aim of these studies  
87 was to inventory major dioxin-emitting sources, identify their temporal trends and assess the  
88 effectiveness of emission reduction policies at national or European level.

89 The inventories by Pacyna et al and Quass et al covered time periods of more than 10 years  
90 (1970-1995 and 1985-2005, respectively) (Pacyna et al., 2003; Quass et al., 2004). These were  
91 built with data reported from each country and dioxin releases were estimated with emission  
92 factors from the literature (or based on national reported emissions and official reported activity  
93 data). They were an important starting point for the assessment of dioxin emissions, and their  
94 environmental and health impacts. They present however three major limitations. Firstly, these  
95 results are generally shown with a coarse spatial resolution and are mostly based on values  
96 averaged over each single nation. Yet, in epidemiological studies, detailed emission estimates  
97 by source at a fine temporal and spatial scale are important. Secondly, despite the number of

98 sources covered by these inventories, some important local sources (like hospital waste  
99 incinerators) may be missing or are not fully characterised (Quass et al., 2004). Thirdly, the  
100 lack of detailed information implies large uncertainties in emission estimates, especially in the  
101 1990s (Webster and Connett, 1998). Even in more recent inventories, uncertainties over  
102 emissions were high, varying by about a factor of three (Pulles et al., 2006).

103 This study aims to build a comprehensive dioxin emission inventory, to include all relevant  
104 sources, for the French metropolitan territory over a long time period, i.e. between 1990 and  
105 2008, with a high spatial resolution, (i.e. location of dioxin-emitting facilities at the stack) and  
106 at a fine temporal scale (i.e. annual dioxin emissions) to be used in an epidemiological study  
107 on dioxin exposure and risk of cancer. The objective is to provide annual estimates of dioxin  
108 emissions, associated uncertainties using historical information on technical abatement  
109 measures and activity rate for each dioxin source. Unlike previous studies, measurements of  
110 source emissions will be used to assess the accuracy of our estimates.

## 111 **2. Materials and Methods**

### 112 **2.1. Identification of dioxin sources from 1990 to 2008**

113 The present inventory was based on the methodology proposed by the "Standardized  
114 Toolkit for the Identification of Quantification of Dioxin and Furan Releases" developed by the  
115 United Nations Environment Program (UNEP; <http://toolkit.pops.int/>). More particularly, our  
116 inventory used the update of the Toolkit, published in 2013 (UNEP, 2013).

117 Furthermore, the application of the methodology of the UNEP Toolkit to the French  
118 context was submitted and approved by a national group of experts from the French  
119 Environment and Energy Management Agency (ADEME); the Regional Directorate for the  
120 Environment, Planning and Housing (DREAL) of the Rhône-Alpes Region; and industry  
121 associations.

122 The UNEP Toolkit methodology suggests a five-step inventory (UNEP, 2013):

- 123 • Use of the UNEP Toolkit's list of activities likely to release PCDD/PCDF into the air to

- 124 identify the main group of sources in the country
- 125 • Production of an inventory of all PCDD/PCDF sources active during the study period
- 126 (1990-2008) and distribution of these source into the appropriate categories
- 127 • Collection of detailed information on technical process and activity characteristics of the
- 128 sources
- 129 • Identification of the suitable emission factor for each source, using the extensive UNEP list
- 130 of emission factors, according to the data collected
- 131 • Quantification of annual source emissions using emission factors

132

133 The UNEP Toolkit provides a comprehensive list of dioxin sources divided into ten groups:

- 134 • Group 1: Waste Incineration, including seven categories of waste incinerator;
- 135 • Group 2: Ferrous and Non-Ferrous Metal Production, including twelve categories for the
- 136 production of metals and metal alloys including recycling operations;
- 137 • Group 3: Heat and Power Generation, including five categories of large and small
- 138 installations using fossil fuels, biomass or gas;
- 139 • Group 4: Production of Mineral Products, including seven categories of processes to
- 140 manufacture mineral products;
- 141 • Group 5: Transport, including four categories (among which are road and ship transport);
- 142 • Group 6: Open Burning Processes, including two categories of burning biomass or waste
- 143 without technical equipment;
- 144 • Group 7: Production and Use of Chemicals and Consumer Goods, including eight
- 145 categories of various industrial activities;
- 146 • Group 8: Miscellaneous Sources, including an array of five source categories that do not
- 147 match the description of any other source group;
- 148 • Group 9: Disposal and Landfill, comprising five categories of waste disposal;
- 149 • Group 10: Contaminated Sites and Hotspots, including thirteen categories.

150

151        Since the study focused on air emissions, source groups 9 and 10, as well as some source  
152 categories of groups 7 and 8, were excluded due to the absence of air emission factors. Also,  
153 traffic emissions (group 5) were disregarded due to their weak contribution to local dioxin  
154 exposure (Coudon et al., 2018). Group 6, covering open burning processes, was omitted due to  
155 the impossibility of collecting consistent information to reliably characterise and locate  
156 emissions. Table 1 provides the detailed list of UNEP source groups and associated source  
157 categories inventoried in this study.

158        Relevant sources between 1990 and 2008 were identified through a combination of regional  
159 and national databases, in particular the Annual Emission Reporting on Pollutant and  
160 Greenhouse Gas Releases (GEREP), the French National Registry of Pollutant Emissions  
161 (IREP) and the Inventory of French Classified Facilities (ICPE). The databases are detailed in  
162 Table S1.

## 163        **2.2. Collection of technological characteristics**

164        The information collected for the characterisation of the facilities includes the following  
165 items: name of the facility; name of the operator; address; geographical coordinates; start and  
166 end dates of the activity; annual activity rate; and technical characteristics (including process,  
167 incoming material, smoke treatment system, physical parameters of flue gases, etc.).

168        **The information was mainly collected through the above-mentioned databases (Table S1)**  
169 **and complemented by these various sources: institutional information sources, contact with**  
170 **industrial unions, nationally recognized associations, company communications, municipal**  
171 **decrees, legislative decrees, measurement data, summaries of public meetings, etc.**  
172 Furthermore, a structured questionnaire was sent to the facilities operating company or group  
173 to collect additional information on their technical characteristics and activity rates.  
174 Technological changes impacting dioxin releases were registered in the database on an annual  
175 basis. **Each data collected was associated with a level of trust based on its origin. Measurement**



176 data, official databases, federations' data, various official decrees and questionnaire replies  
177 were considered as the most reliable sources of information. In addition, to the extent possible,  
178 we have crossed the information gathered to reduce uncertainties.

179

### 180 **2.3. Default air emission factors and emission estimates**

181 The Toolkit provides a comprehensive range of emission factors based on the international  
182 literature, experimental data, and governmental and institutional reports. Within each source  
183 category, the UNEP Toolkit distinguishes several classes and provides a default emission factor  
184 specific to each class. The default emission factors are expressed as TEQ (toxic equivalent) per  
185 unit of activity (e.g., µg-TEQ per ton of material produced, per ton of fuel burned, etc.). Based  
186 on the information collected on technology, process characteristics, and technical abatement  
187 measures and operating practices, each source is assigned to one of the several classes and  
188 corresponding default emission factor for this class.

189 Classification and assignment of the corresponding default emission factor, performed for  
190 each calendar year, were carried out independently by two engineers, with complementary  
191 expertise in chemistry and physics, blinded to each other's classification. Disagreement in  
192 classification was resolved by discussion to reach consensus. Furthermore, a national group of  
193 experts from ADEME, DREAL and industry associations has reviewed the classification with  
194 the UNEP toolkit of a subsample of facilities in the Rhône-Alpes Region.

195 The estimate of the annual dioxin emission for each source in grams TEQ per year was  
196 calculated as the product of the assigned air emission factor and the annual activity rate of the  
197 source.

198

### 199 **2.4. Estimates for emission uncertainty**

200 A confidence level of the annual emission intensity was established by source based on  
201 the quality of the information on technical parameters and activity rate for each source. The

202 quality of the information collected on technical parameters was classified as High (in-depth  
203 knowledge of technological parameters of interest), Medium (approximate knowledge of  
204 parameters of interest) or Low (parameters of interest are only known in a summary way).  
205 Similarly, the quality of the information on the activity rate for each source was classified as  
206 High (activity rate known), Medium (activity rate known for at least one year of the source) or  
207 Low (activity rate unknown: approximated using an average activity rate for similar sources).  
208 The overall confidence level of the annual emission intensity of each source was then derived  
209 by combining the confidence levels for the activity rate and the emission factor; this  
210 distinguished four categories (High, Medium+, Medium-, and Low) corresponding to the  
211 uncertainty in the annual emission estimates ( $\pm 30\%$ ;  $\pm 50\%$ ;  $\pm 75\%$  and  $\pm 100\%$ ) for each  
212 inventoried source (Coudon et al., 2018).

213

## 214 **2.5 Specific methodology for medical waste incineration**

215 A specific process was put in place for medical waste incineration due to the large number  
216 of sources over the French territory between 1990 and 2008, the absence of a specific database  
217 and difficulties in obtaining reliable historical information on technical characteristics and  
218 activity rates for each medical waste incinerator. According to (Tabeaud and Hamez, 2000), in  
219 the early 1990s, 54% of the 2676 medical centres in France were equipped with an internal  
220 incinerator. These incinerators were small ( $< 6$  t/h) and most of them had no air pollution  
221 control system and emission abatement (APC). Without any available information on the  
222 technologies, emissions from medical waste incinerators were estimated using a unique  
223 emission factor provided by the UNEP Toolkit for incinerators with “discontinuous operation”  
224 and “no or minimal APC”.

225 The tonnage of medical waste incineration was estimated based on the number of beds in  
226 the hospital. An average annual activity ratio of 0.38 t/bed was considered according to a study  
227 on medical waste incinerators in the French Rhône-Alpes Region (Vela, 2010). Given these  
228 assumptions, the lowest confidence level was assigned to medical waste incineration emissions.

229 Based on the French decree of January 1991 (restricting waste from incineration plants  
230 with a nominal capacity < 6t/h), small incinerators in hospital centres, local hospitals and  
231 dialysis centres were considered as inactive by the end of 1995.

## 232 **2.6. Geolocation of dioxin-emitting sources**

233 All identified dioxin-emitting facilities were manually geolocated using ARCGIS software  
234 (ESRI, ArcMap 10.1) based on aerial photography available from the Geographical National  
235 Institute (<https://www.geoportail.gouv.fr/>). When, facilities still existing and the chimney is  
236 visible, they were positioned at the chimney stack. When several stacks were identified on the  
237 aerial photography, the facility was positioned at equal distance from each of the chimneys. As  
238 necessary, to verify the correct location of the chimney on existing facilities, we used Google  
239 Street View images. When no chimney could be identified on the aerial photography, the  
240 facility was positioned at the centroid of the building. For facilities that had been closed for  
241 several years, we used historical aerial photography available from the Geographical National  
242 Institute (<https://remonterletemps.ign.fr/>). When neither a chimney nor the building could be  
243 identified, facilities were positioned, in descending order of precision, at the centroid of the  
244 parcel, the town hall of the municipality of the facility. For hospital waste incinerators, a  
245 specific geolocation by the French Ministry of Health was used  
246 (<https://www.atlasante.fr/accueil>) locating these facilities at the address plate or street segment.

247 Information on geographic coordinates and positional accuracy was stored for each source  
248 in the database.

## 249 **2.7. Comparison with emission measurements and other emission inventories**

250 The emission estimates provided by the UNEP Toolkit were compared with the dioxin  
251 declaration from the IREP database, available since 2003. Since 2003, the IREP registers  
252 mandatory declarations of facilities exceeding the reporting threshold of annual dioxin  
253 emissions of 0.1 g-TEQ/year. We compared each registration in the IREP database with the

254 UNEP Toolkit estimate for the corresponding source and activity.

255 At departmental level, we compared our estimates to the data of the French National Spatial  
256 Inventory (INS) from the French National Institute for Industrial Environment and Risk  
257 (INERIS). Dioxin emissions were estimated as the product of the activity rate and a specific  
258 emission factor (established for French emissions according to measurement data). These data  
259 were only publicly available for 2004, 2007 and 2012 ([http://emissions-air.developpement-](http://emissions-air.developpement-durable.gouv.fr)  
260 [durable.gouv.fr](http://emissions-air.developpement-durable.gouv.fr)).

261 Furthermore, we compared the emission estimates provided by the UNEP Toolkit with the  
262 annual emission inventory for France by the European Monitoring and Evaluation Program  
263 (EMEP; [www.ceip.at](http://www.ceip.at)). Since they used a different source classification than the UNEP Toolkit,  
264 we could only compare estimates for a limited number of source groups and categories  
265 matching the UNEP classification, i.e. group 2 (ferrous and non-ferrous metal production),  
266 group 4 (production of mineral products) and group 8 (miscellaneous), as well as categories 1.b  
267 (hazardous waste incineration) and 1.c (medical waste incineration).

268 The EMEP distinguishes municipal solid waste incineration (MSWI) with heat recovery  
269 (classified in the heat production sector) and MSWI without heat recovery (classified in the  
270 incineration sector). As we were unable to perform this distinction due to lack of data, we  
271 compared the sum of the EMEP emissions estimates for MSWI with heat recovery, and MSWI  
272 without heat recovery and heat production to the sum of the UNEP Toolkit estimates for  
273 category 1.a (municipal solid waste incineration) and group 3 (heat and power generation).

274

## 275 **3. Results**

### 276 **3.1. National inventory of dioxin sources from 1990 to 2008**

277 The overall response rate to the questionnaire sent to 447 facilities companies or operating  
278 groups still operating was 32%, despite mail and phone reminders. The highest response rates  
279 were observed for cement production (4.a, 100%); ferrous and non-ferrous metal production

280 (80%); lime production (4.b, 80%); sewage sludge incineration (1.e, 78%); and hazardous  
281 waste incineration (1.b, 70%). It should be noted that sole confirmation of information through  
282 phone contact with operators was not accounted for in the response rate. Some companies  
283 explicitly refused to respond. The main reasons for refusal were lack of time, information being  
284 difficult to access or lost, in particular for facilities which were no longer active, and mistrust  
285 in the study.

286 Overall, 2620 dioxin sources were identified over the French territory between 1990 and  
287 2008 (Table 1). Waste incineration (n=1913), and ferrous and non-ferrous metal production  
288 (n=241) represent, respectively, 73% and 9% of the sources inventoried. The remaining 18%  
289 correspond to heat and power generation (6%, n=165), production of mineral products (6%,  
290 n=156), production and use of chemicals and consumers goods (1%, n=13), and miscellaneous  
291 sources (5%, n=131). Medical waste incineration represents the most frequent source category  
292 (category 1.c, n=1442), followed by municipal waste incineration (1.a, n=377), fossil fuel  
293 plants (3.a, n=147) and crematoria (8.b, n=131). The categories of iron and steel production  
294 (2.ci, n=46; 2.c.ii, n=60), and aluminium production (2.e, n=54) constitute further categories  
295 comprising numerous sources. Setting aside medical waste incinerators, the total annual  
296 number of dioxin sources only slightly decreased over time from 905 in 1996 to 853 in 2008  
297 (Table S2). Of the 2620 sources identified, only 920 (35%) were active in 2008.

### 298 **3.2. Geolocations of the inventoried sources**

299 Figure 1 shows the geolocation of the inventoried sources. The location accuracy of the  
300 hospital waste incinerators (n=1444) is as follows: 62% are located at the address plate, 18% at  
301 the town hall, 18% at the track and 2% nearby. Of the remaining sources types (n=1176), 82%  
302 are located at the stack, 12% at the centroid of the building, 5% at the address plate and 1% at  
303 the town hall. **The sources with a coarse geographic scale (namely at the town hall) are almost**  
304 **exclusively waste incinerators destroyed in the early 1990s and located in the countryside. The**  
305 **source located at the address plate are factories destroyed during the study period and for which**  
306 **old aerial photos do not allow to improve the location.**

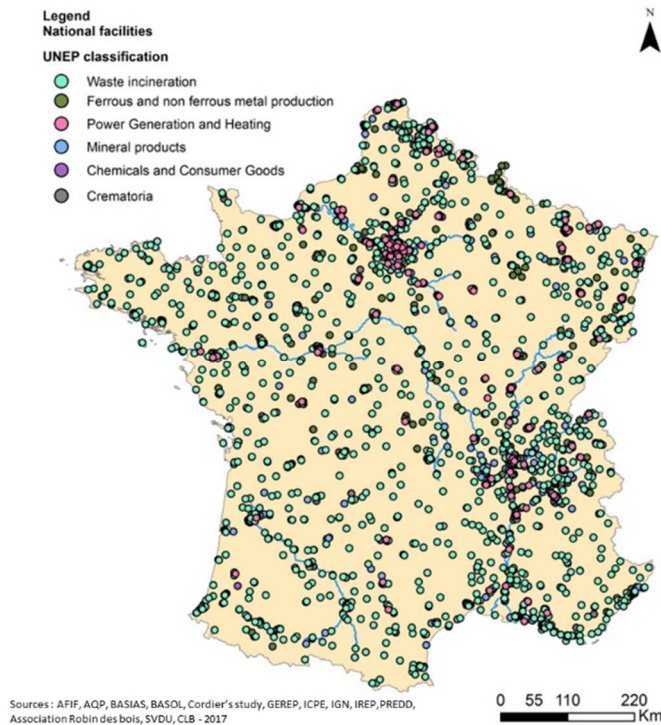


Figure 1 – Geolocations of the dioxin sources between 1990 and 2008 in France

307  
308  
309

### 3.3. Emission intensity estimates

310

311 The distribution of emission intensities by the UNEP Toolkit group and activity category  
312 is shown in Table 1. The median annual emission per source for waste incineration (0.402 g-  
313 TEQ/year) largely exceeds that of other groups, i.e. ferrous and non-ferrous metal production  
314 (0.017 g-TEQ/year), heat and power generation (0.001 g-TEQ/year), production of mineral  
315 products (0.008 g-TEQ/year), production and use of chemicals and consumer goods (0.017 g-  
316 TEQ/year) and miscellaneous sources (0.008 g-TEQ/year).

317 Municipal waste incineration (1.a) and iron ore sintering (2.a) are the categories with the  
318 most important median intensity per source of 3.85g TEQ/year and 1.3 g-TEQ/year,  
319 respectively. The median annual emissions of two additional categories are above 0.1 g-  
320 TEQ/year: medical waste incineration (1.c; 0.323 g-TEQ/year) and cement production (4.a;  
321 0.147 g-TEQ/year). A further six categories (Pulp and paper production; Hazardous waste  
322 incineration; Iron and steel making; Aluminium production; Biomass power plants and Waste  
323 wood and biomass incineration) display median emissions close to 0.5 g-TEQ/year (Table 1).

324 The estimated uncertainties of dioxin emission estimates vary across categories ranging  
325 from  $\pm 30\%$  (1.g, 2.a, 3.b, 7.a and 7.f) to  $\pm 100\%$  (1.c, 4.f and 8.b) (Table 1). At the national  
326 level, we observe an average uncertainty of  $\pm 62\%$  in the total emission estimates. Setting aside  
327 hospital waste incinerators (1.c, n=1442), characterised by the lowest confidence level, the  
328 uncertainty in the total emission estimates is  $\pm 53\%$ .

329 Table 1 – Median annual intensity of emissions (g-TEQ/year) by UNEP Toolkit group and category

Group	Nb of sources	Median (g-TEQ/year)	IQR	Uncertainty (%)	
1	Waste incineration	1913	0.402	0.180 – 0.981	96%
2	Metal production	241	0.017	0.002 - 0.105	53%
3	Heat and power generation	165	0.001	0.0002 - 0.007	40%
4	Production of mineral products	156	0.008	0.001-0.051	63%
7	Chemicals and consumer goods	14	0.048	0.009-0.080	29%
8	Miscellaneous	131	0.008	0.006-0.014	100%
Category	Nb of sources	Median (g-TEQ/year)	IQR	Uncertainty (%)	
1.a	Municipal solid waste incineration	377	3.850	1.750-8.467	49%
1.b	Hazardous waste incineration	67	0.055	0.010-0.229	63%
1.c	Medical waste incineration	1442	0.323	0.174-0.630	100%
1.e	Sewage sludge incineration	21	0.018	0.0040-0.029	45%
1.f	Waste wood and biomass incineration	1	0.04	NA	75%
1.g	Destruction of animal carcasses	5	0.011	0.045-0.146	30%
2.a	Iron ore sintering	7	1.31	0.603-6.000	30%
2.b	Coke production	5	0.018	0.010-0.042	75%
2.c.i	Iron and Steel Making	46	0.05	0.010-0.419	51%
2.c.ii	Iron Foundries	60	0.009	0.002-0.026	56%
2.c.iii	Hot-dip Galvanizing Plants	9	0.008	0.001-0.013	32%
2.d	Copper production	23	0.001	0.0001-0.056	75%
2.e	Aluminium production	54	0.05	0.014-0.179	59%
2.f	Lead production	14	0.01	0.002-0.020	42%
2.g	Zinc production	7	0.002	0.001-0.020	50%
2.h	Brass and bronze production	2	0.021	0.011-0.032	50%
2.j	Others non-ferrous metal production	14	0.003	0.002-0.022	36%
3.a	Fossil fuel power plant	147	0.001	0.0002-0.005	37%
3.b	Biomass power plants	9	0.05	0.028-0.059	30%
3.c	Landfill Biogas combustion	9	0.001	0.001-0.001	75%
4.a	Cement production	39	0.147	0.043-0.285	58%
4.b	Lime production	36	0.008	0.004-0.031	66%
4.c	Brick production	5	0.003	0.002-0.003	32%
4.d	Glass production	30	0.029	0.006-0.040	37%
4.f	Asphalt mixing	46	0.001	0.0001-0.001	100%
7.a	Pulp and paper production	13	0.075	0.012-0.080	30%
7.f	Chlorinated inorganic chemicals	1	0.007	NA	30%
8.b	Crematoria	131	0.008	0.006-0.014	100%

330

331

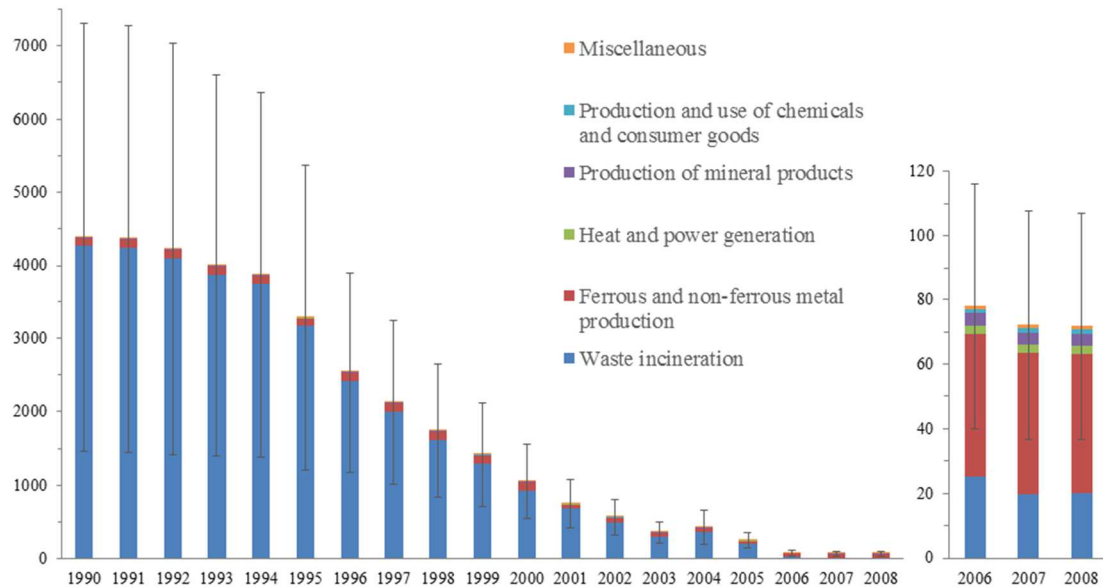
### 332 **3.4. Emission trends (1990-2008)**

333 Results show a 98.4% decrease in total dioxin emissions between 1990 and 2008 (Figure  
334 2). Waste incineration (UNEP Toolkit Group 1) constitutes the main contributor to total annual  
335 dioxin emissions until 2005, representing 95%, 85% and 75% of the total emissions  
336 respectively in 1990, 2000 and 2005. Over the last three years of the study period, 2006 -2008,  
337 emissions from ferrous and non-ferrous metal production (Group 2) instead are dominant.  
338 Emissions from waste incineration decreased by 99.5% over the whole period and two sharp  
339 decreases are observed in 1996 and 2005, subsequent to changes of regulatory standards in  
340 January 1991 and September 2002 involving the implementation of new abatement  
341 technologies for incinerators and the closing of hospital waste incinerators (Table S3). A similar  
342 decrease is observed over the whole period for ferrous and non-ferrous metal production, and  
343 for the production of mineral products (57% and 74%, respectively) (Table S4). The trend is  
344 different for the production and use of chemicals and consumer goods, and miscellaneous  
345 groups, whose emissions increased over the entire period by 31% and 100%, respectively  
346 (Table S5). However, the annual emissions of these two groups are low (1.27 and 1.26 g-  
347 TEQ/year, respectively, in 2008), representing less than 3% each of total emissions. Emissions  
348 from the heat and power generation group vary slightly over the study period (-8%) and remain  
349 negligible at 4% of total emissions. Overall uncertainties in emissions decrease over the years,  
350 from 67% in 1990 to 49% in 2008 (Table S2). Total uncertainties in emissions represent almost  
351 3 kg-TEQ/year in 1990, whereas in 2008, they represented 35 g-TEQ/year.

352 Except for Iron and steel making (category 2.c.i), all categories with emissions above 10  
353 g-TEQ/year in 1990 (1.a, 1.b, 1.c, 2.a, 1.e and 4.a), strongly decrease over the 18-year period  
354 by between 57% and 99% (Tables S3, S4 and S5). We observe an increasing trend in emissions  
355 for 8 categories out of 28. For crematoria (category 8.b), this increase in emissions is due to an  
356 almost three-fold rise of the number of crematoria from 41 in 1990 to 120 in 2008. For other  
357 categories, displaying a reduced number of sources ( $n < 10$ ) and overall weak emissions, an  
358 increased sensitivity to emission variations of a single source is observed. As for uncertainties  
359 in group emissions, we observe a decrease in the uncertainties by category (Tables S3, S4 and



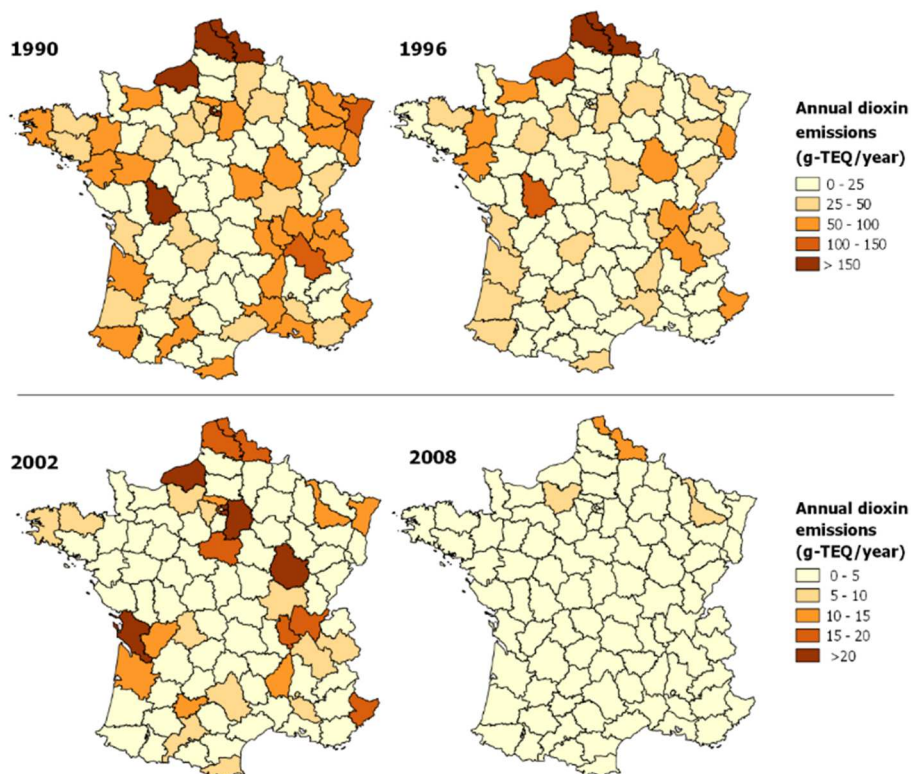
360 S5). Except for hospital waste incineration, the emission estimates for the 8 categories with a  
 361 median emission  $\geq 0.05$  g-TEQ/year over the study period were associated with relative low  
 362 uncertainties, ranging from 30% to 63%.



363

364 Figure 2 - Trends in dioxin emission estimates (g-TEQ/year) by UNEP group in France from 1990 to  
 365 2008

366 The decrease in emissions varies spatially (Figure 3). In 1990, we observe high spatial  
 367 differences in emissions over the French territory, with the highest emissions in the North and  
 368 strong emissions in the Rhône-Alpes region. While in 1990, 33 French departments (34%) have  
 369 an annual emission above 50 g-TEQ (including 4 departments with emissions above 150 g-  
 370 TEQ/year), there are only 12 (13%) with an annual emission above 50g-TEQ in 1996 (including  
 371 two departments with emissions above 150 g-TEQ/year) and the large majority of the French  
 372 departments have emissions between 0 and 25 g-TEQ/year. In 2002, the emissions of the  
 373 majority of the departments are below 5 g-TEQ/year. Some high emission values remain in the  
 374 most populated areas (Paris, Lyon and Bordeaux), often due to a single major source. In 2008,  
 375 almost all French departments have annual emissions below 5 g-TEQ/year (Figure 3).



376

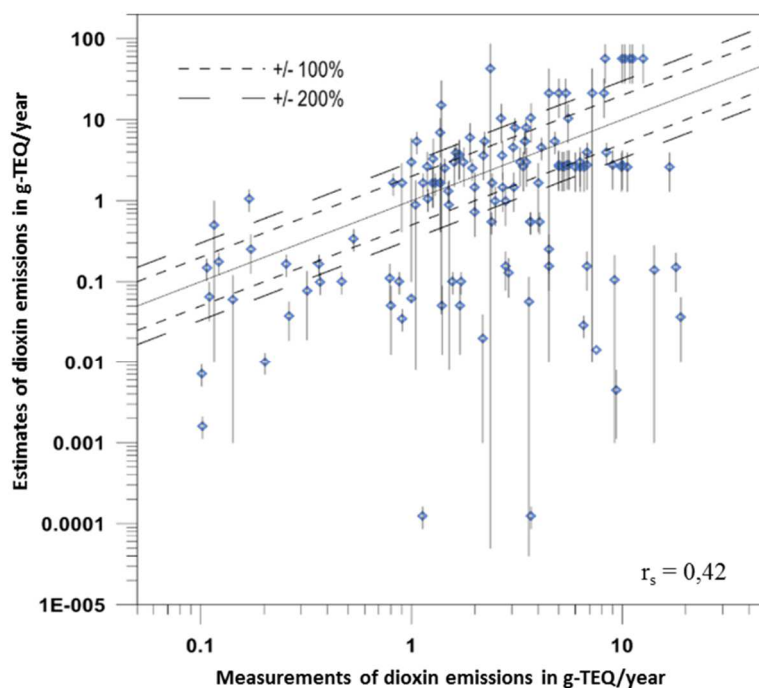
377 Figure 3 - UNEP Toolkit estimates of dioxin emissions by department in France from 1990 to 2008

378

### 379 3.5. Comparison of the estimates with available measurements (2003-2008)

380 We compared annual UNEP Toolkit emission estimates and dioxin emissions declared in  
 381 the IREP database, i.e. dioxin emissions exceeding the reporting threshold of annual dioxin  
 382 emissions of 0.1 g-TEQ/year in place since 2003 (Figure 4 and Table S6). Between 2003 and  
 383 2008, we identified 128 dioxin air emission declarations, corresponding to 59 distinct sources  
 384 (2.3% of the inventoried sources over the whole period). Overall, UNEP Toolkit estimates of  
 385 annual averaged emissions exceed emission measurements by 34%, while median source  
 386 annual emissions underestimate measurements by 40% (Median (IQR): 1.65 (0.16-3.64) g-  
 387 TEQ/year; 2.76 (1.27-5.51) g-TEQ/year). For waste incineration (63 declarations from 36  
 388 distinct sources), the median emission estimates (median (IQR): 2.51 (0.13-5.40) g-TEQ/year)  
 389 are close to the median emission measurements declared in the IREP (2.80 (1.64-4.73) g-  
 390 TEQ/year), with a difference below 10% (Table S6). For ferrous and non-ferrous metal

391 production (65 declarations from 23 distinct sources), median source annual emissions  
 392 underestimate median emission measurements declared in the IREP by 32% (1.65 (0.38-2.76)  
 393 versus 2.41 (0.93-6.54) g-TEQ, respectively). Yet, the declared measurements remain within  
 394 the range of the estimated uncertainty in the UNEP Toolkit estimates. Overall, the majority  
 395 (52%) of our estimates are within  $\pm 200\%$  of the IREP data for the corresponding facilities and  
 396 activity period (Figure 4) and the Spearman's rank correlation coefficient shows a “moderate”  
 397 correlation ( $r_s = 0.42$ ).



398  
 399 Figure 4 - Comparison between annual average estimates of dioxin emissions and declared dioxin  
 400 measurements exceeding the reporting threshold of annual dioxin emissions (0.1 g-TEQ/year) (IREP  
 401 database) in France between 2003 and 2008

402

### 403 3.6. Comparison with emission inventories

404 By using the EMEP emission inventory, we performed comparisons for the following  
 405 groups and categories: ferrous and non-ferrous metal production (group 2), production of  
 406 mineral products (group 4), miscellaneous sources (group 8), hazardous waste incineration  
 407 (category 1.b), medical waste incineration (1.c), and municipal solid waste incineration and  
 408 heat and power generation combined (group 3 and category 1.a) (see Table S7).

409 We observe discrepancies between the two inventories in emission estimates and historical  
410 trends for the different groups and categories. The UNEP Toolkit total emission estimates from  
411 1990 to 2008 underestimate the EMEP estimates for ferrous and non-ferrous metal production  
412 and hazardous waste incineration (by 37% and 60%, respectively) and overestimate the EMEP  
413 estimates for production of mineral products, crematoria and medical waste incineration (by  
414 approximately 130%, 1200% and 1800%). For group 8, the relative difference is high but the  
415 absolute total difference is low (0.86 g-TEQ/year on average).

416 The largest absolute difference is observed in 1990 for the group including both municipal  
417 solid waste incineration and heat and power generation, with an overestimation of the EMEP  
418 annual dioxin emissions by the UNEP Toolkit of nearly 2.4 kg (+200%) (Table S7). This  
419 difference progressively decreases over time: 1467 g-TEQ in 1995, 564 g-TEQ in 2000, 167 g-  
420 TEQ in 2004 and 12 g-TEQ in 2008. From 1990 to 2008, the difference between the EMEP  
421 estimates and UNEP Toolkit estimates falls by more than 95% for ferrous and non-ferrous  
422 metal production, municipal solid waste incineration and heat and power generation, production  
423 of mineral products groups and medical waste incineration. For crematoria and hazardous waste  
424 incineration, the difference increases over the years by 100% (0.6 g-TEQ) and 184% (22.5 g-  
425 TEQ), respectively.

426 We also performed a comparison at departmental level (Figure S1) with the data of the INS  
427 from the INERIS. At this scale, data were only available for two recent years: 2004 and 2007.  
428 Overall, results were similar to those at the national level (SM8).

429

## 430 **4. Discussion**

431 This study reports a national inventory to estimate annual dioxin emissions for France at  
432 the source level, from 1990 to 2008 as a basis for future use in epidemiological studies. To our  
433 knowledge, the present study is the first European historical inventory to include a systematic  
434 geolocation of sources from 1990 onwards, with a high spatial resolution taking into account

435 source's technological characteristics and the activity rates of a large number of dioxin sources  
436 (n=2620) including the assessment uncertainty of emission estimates. Our study covered older  
437 and longer periods as compared to previous published studies from US (Jones et al., 2018). We  
438 further compared the UNEP Toolkit estimates with emissions measurements, as well as  
439 inventory at national level (EMEP).

440 Our results showed a large decrease in atmospheric dioxin emissions in France between  
441 1990 and 2008 (-98.4%). Similar decreases over the same period in emissions from controlled  
442 sources have been described by others: for Europe (Quass et al., 2004), the United States  
443 (Dwyer and Themelis, 2015) and Japan (Government of Japan, 2012). The decrease in total  
444 dioxin emissions is due to the implementation of legislations and reduction measures more  
445 repressive towards dioxin-emitting sources. This has led to the closure of numerous facilities  
446 and implementation of emission-reduction practices. According to the Interprofessional  
447 Technical Centre for Air Pollution Studies (CITEPA), total dioxin emissions in France  
448 decreased by more than 90% between 1990 and 2008 (Serveau et al., 2013). It is worth noting  
449 that the observed reduction of emissions in our study and contribution of different dioxin source  
450 groups to total dioxin emissions was heterogeneous over the French territory over time.

451

452 In the absence of dioxin atmospheric monitoring data, the use of technical parameters to  
453 reconstruct historical dioxin atmospheric emissions has been shown to be a robust methodology  
454 (Cordier et al., 2010; Coudon et al., 2018; Fabre et al., 2008). The UNEP Toolkit provides a  
455 series of emission factors that may differ by a factor of three to ten for different technologies  
456 in many categories. The reliability of the information used to derive the emission factor and  
457 subsequent choice of the emission factor based on technical characteristics, as well as  
458 uncertainties in the activity rate may lead to large differences in the emission estimates for a  
459 specific source or group of sources. Uncertainty estimates are therefore an essential element of  
460 an emissions inventory. However, little is known about the accuracy of existing emission  
461 estimates and few emission inventories report uncertainties explicitly. Our observation that  
462 dioxin emission estimates are associated with large uncertainties, that vary across categories,

463 are consistent with observations from the literature (Breivik et al., 2006). Pulles et al., using  
464 Monte Carlo modelling, showed that the annual total emission estimates with the UNEP Toolkit  
465 for 2000, in 13 Central and Eastern European countries, was situated in a range of 2-3 times  
466 lower and 3–5 times higher than the mean value (Pulles et al., 2006). Quina et al. (2011), using  
467 the UNEP Toolkit, estimated that the total amount of PCDD/PCDF emissions was situated in a  
468 range of 2 times lower to 2.5 times higher the most likely value of the total annual amount of  
469 PCDD/PCDF released in Portugal in 2006 (Quina et al., 2011). An inventory of PCDD/PCDF  
470 releases estimates total annual emissions from 15 countries to be 10500 g I-TEQ/year for 1995,  
471 with an inferior value 1.3 times lower and an upper value approximately 3.5 times higher  
472 (Fiedler, 1999).

473 Similarly to Pulles et al., **who performed uncertainty analyses of dioxin emission estimates**  
474 **in 13 central and eastern European countries for the year 2000**, the largest contribution to the  
475 overall uncertainty was due to the uncertainty in emissions from hospital waste incinerators  
476 (Pulles et al., 2006). The large number of sources and the poor quality of available information  
477 strongly affected the average confidence level and subsequent uncertainty estimates. **These**  
478 **results are consistent with findings reported by Quass et al in a 1985-2005 European Dioxin**  
479 **Emission Inventory, observing considerable uncertainties regarding dioxin emissions from**  
480 **hospital incinerators during this period (Quass et al., 2004).** Our comprehensive data search  
481 that included multiple databases and use of historical information sources as well as direct  
482 contact with operators allowed for numerous facilities to obtain and confirm information on  
483 technical characteristics and activity rates. **Similarly to other studies, the lack of information**  
484 **on technical characteristics and activity rates of dioxin sources especially in the early 1990s**  
485 **constitutes an important limitation in our study. Furthermore, the overall response rate to the**  
486 **questionnaire sent to companies or groups of 447 still operating facilities was low (32%) despite**  
487 **mail and phone reminders. While response rates differed between groups and categories, we**  
488 **did not observe any overall difference between responding and non-responding groups, in terms**  
489 **of company size, activity rate or intensity of emissions. It is however worth noting that facilities**  
490 **still operating during the inventory phase (2015-2017) represent less than 20% of the total**

491 number of sources inventoried sources and a much lower proportion of total emissions, given  
492 that the most emitting sources have been closed during the 1990s.

493 In response to this and to assess the reliability of the UNEP Toolkit estimates, we compared  
494 our estimates with both measurements at the individual source level and an inventory providing  
495 global annual average dioxin emission estimates at the national (EMEP) scales.

496 Measurements of dioxin emissions are scarce (Coudon et al., 2018). The comparison of our  
497 annual UNEP Toolkit emission estimates to 128 dioxin emissions measures from 59 sources,  
498 declared since 2003 in the IREP database, showed a “moderate” correlation overall. The median  
499 measurements were situated within 1.5 times the median values of the annual Toolkit estimates.  
500 The discrepancies observed can be explained by several factors. The one-off measurements  
501 declared in the IREP database by the facility operators once a year have been taken for another  
502 purpose and may not be representative of the annual average emission. Furthermore, the  
503 variability at a single plant can be an order of magnitude between different sampling periods  
504 (Pacyna et al., 2003). Also, measurements were available only for municipal waste incinerators  
505 and the metal industry, and the number of sources was small.

506 The comparison at the national scale showed that the annual estimates from the EMEP  
507 emission database were up to 3.5 times lower compared to the annual UNEP Toolkit estimates  
508 for municipal solid waste incineration and heat and power generation, and production of  
509 mineral products. The largest differences were found for medical waste incineration.  
510 Conversely, EMEP emission estimates exceeded the UNEP Toolkit estimates for ferrous and  
511 non-ferrous metal production and hazardous waste incineration. The major differences were  
512 observed between 1990 and 1999 corresponding to the period with the highest dioxin  
513 emissions. The UNEP Toolkit estimates tend to get closer to the national inventory estimates  
514 for more recent years, probably due to better access to technical source data but also to the  
515 overall decline in emissions. These differences can be explained by differences in the emission  
516 factors and activity rates used to estimate emissions, the lack of technical data on facilities, and  
517 the absence of information on the right number of dioxin sources, such as hospital waste  
518 incinerators (Quass et al., 2004). For the latter, the factor of 10 difference is likely to be due to



519 the large number of sources in our inventory (n=1442) with low-quality information collected  
520 for this specific category. For the crematoria, the discrepancies are due to differences in the  
521 emission factors varying by a factor of 10, and to the low emissions of this category that favours  
522 large relative differences. Also, several studies suggest that for the period 1990-2000, dioxin  
523 emissions were underestimated due to a poor characterisation of emission sources (Brzuzy and  
524 Hites, 1996; Pacyna et al., 2003; Webster and Connett, 1998). In our study regarding municipal  
525 solid waste incineration, we inventoried numerous small MSWI in rural areas; most of them  
526 did not have an abatement system and were closed before 1996 due to their high emissions.  
527 These sources may not have been included in the EMEP inventory. For example, using specific  
528 databases from the Rhône-Alpes region, we inventoried numerous small waste incinerators  
529 located in the mountainous area.

530 *Additional comparison with the data of INS from INERIS at departmental level for two*  
531 *recent years (2004 and 2007) were consistent with those at the national level (SM8).*

532 Industrial emissions today are low in Europe and further refinement should incorporate  
533 other punctual and non-industrial sources that can emit relatively high amounts of dioxins  
534 (Coudon et al., 2018; Lee et al., 1999; Serveau et al., 2013) locally on an ad hoc basis over  
535 time, such as biomass fires, the burning of manufactured goods and cable, and outdoor burning  
536 and illegal landfills in the early 1990s. Yet, these poor or uncontrolled emission sources could  
537 not be considered here due to the lack of reliable activity data and estimates of such emissions,  
538 in particular retrospectively, and the difficulty of their geolocation. Furthermore, considering  
539 only air emissions from controlled sources may also lead to an underestimation of the total  
540 amount of PCDD/PCDF released (Brzuzy and Hites, 1996; Quina et al., 2011).

541

## 542 **5. Conclusion**

543 In the framework of an epidemiological study on the long-term effects of dioxin exposure  
544 on cancer risk, we created the first database containing annual dioxin emissions at the facility  
545 scale for 2620 facilities in France between 1990 and 2008, including geolocation at the



546 facilities' stack and the technical information needed to characterise emissions over time. Our  
547 study shows that the UNEP Toolkit estimates provide reliable estimates for some comparison  
548 and that the main difficulty is the availability of data and subsequent uncertainties in the  
549 emission estimates. The differences observed in the comparisons at the source, and  
550 departmental and national scales were within the estimated uncertainty ranges from our study  
551 and as reported in the literature (Breivik et al., 2006).

552 The final database contains further complementary information on the sources' technical  
553 parameters (namely stack height, smoke temperature and velocity). These served as a basis to  
554 develop a GIS-based metric in order to assess industrial airborne dioxin exposure in  
555 epidemiological studies (Danjou et al., 2019). For this purpose the sources' annual emission  
556 intensity (in g-TEQ/year), stack height and geographic location were integrated into the GIS-  
557 metric with information on wind direction, the residential history of study participants over the  
558 study period as well as the subject's residence-to-source distance. The development and  
559 performance evaluation of the GIS-based metric has been described in detail elsewhere  
560 (Coudon et al., 2019). This database could also be valuable for future studies to assess human  
561 exposure to other pollutants such as heavy metals (Coudon et al., 2018).

562

### 563 **Competing interests**

564 The authors declare to have no competing interests.

565

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575

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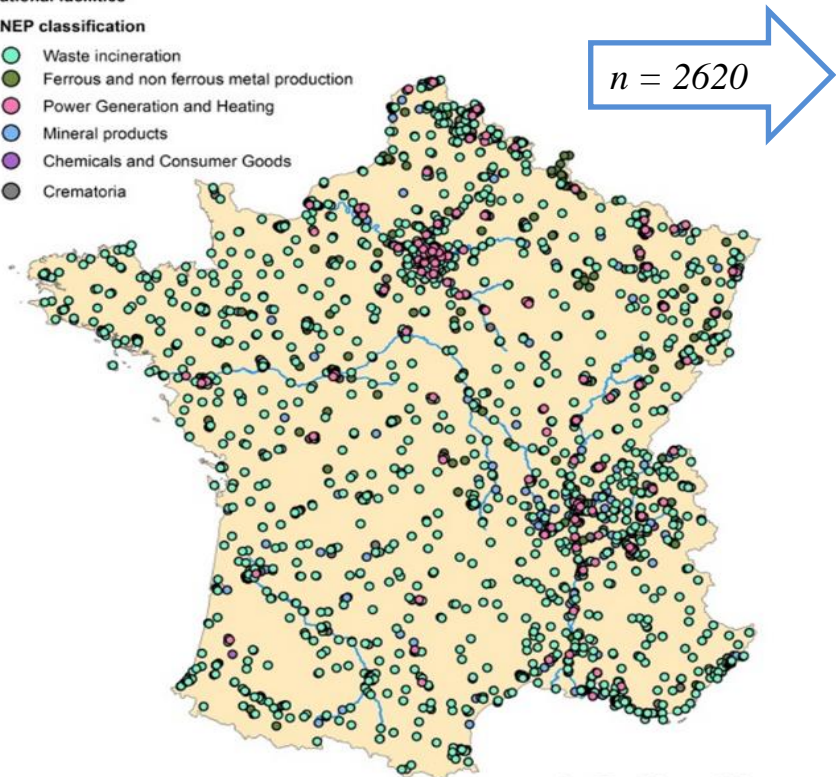
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# Inventory, geolocation, and atmospheric emissions estimates of dioxin sources in France between 1990 and 2008

## Legend National facilities

## UNEP classification

- Waste incineration
- Ferrous and non ferrous metal production
- Power Generation and Heating
- Mineral products
- Chemicals and Consumer Goods
- Crematoria



High location precision :  
Sources were located at the stack (43%) or at the building (27%)

