

2 **Atmospheric emissions of some POPs in Europe – a** 3 **discussion of existing inventories and data needs**

4 Knut Breivik ^{a,*}, Vigdis Vestreng ^b, Olga Rozovskaya ^c, Jozef M. Pacyna ^{a,d}

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6
7 ^a Norwegian Institute for Air Research (NILU), P.O. Box 100, NO-2027 Kjeller,
8 Norway.

9 ^b Norwegian Meteorological Institute, P.O. Box 43, Blindern, NO-0313 Oslo,
10 Norway.

11 ^c Meteorological Synthesizing Centre – East (MSC/E), Leningradsky prospekt
12 16/2, 125040 Moscow, Russia.

13 ^d Gdansk University of Technology, 11/12 G. Narutowicza Str, 80-952 Gdansk,
14 Poland.

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16 * Corresponding author. Phone: + 47 63 89 80 00, Fax: + 47 63 89 80 50
17 E-mail addresses: knut.breivik@nilu.no, vigdis.vestreng@met.no,
18 olga.rozovskaya@msceast.org, jozef.pacyna@nilu.no

19 20 **Abstract**

21
22 Accurate and complete information on the emissions of persistent organic
23 pollutants (POPs) are essential for interpreting historical, current and future
24 contamination levels in remote areas. However, this information is also crucial
25 for decision makers aiming at further reduction of the environmental loading of
26 these substances on a regional scale. In this study, we identify and discuss
27 specific POP emission inventory features that are deemed essential to
28 understand, predict and control the behaviour of such substances on a
29 European scale. It is shown that the scientific value of official emission data is
30 limited (e.g. for deriving source-receptor relationships on a European scale),
31 as there is insufficient information on spatial, temporal and compound
32 coverage. Likewise, we argue that non-official emission data (i.e. research-
33 driven emission inventories), which are based on aggregated statistics, may
34 be of limited value for the identification of further emission control strategies. It
35 is thus argued that future emission inventories should be developed in a
36 format that is suitable to serve both policy- and research oriented applications.
37 Further improvement of official emission data with respect to research-driven
38 features seems to be the most sensible way to proceed. Finally, the empirical
39 basis of current emission inventories remains weak, and further research on
40 emission identification and characterisation seem needed (a) to gain
41 confidence in predicted source-receptor relationship as well as (b) for the
42 development of sound control strategies.

44 1. Introduction

45

46 Persistent Organic Pollutants (POPs) are chemicals that are recognised as
47 persistent, bio-accumulative, toxic and susceptible to long-range atmospheric
48 transport (PBT-LRT). POPs generally fall into two classes dependent on their
49 origin; intentionally produced chemicals (typically organochlorinated pesticides
50 and industrial chemicals) and unwanted by-products of combustion
51 (polyaromatic hydrocarbons – PAHs, and dioxins - PCDD/Fs).

52 Elevated concentrations of several POPs have been observed in remote
53 environments far from global source regions, such as the Arctic (AMAP,
54 2004). Ottar (1981) hypothesised that several chlorinated hydrocarbons have
55 a potential for reversible atmospheric deposition, and that they therefore may
56 be subject to a systematic long-term transport from warmer to colder regions.
57 This theory was further elaborated by Wania and Mackay (1993) who offered
58 more detailed explanations how the temperature-dependent partitioning of
59 persistent and low volatility compounds could lead to an enrichment in the
60 Arctic through global fractionation and cold condensation processes. Later,
61 several modelling studies have been undertaken to understand and predict
62 source-receptor relationships for POPs on a European (e.g. VanJaarsveld et
63 al., 1997; Prevedouros et al., 2004) hemispherical (e.g. Malanichev et al.,
64 2004; Hansen et al., 2004) and global scale (e.g. Wania and Su, 2004).
65 International agreements are now also coming into force to reduce further
66 environmental exposure of POPs on a regional and global scale. One of such
67 international agreements is the Stockholm Convention on POPs (UNEP,
68 2001). The Stockholm Convention entered into force in May 2004 (151
69 signatories and 118 parties as of February 17, 2006). The Stockholm
70 Convention targets 12 POPs (the so-called “dirty dozen”) for reduction and
71 eventual elimination, and sets up a system for evaluation of additional
72 chemicals for consideration. Another international agreement is the 1979
73 Geneva Convention on long-range transboundary air pollution (LRTAP), which
74 has 49 parties (UN/ECE, 1979). The LRTAP convention has been extended
75 by the 1998 Aarhus protocol on Persistent Organic Pollutants (POPs), which
76 entered into force by the end of 2003 (25 ratifications as of January 13, 2006).
77 Following their entry into force, officially reported emission inventories by
78 parties are increasingly needed to (a) understand and predict source-receptor
79 relationships for such contaminants within both a scientific and regulatory
80 context, and (b) develop sound emission reduction strategies.

81 The key objectives of this evaluation have been:

82 (i) To identify and discuss specific data needs and requirements regarding
83 emission data for POPs by main users of such information (policy-makers,
84 scientists).



85 (ii) To compare and contrast policy-driven (official emission data) and
86 research-driven emission estimates¹ in terms of major features and selected
87 outputs.

88 (iii) To assess temporal trends in relative source contribution for selected
89 POPs on the basis official emission data.

90 (iv) To discuss if existing emission data include the necessary information
91 and features for source-receptor relationships to be predicted and understood
92 and for sound control strategies to be elaborated.

93 We emphasise that the discussion around official emission data in this study
94 solely relates to the information submitted by the Parties to the European
95 Monitoring and Evaluation Programme (EMEP) under the UN/ECE LRTAP
96 convention by 10th March 2005 (Vestreng et al., 2005).

97

98 **2. Policy and research-oriented features**

99

100 The features of a specific emission inventory commonly mirrors the specific
101 needs of main user(s) for whom the inventory is targeted. The two key groups
102 of users of emission inventories are: (i) policy-makers developing strategies
103 for the reduction of the environmental exposure to pollutants on a regional or
104 global scale, and (ii) scientists studying past, current and future source-
105 receptor relationships for pollutants on a regional and global scale. It may
106 therefore be useful to distinguish between policy-driven (officially reported
107 emission data by Parties to various international conventions) and research-
108 driven emission estimates. Policy-driven emission estimates tend to focus on
109 identification and control of POP sources, while research-driven emission
110 estimates provide information that helps understanding the impact of POP
111 emissions on the environment and human health. Official policy-driven
112 emission data for POPs are commonly developed and maintained by national
113 emission experts within various environmental protection authorities in the
114 Parties to the LRTAP convention (Vestreng et al., 2005). Research-driven
115 estimates are typically developed by groups of international emission experts
116 and target the specific needs of various research projects or environmental
117 assessments. Table 1 contains a simplified classification of various European
118 emission data according to identified policy- and science-oriented features,
119 each which are described and discussed in turn. It should be noted that there
120 are also other emission inventories for POPs available at present. These
121 inventories are not included in Table 1, but they were recently reviewed by
122 Breivik et al. (2004). The main reason for not including all studies conducted
123 so far is the limited geographical scope of some these inventories, i.e. for

¹ Research-driven emission inventories are sometimes referred to as emission expert estimates. It is our opinion that the latter term is inappropriate.

124 single countries (e.g. Alcock et al. 2001) or for certain parts of Europe only
125 (e.g. Quass et al. 2000; 2004).

126

127 **2.1 Policy-oriented features**

128

129 An essential policy-oriented feature of an emission inventory is a clear
130 identification of the individual sources and source categories of POP
131 emissions. Official data submitted by the Parties to the LRTAP convention
132 (Vestreng et al., 2005) are thought to be superior over the research based
133 emission inventories, because these national emission inventories are
134 collected from various industries at national and even international level or
135 they are estimated by emission experts in individual countries. These national
136 experts are expected to know the detailed characteristics in their respective
137 countries concerning relevant activity data, such as usage of individual
138 pesticides and industrial chemicals as well as relevant details concerning the
139 operation of individual plants (e.g. abatement technologies at individual waste
140 incineration facilities).

141 The officially reported POP emissions under the Convention on LRTAP are
142 submitted annually to the UN/ECE Secretariat. It is emphasised that the
143 EMEP emission database is a dynamic database, and the values contained in
144 this database are frequently updated and improved. The requirements for
145 reporting of these data are given in the Emission Reporting Guidelines
146 (UN/ECE, 2002). Emission inventories should be transparent, consistent,
147 comparable, complete, and accurate. The comparability of emission
148 inventories is attempted by encouraging all Parties to apply the guidance
149 given in the EMEP/CORINAIR Atmospheric Emission Inventory Guidebook
150 (EEA, 2005). Consistent time series of POPs sectoral and total emissions
151 should be reported from 1990 to the latest year (i.e. emission year 2003 for
152 2005 submissions) annually. The emissions should be complete with respect
153 to source categories included. More than 100 source categories are defined in
154 the reporting templates. Gridded data for the priority POPs (PAHs, HCB,
155 PCDD/Fs) are requested every fifth year starting in 1990. For the sake of
156 transparency, all parties are also encouraged to submit an Informative
157 Inventory Report (IIR) together with their emission data in which as a
158 minimum, methodologies other than those found in the Guidebook (EEA,
159 2005) and re-calculations should be described and a key source analysis
160 performed. More elaborated IIRs are welcomed, and may include e.g.
161 information on emission factors, emission trend analysis and uncertainty of
162 emission estimates. Since the 2003 trial review of emission data reported to
163 the Convention of LRTAP and under the NEC (National Emission Ceiling)
164 Directive, annual review of emission data has been undertaken and
165 documented (e.g. Vestreng et al., 2005). The IIRs have proven to be crucial
166 for a complete understanding of the review results. Like the other



167 requirements listed above, the accuracy of the emission data is the
168 responsibility of the parties. The officially reported and review emission data
169 are published in the UN/ECE web database, WEBDAB
170 (<http://webdab.emep.int/>), hosted and maintained by the EMEP Meteorological
171 Synthesizing Centre – West (MSC-W) in Oslo. For most other inventories
172 listed in Table 1, the information is usually compiled on an aggregated level
173 taking advantage of aggregated international statistics. It may therefore in
174 many cases be more difficult to take advantage of the “non-official” emission
175 inventories in the policy-oriented context of evaluating possible control
176 measures on a local scale (i.e. for specific emission sources within a country).
177 Prime examples of such aggregated inventories may be the dynamic PCB and
178 PBDE emission mass balance estimates by Breivik et al., (2002a,b) and
179 Prevedouros et al., (2004a), respectively. Other studies seem to rely on
180 official emission data, whenever available, thus only presenting additional
181 emission estimates when there are spatial and/or temporal gaps in the official
182 data (Berdowski et al., 1997; Pacyna et al., 2003; van der Gon et al., 2005).

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184

185 **2.2 Research-oriented features**

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187 2.2.1 Spatial coverage

188

189 For policy-oriented purposes, emission inventories are needed as a useful
190 documentation on the level of reduction of POP emissions agreed within the
191 LRTAP Convention. So far, only a limited number of Parties to the Aarhus
192 POPs protocol within the LRTAP Convention have reported the POP emission
193 data to EMEP. As previously recognised by Berdowski et al. (1997), the
194 EMEP emission database has significant gaps in the spatial coverage in
195 reporting of emission data for individual POPs or POP groups (Vestreng et al.,
196 2005). As previously mentioned, reliable emission data are essential for
197 research efforts aiming to understand and predict source-receptor
198 relationships POPs (e.g. on a European scale). Therefore, scientists studying
199 these issues request emission data that are complete across respective
200 model domain. In other words, the modeller needs to make sure that there are
201 no regions with no emission data in parts of the spatial domain of the model (if
202 significant emissions are assumed to occur in these regions). Most of the
203 emission data are reported on a country specific level (Table 1), while some
204 emission inventories are further distributed on a grid system, typically a 1° by
205 1° grid system for the global emission estimates (Li et al., 2000; 2003, Breivik
206 et al., 2002a) or finer resolution for some of the European emission estimates.
207 For example, the recent European inventory by van der Gon et al. (2005) is
208 presented on a 50 by 50 km² grid system.

209



210 2.2.2 Temporal coverage

211

212 POP emissions may vary considerably over a variety of time scales (e.g.
213 diurnal, seasonal, inter-annual). In the context of POPs, the key interest is
214 typically to obtain long-term (inter-annual) emission data trends (Table 1),
215 reflecting the potential environmental lifetime of the pollutant of concern. In the
216 case of POPs, this could be decades or even more (e.g. Sinkkonen and
217 Paasivirta, 2000; Wania and Daly, 2002). Many POPs have the potential for
218 undergoing reversible atmospheric deposition (Larsson, 1985; Jeremiason et
219 al., 1994). Atmospheric levels measured today, may thus be a legacy of
220 emissions that had occurred in the past. Several studies have thus attempted
221 to infer the relative importance of primary and secondary emissions in
222 controlling the contemporary atmospheric burden of POPs (e.g. Harrad et al.,
223 1994; Bailey, 2001; Bidleman and Leone, 2004; Hung et al., 2005; Kurt-
224 Karakus et al. 2006).

225

226 2.2.3 Speciation

227

228 Modellers also need information on the physical and/or chemical speciation of
229 individual POP substances (e.g. isomer or congener patterns). This issue may
230 appear to be of particular interest to scientists, but it is argued that it also has
231 important implications for the development of sound control strategies.
232 Information on speciation or congener patterns may be critical because it
233 influences the distance of POP transport within the air masses. PAHs, HCHs,
234 PCBs and PCDD/Fs contain several individual species with different physical-
235 chemical properties, environmental degradation rates and thus potential for
236 atmospheric long-range transport (e.g. Beyer et al., 2003). For example Li et
237 al. (2002) have illustrated how β -HCH, unlike α -HCH, exhibited only limited
238 propensity for transport into the Arctic in spite of identical chemical formulas
239 and similar emission histories of these two HCHs.

240

241 2.2.4 Multimedia features

242

243 Under certain circumstances and for the more volatile POPs, the multimedia
244 feature of POP releases into the environment could be crucial for
245 understanding atmospheric source-receptor relationships. For example,
246 subsequent transfer of PCBs from contaminated soils and sediments to the
247 atmosphere have been hypothesised to have significant implications for the
248 PCB mass balance (Chiarenzelli et al., 1997). Multimedia emission inventories
249 may thus be of particular interest for attempts to evaluate the relative impact
250 of various contaminant pathways to polluted water bodies (e.g. Breivik and
251 Wania, 2002), or studies aiming to evaluate the relative significance of primary
252 anthropogenic (controllable) emissions versus secondary re-emissions from



253 environmental reservoirs as contaminated in the past (e.g. Harrad et al. 1994;
254 Bailey, 2001; Hung et al. 2005).

255

256 2.2.5 Additional data needs

257

258 Some modellers may in addition require more detailed emission input
259 information. These specific needs are typically dependent on the specific
260 geometries of each individual model, and it is therefore considered out of the
261 scope to discuss these specifics in detail. However, it may be mentioned that
262 many of these additional input needs are typically related to physical
263 properties governing emission data (such as emission release heights,
264 particle-size distribution, stack exhaust temperatures and velocities etc).

265

266 **2.3 Common data needs**

267

268 Common to the needs of both policy-makers and scientists is the request for
269 accurate emission information. Information on emission data uncertainty is
270 highly valuable when evaluating the uncertainty of predicted source-receptor
271 relationships as well as the potential risk of choosing erroneous control
272 strategies. Little is known about the accuracy of available emission estimates,
273 although a few of the emission inventories listed in Table 1 contain some
274 information on emission data uncertainties. For example, Berdowski et al.
275 (1997) report uncertainty factors for the emission estimates of individual
276 groups of pollutants at European scale (e.g. PAHs: 2-5; PCBs: 2-5 and
277 PCDD/Fs: 5-20), whilst Breivik et al. (2002a,b) and Prevedouros et al. (2004a)
278 present various high and low emission estimates in addition to default
279 estimates. Information on emission data uncertainties in the official emission
280 inventories are contained in the informative inventory reports (IIRs) previously
281 mentioned. IIRs containing information on emission data uncertainties for
282 POPs have recently been submitted by Denmark (Illerup et al. 2005), Finland
283 (SYKE, 2005) and France (CITEPA, 2004). Interestingly, attempts have also
284 recently been made to quantify uncertainties in a study of dioxin emission
285 estimates for Central Europe, using Monte-Carlo analysis (Pulles et al. 2006).

286

287 **3. A comparison of different inventories for the year 1990**

288

289 **3.1 National totals**

290

291 One potential approach to evaluate uncertainties of an emission inventory is
292 to compare and contrast emission estimates prepared through different
293 methodologies. Figure 1 compares results from three different emission
294 inventories for selected POPs for the reference year 1990. The reference year

295 1990 is considered to be of specific interest, as the UN/ECE Aarhus protocol
296 on POPs obliges Parties to reduce their emissions of PCDD/Fs, PAHs and
297 HCB below their levels in 1990 (or an alternative year between 1985 and
298 1995). In addition to the EMEP data, Figure 1 also includes results from a
299 comprehensive study by Berdowski et al. (1997), as well as a study by
300 Pacyna et al. (2003). We emphasise that the data from Berdowski et al.
301 (1997) and Pacyna et al. (2003) are based on the quantitative knowledge on
302 POP emissions at the time these studies were published, whereas the EMEP
303 database is frequently updated as new and better information becomes
304 available. Thus, differences between these studies may in part be explained
305 by improved characterisation of some specific POP emission sources over the
306 last decade.

307

308 3.1.1 PAHs

309

310 PAHs and dioxins (PCDD/Fs) are the POPs that are most extensively
311 reported to EMEP by the Parties to the LRTAP convention (Figure 1). This
312 may not come as a surprise, as the source categorisation and emission
313 inventory methodologies that were originally developed for classical air
314 pollutants (EEA, 2005), clearly fit the typical source-profiles of these pollutants
315 better than for pesticides and industrial chemicals. For PAHs, there are both
316 official reported EMEP data² and estimates from Berdowski et al. (1997)
317 available for 23 Parties (Fig. 1A). The study by Berdowski et al. (1997)
318 includes the sum of the Borneff six PAHs (benzo[*a*]pyrene,
319 benzo[*b*]fluoranthene, benzo[*g,h,i*]perylene, benzo[*k*]fluoranthene,
320 fluoranthene and indeno[1,2,3-*c,d*]perylene), while EMEP request the parties
321 to address four out of these six PAHs (benzo[*a*]pyrene, benzo[*b*]fluoranthene,
322 benzo[*k*]fluoranthene, and indeno[1,2,3-*c,d*]perylene). A comparison with the
323 European emission data from Pacyna et al. (2003) was omitted, as this study
324 considered benzo[*a*]pyrene only. Differences in speciation are likely to explain
325 in part why the total PAH emissions for the 23 Parties are about three times
326 higher by Berdowski et al. 1997 (in comparison to the EMEP data). However,
327 the estimates for individual countries often deviate substantially, and in
328 several cases this difference is more than one order of magnitude (for AT, BG,
329 DE, DK, EE, FR, IS, RU)³. Some Parties report higher national emissions than
330 Berdowski et al. (1997) (i.e. BG, CZ, NL). Hence a more limited compound
331 coverage cannot explain the differences between these two inventories.

² The EMEP database reports a high value of PAH emissions from “Accidental and Natural Fires” (NFR source category 5E) in GB (about twice as high as the remaining total emissions from the 23 countries included in Figure 1). This value was disregarded in this study, as it is reported as a memo item and should not be included in the national total. The emissions reported from IS are too small (0.087 Mg) to show up in the figure 1A.

³ For interpretation of ISO2 country codes, please see www.emep.int/grid/country_numbers.txt



332

333 3.1.2 PCDD/Fs

334

335 Dioxins are among the POPs that have received a lot of attention, primarily
336 because of their toxicity at very low concentrations. As an unintentional by-
337 product of combustion, emissions of dioxins are also expected to take place in
338 all countries⁴. The results of two independent estimates are compared with
339 the EMEP data in Figure 1B. Although there is a fair agreement between
340 these estimates for some countries, large discrepancies (more than 100%)
341 between the EMEP data and one or both independent estimates are evident
342 for BG, CZ, DK, FI, HU, IS, NO, SE, SK.

343

344 3.1.3 PCBs

345

346 Only 9 Parties to the LRTAP Convention have submitted official PCB emission
347 data (greater than zero) for 1990. For the total emissions of PCBs from these
348 9 countries, it can be seen that the estimate by Berdowski et al., (1997) is
349 about twice as high as the EMEP data (Figure 1C). Again, difference in
350 compound coverage within the group of PCBs is an issue that may help to
351 explain deviations between these two estimates. The estimates by Berdowski
352 et al. (1997) address total PCBs (i.e. the sum of 209 different compounds)
353 when dealing with emissions from leakage or evaporation - or the sum of six
354 frequently reported congeners (PCB-28, PCB-52, PCB-101, PCB-118, PCB-
355 153 and PCB-180) for other emission categories. It is not known which
356 congeners of PCBs are included in the EMEP database. Some countries (e.g.
357 The Netherlands and Spain) are reporting zero PCB emissions to EMEP, and
358 it may be questioned if this is plausible based on the widespread global usage
359 of PCBs (Breivik et al., 2002b).

360 The limited availability of emission data for PCBs from various countries
361 indicates that control strategies for PCBs (and other industrial chemicals) may
362 be different in comparison to the PAHs and PCDD/Fs. For example, current
363 control strategies in Norway are typically targeted towards the safe collection
364 and destruction of remaining materials and products containing PCBs, rather
365 than based on differences in quantitative source strength with respect to
366 atmospheric emissions. Secondly, there are fundamental methodological
367 difficulties addressing emissions of industrial chemicals like PCBs (Breivik et
368 al. 2002b; Prevedouros et al. 2004a). In order to quantify atmospheric
369 emissions of substances that are intentionally produced, consideration must
370 be given to the potential for atmospheric losses that may occur during the
371 entire lifecycle of these chemicals (production, transport, storage, use and
372 disposal). Furthermore, there are also fundamental difficulties in determining
373 suitable emission factors for PCBs for volatilisation processes (i.e.

⁴ Denmark reports zero emissions of PCDD/Fs in 1990.

374 evaporation from contaminated products or environmental surface media in
375 contact with air). This is because (a) such emissions depend on the
376 equilibrium status between the air and the product / surface media being
377 contaminated with PCBs (e.g. Mackay, 2001), and (b) emissions due to
378 volatilisation are expected to vary significantly in response to temperature
379 changes (e.g. Haque et al. 1974; Haugen et al. 1998; Breivik et al. 2002b).
380 Thus elevated emissions due to volatilisation are expected to take place when
381 “clean” air passes over a contaminated surface at elevated temperatures.

382

383

384 **3.2 Source categories**

385

386 Overall, the simple comparison presented in Figure 1 provides an idea of the
387 uncertainties in available emission estimates for the individual European
388 countries. A closer look at differences in predicted emissions from various
389 source categories may provide further insight into why there are notable
390 differences between these inventories. Since 2002, emissions reported to the
391 LRTAP Convention, have been reported according to the UN/ECE Emissions
392 reporting Guidelines (UN/ECE, 2002) along the so-called Nomenclature For
393 Reporting (NFR) source categories. In order to compare the research-based
394 emission estimates and the EMEP emission data, the NFR categories have
395 been aggregated to the Selected Nomenclature for Air Pollutants (SNAP)
396 according to the corresponding allocation Table III A in the Guidelines
397 (UN/ECE, 2002). For some countries, reported emissions in the SNAP
398 categories have been used directly, since the 1990 emissions were only
399 available in SNAP sectors.

400 Figure 2 shows a comparison of the EMEP data against data presented by
401 Berdowski et al., (1997) and Pacyna et al., (2003). The data are presented at
402 the SNAP Level I for the sake of comparability⁵. Table 2 presents the relative
403 importance of emissions from various SNAP source categories in controlling
404 emissions in selected European countries in 1990. It can be observed that the
405 number of countries considered in Figure 2 and Table 2 is lower than the one
406 in Figure 1. This is because some countries only report their national total
407 emissions to EMEP. Also given in the last column in Table 2 are the total
408 emissions in the countries considered.

409

410 3.2.1 PAHs

411

412 Table 2 shows that the estimates of PAH emissions by Berdowski et al.,
413 (1997) attribute almost half of the emissions in selected European countries to

⁵ Level 1 is the more coarse aggregation of emission source categories (as opposed to SNAP Levels 2 and 3). A comparison of data at SNAP Level 2 and 3 has not been feasible in this study.



414 SNAP 2 (non-industrial combustion), while the EMEP data attribute about one
415 third of the emissions in this year to SNAP 6 (solvent use). In general, there is
416 a very poor correlation between reported emission data from Berdowski et al.,
417 (1997) and the EMEP data aggregated to SNAP Level I that are reported for
418 individual countries (Figure 2A). In many cases, the two estimates differ by
419 several orders of magnitude. A closer inspection at the EMEP data in
420 comparison to the data from Berdowski et al. (1997) also indicates that
421 important sources in some countries may have been left out in the EMEP
422 data. Prime examples are Estonia and Germany, which both only reports
423 emissions of PAHs from road transport (SNAP 7). It seems safe to claim that
424 for these countries, there is a lack of completeness in terms of source
425 coverage in the EMEP data. As can be seen from Figure 1A, the EMEP data
426 are lower than the data from Berdowski et al., (1997) in 20 out of 23 countries.
427 Figure 2 A also demonstrates this underestimation of EMEP data compared to
428 estimates by Berdowski et al., (1997). Although this may in part be explained
429 by the more limited coverage of PAH species in the EMEP database, it could
430 also be explained by a more complete coverage of emission sources in the
431 study by Berdowski et al., (1997). There are other examples where the data
432 for individual source categories are far higher in the study by Berdowski et al.,
433 (1997) in comparison with the EMEP data, but it is not possible to conclude
434 which one is the more correct estimate. A specific example is the emissions of
435 PAHs from non-industrial combustion (SNAP 2) in France. Berdowski et al.,
436 (1997) suggest that these emissions are higher than 3000 tonnes, while the
437 official EMEP data are almost two orders of magnitude lower (39 tonnes).

438

439 3.2.2 PCDD/Fs

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441 A comparison of PCDD/Fs emissions data in EMEP with inventories made by
442 Berdowski et al. (1993) and Pacyna et al. (2003) is shown in Table 2 and
443 Figures 2 B,C. Specifically, the total emissions from selected countries are
444 comparable, and all inventories identify waste treatment and disposal (SNAP
445 9) as the primary source category for dioxins on a European scale (Table 2).
446 It should be cautioned, however, that waste incineration may not necessarily
447 be a key source of dioxins in all European countries, as previously recognised
448 for Belarus (Kakareka, 2002). As can be seen from Table 2, the EMEP
449 database also identifies public power (SNAP 1) and industrial combustion
450 (SNAP 3) as two important source categories for dioxins, while Pacyna et al.,
451 (2003) suggest that significant emissions from non-industrial combustion
452 (SNAP 2) as well as production processes (SNAP 4) took place in the
453 selected European countries in 1990. These differences may in part be
454 explained by differences in SNAP allocation in individual countries with
455 significant national emissions (Figure 2C). France may serve as an example,
456 although the total emissions in France are suggested to be fairly similar in the

457 reported emission inventories (Figure 1). The EMEP data report the national
458 total emissions to be 1.77 Kg I-TEQ year⁻¹, and the three major source
459 categories for which these emissions are attributed are SNAP 1 (47.2%),
460 SNAP 9 (28.0%) and SNAP 3 (20.0%). Likewise, Berdowski et al., (1997)
461 report the national total emissions in France to be 1.64 Kg I-TEQ year⁻¹, for
462 which the key sources are SNAP 3 (54.5%), SNAP 9 (31.4%) and SNAP 2
463 (10.6%). Finally, Pacyna et al., (2003) report the national total emissions in
464 France to be 1.23 Kg I-TEQ year⁻¹, with the key sources being SNAP 9
465 (41.8%), SNAP 2 (30.9%) and SNAP 4 (22.2%).

466 For dioxins, there is an additional concern whether all significant sources have
467 been included in the inventory (completeness). The dioxin emissions from the
468 open burning of household waste have received considerable attention in
469 recent years (e.g. Lemieux et al., 2000/2004; Wevers et al., 2004). However,
470 reliable estimates of the relative importance of such emissions are considered
471 difficult because of the lack of reliable activity data and emission factors
472 related to open burning.

473

474 3.2.3 PCBs

475

476 Only six European countries have reported official emission data for PCBs by
477 source category for the year 1990. The total PCB emissions from these six
478 countries amount to 10.4 tonnes according to EMEP, and 27.1 tonnes
479 according to Berdowski et al., (1997). In the case of the EMEP data, 78.6% of
480 these emissions are attributed to SNAP 6. According to the explanatory notes
481 from United Kingdom (which reported the highest PCB emissions among the
482 countries listed in Table 2 – see also Figure 1), this lumped source category
483 includes emissions from capacitors and transformers and the use of
484 halogenated chemicals in the chemical industry. Berdowski et al., (1997) also
485 report that most of the PCB emissions were attributed to electrical equipment,
486 but did not assign these emissions to any existing SNAP source category
487 (Table 2).

488

489 **4 Temporal changes in key source categories**

490

491 It is argued that official emission data should be the preferred choice of
492 emission information, as official estimates seem to be most suitable for
493 evaluation of emission reductions. Table 3 presents a key source analysis for
494 selected POPs according to the EMEP data alone. The determination of key
495 sources is a fairly simple ranking technique, which is considered useful to
496 identify the importance of various sources in drawing conclusions about the
497 emission current levels and emission trends (e.g. Rypdal and Flugsrud, 2001;
498 Rypdal, 2002). Inventory improvements may then be directed towards the key

499 sources that have been identified, although there is a general concern if all
500 key sources are included in official national emission inventories (see
501 discussion above). For simplicity, only the top ten source categories are listed
502 in Table 3 for those pollutants that have more than ten source categories
503 contributing to 95% of the total emissions. It should be observed that the
504 number of countries included in the comparison is rather limited. This is
505 because we only included Parties for which data in NFR (Nomenclature for
506 Reporting) format are available (Vestreng et al., 2005) for the years
507 considered in this study. Table 3 includes data for both 1990 and 2003 in
508 order to evaluate potential temporal changes of the key sources. The reported
509 emission reductions in selected European countries from 1990 to 2003 are
510 ~80% in the case of PCDD/Fs and PCBs, whereas a reduction of ~47% is
511 suggested for PAHs in the same period (2nd last row of Table 3). Only a few
512 Parties report increasing emission from 1990 to 2003 (BE, DK and IS in the
513 case of PAHs, and FR in the case of PCBs).

514 The frequent assignment of POP emissions to “other” source categories as
515 well as the limited number of sources listed, serve to illustrate that the official
516 reporting scheme (originally developed for classical air pollutants) may be
517 considered less suitable when applied to industrial chemicals and pesticides.
518 An extension of the NFRs to increase the completeness and transparency in
519 the reporting of POPs should therefore be considered when the UN/ECE
520 Emission Reporting Guidelines are to be revised in 2007.

521 For PAHs, various (other) processes in the chemical industries and metal
522 production were the two key sources in 1990 (Table 3). By 2003, residential
523 plants are suggested to be the key source of PAHs, contributing almost a third
524 of the total emissions from the European countries included in Table 3. The
525 data indicate that a significant shift has occurred over the time, in which
526 emissions from residential plants have increased in relative importance on the
527 expense of various industrial emissions. Likewise, residential plants are also
528 suggested to be the most significant source of dioxin emissions in 2003,
529 following stringent emission controls in other sectors (e.g. waste incineration
530 from 1990). For PCBs, only five source categories are addressed and only
531 two countries are included (GB, FR)⁶. The total PCB emissions from these
532 two countries are dominated by emissions from GB (99% and 94% in 1990
533 and 2003, respectively), and other processes in the chemical industries (i.e.
534 emissions from capacitors and transformers in the case of G.B.) are attributed
535 as the key source in both years.

536

537 **5 Final remarks**

538

⁶ SE reports minor national emissions of PCBs, but for “national navigation” only. In addition, SE also reports PCB emissions from “international navigation”.



539 While a number of efforts have been made to improve emission inventories
540 needed for better understanding major environmental problems, such as
541 stratospheric ozone depletion, climate change and acid deposition, inventories
542 for POPs have for many years been considered unreliable and inaccurate
543 (Graedel et al., 1993; Pacyna and Graedel, 1995). A number of studies have
544 concluded that emission data are frequently the most uncertain input
545 information that determines the overall uncertainty of model predictions for
546 POPs (Vallack et al., 1998; Cohen et al., 2002; Malanichev et al., 2004). In
547 fact, it has often been claimed that the emissions of POPs remain the least
548 understood part of the research on overall distribution and fate of these
549 chemicals in the environment (Vallack et al., 1998; Wania and Mackay, 1996;
550 Jones and de Voogt, 1999; Breivik and Alcock, 2002). The simple comparison
551 of emission data in this study illustrates some of the difficulties with current
552 emission data for POPs, and in comparing and contrasting emission data of
553 different origin.

554 At present, it is not possible to firmly conclude which of the available emission
555 inventories are the most accurate. Even so, it is suggested that individual
556 countries may take further advantage of the research-based emission
557 inventories (i.e. non-official data) and use this information in search for
558 improvement of their official national emission inventories. For example,
559 research-based emission inventories may assist in identifying and filling gaps
560 with respect to some significant source categories that may have been
561 ignored or missing in the official emission data. The lack of consideration of
562 other PAH sources than road transport in the German and Estonian emission
563 inventories may serve to illustrate this point. It is also concluded that the non-
564 official data are at elevated risk of providing erroneous estimates, due to a
565 lack of knowledge on specific factors or control strategies that may have been
566 implemented in various individual countries.

567 Whilst official emission data should be the preferred choice of emission
568 information, it is shown that the EMEP data contain incomplete information on
569 spatial, temporal, and speciation features. This generally mitigates the use of
570 official emission data in the context of source-receptor modelling on a regional
571 (e.g. European) scale. For this reason, modellers may still have to rely on
572 research-driven estimates, which have typically aimed to present the more
573 general picture of emissions of individual POP substances in quantitative
574 terms (e.g. Breivik et al., 2002a,b; Prevedouros et al., 2004a).

575 The upcoming revision of the UN/ECE Emissions Reporting Guidelines and
576 future studies on emission inventories of POPs shall recognise and address
577 more clearly the specific features of POP emission inventories related to their
578 accuracy and completeness⁷. This consideration is needed for both scientific
579 and regulatory purposes of developing emission inventories of POPs.

⁷ The interested reader may note that a new report on the EMEP emission data has been issued since the submission of this manuscript (Vestreng et al. 2006)

580 Further development of official emission data is also dependent on the
581 outcome of future research on the generation of POPs during various
582 anthropogenic and natural processes. This research should focus on the
583 improvement of methodological approaches leading to the establishment of
584 more accurate methods of POP emission estimates.

585 The outcome of this study also calls for further efforts on the standardisation
586 of POP emission inventories and data parameters. This is an important issue
587 that still needs further consideration for future comparative studies and inter-
588 regional decision-making on a regional European basis. This study may in this
589 respect also have potential implications for relevant regional activities taking
590 place on other continents and even globally (i.e. work under the Stockholm
591 Convention on POPs; UNEP, 2001). As for this latter agreement, an important
592 step for dioxins and furans has already been made. For these chemicals a
593 standardised toolkit for identification and quantification of dioxin and furan
594 releases have been issued to assist countries to establish emission
595 inventories (UNEP, 2005). Use of this guidebook among countries is thus
596 expected to contribute to consistent emission data over time and between
597 countries.

598

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602 three anonymous reviewers for helpful comments on this manuscript.

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853



854 **Biographical note**

855

856 Knut Breivik is a senior scientist at the Norwegian Institute for Air Research
857 (NILU). He is also project manager for activities related to POPs that are
858 carried out at the Chemical Co-ordinating Centre (CCC) under the UN/ECE
859 European Monitoring and Evaluation Programme (EMEP). A key research
860 interest is on bridging the gap in the quantitative understanding between
861 sources and environmental fate of POPs.

862

863 Vigdis Vestreng is a scientist at the Norwegian Meteorological Institute. She is
864 project manager for the quality control, storage, and distribution of information
865 on emissions that are compiled at the Meteorological Synthesizing Centre -
866 West (MSC-W) under the EMEP Programme.

867

868 Olga Rozovskaya is a scientist working with emission inventories for POPs
869 and Heavy Metals at the Meteorological Synthesizing Centre-East (MSC-E) in
870 Moscow. MSC-E is responsible for the development and operational use of
871 numerical models of HMs and POPs under the EMEP Programme.

872

873 Jozef M. Pacyna is department director at the Center for Ecological
874 Economics at the Norwegian Institute for Air Research. Prof. Pacyna's
875 expertise is based on studies on biogeochemical cycling and fluxes of various
876 chemicals and radionuclides through the environment and source-receptor
877 relationships of various chemicals in the local, regional, and global
878 environment.

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897 **Figure captions**

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899 **Figure 1** Comparison of different emission estimates for selected POPs in
900 1990. The data are sorted from EMEP high to EMEP low. The
901 numerical values are reported in Vestreng et al., (2005)⁸.

902

903 **Figure 2** Comparison of emission estimates from Berdowski et al., (1997)
904 and Pacyna et al., (2003) versus EMEP data at SNAP Level 1 for
905 the reference year 1990. A) PAHs (unit in tonnes year⁻¹), B) and C)
906 PCDD/Fs (unit in g I-TEQ year⁻¹).

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⁸ The data reported by Berdowski et al. (1997) sometimes refer to officially reported data submitted for a different reference year than 1990. Data for PAHs and PCBs from Germany refer to data for 1985-1990, data from Luxembourg and the Netherlands refer to data from 1993, while data from Sweden refer to the years 1987 and 1991.



929 **Table 1.** Simplified classification of selected emission inventories for POPs
 930 with European coverage, according to policy- and science-oriented
 931 features (see *text for discussion*). A parenthesis indicates that some
 932 information is available on the subject. NA = Not applicable.

	Policy-oriented features	Research-oriented features				Common
	Source identification	Spatial (by country)	Temporal (inter-annual)	Speciation	Multimedia	Uncertainty
EMEP (official data)	+	(+)	(+)	(+)	-	(+)
Berdowski et al., 1997	(+)	+	-	-	-	(+)
Pacyna et al., 2003	(+)	+	+	(+)	-	-
Prevedouros et al., 2004a	(+)	+	+	+	-	(+)
Van der Gon et al., 2005	(+)	+	+	(+)	-	-
Li et al., 2000/2003	(+)	+	+	+	-	-
Breivik et al., 2002a,b	(+)	+	+	+	-	(+)
Bailey (2001)	(+)	-	-	NA	-	(+)

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945 **Table 2.** Relative importance of emissions from various SNAP source categories^{A)} in controlling emissions in selected European
 946 countries^{B)} in the reference year 1990 (%). The latter column gives the total emissions in the European countries
 947 considered (Units are thousand tonnes per year for PAHs, kg I-TEQ per year for PCDD/Fs and tonnes per year for
 948 PCBs). NI = No information.

		SNAP (% of Total)												Total
		1	2	3	4	5	6	7	8	9	10	11	Other	
PAHs	EMEP (2005)	2.7	20.5	6.4	19.9	<0.1	34.4	10.6	1.5	2.1	2.0	<0.1	NI	3.5
	Berdowski et al. (1997)	0.3	45.2	1.3	11.5	0	28.8	11.2	1.1	<0.1	0	0.4	NI	12.5
PCDD/Fs	EMEP (2005)	20.1	18.2	19.9	12.2	0.1	0.1	1.3	0.9	25.6	0.8	0.8	NI	8.4
	Berdowski et al. (1997)	7.1	19.7	15.7	23.0	<0.1	<0.1	1.0	0.5	33.0	0	<0.1	NI	7.5
	Pacyna et al. (1997)	5.7	30.0	6.0	26.4	0	<0.1	1.9	0.2	29.5	0	0.3	NI	8.9
PCBs	EMEP (2005)	6.5	2.6	2.4	5.7	0	78.6	1.4	0.2	2.6	0	0	NI	10.4
	Berdowski et al. (1997)	3.1	0.5	0.4	1.3	0	0	0.3	0	0.1	0	0	94.2	27.1

949 A) Definition of SNAP source categories: 1. Combustion in energy and transformation industries; 2. Non-industrial combustion;
 950 3. Combustion in manufacturing industry; 4. Production processes; 5. Extraction and distribution of fossil fuels and geothermal
 951 energy; 6. Solvent and other product use; 7. Road transport; 8. Other mobile sources and machinery; 9. Waste treatment and
 952 disposal; 10. Agriculture; 11. Other sources and sinks. Other: Electrical equipment.

953 B) PAHs: NL, BG, GB, BE, ES, PL, IT, FR, SK, SE, SI, RU, AT, FI, HR, NO, DK, MD, DE, EE, IS. PCDD/Fs: FR, GB, DE, RU,
 954 BE, BG, PL, IT, CH, SK, ES, AT, NO, SE, LU, FI, IS. PCBs: GB, PL, SI, BG, SK, FR.

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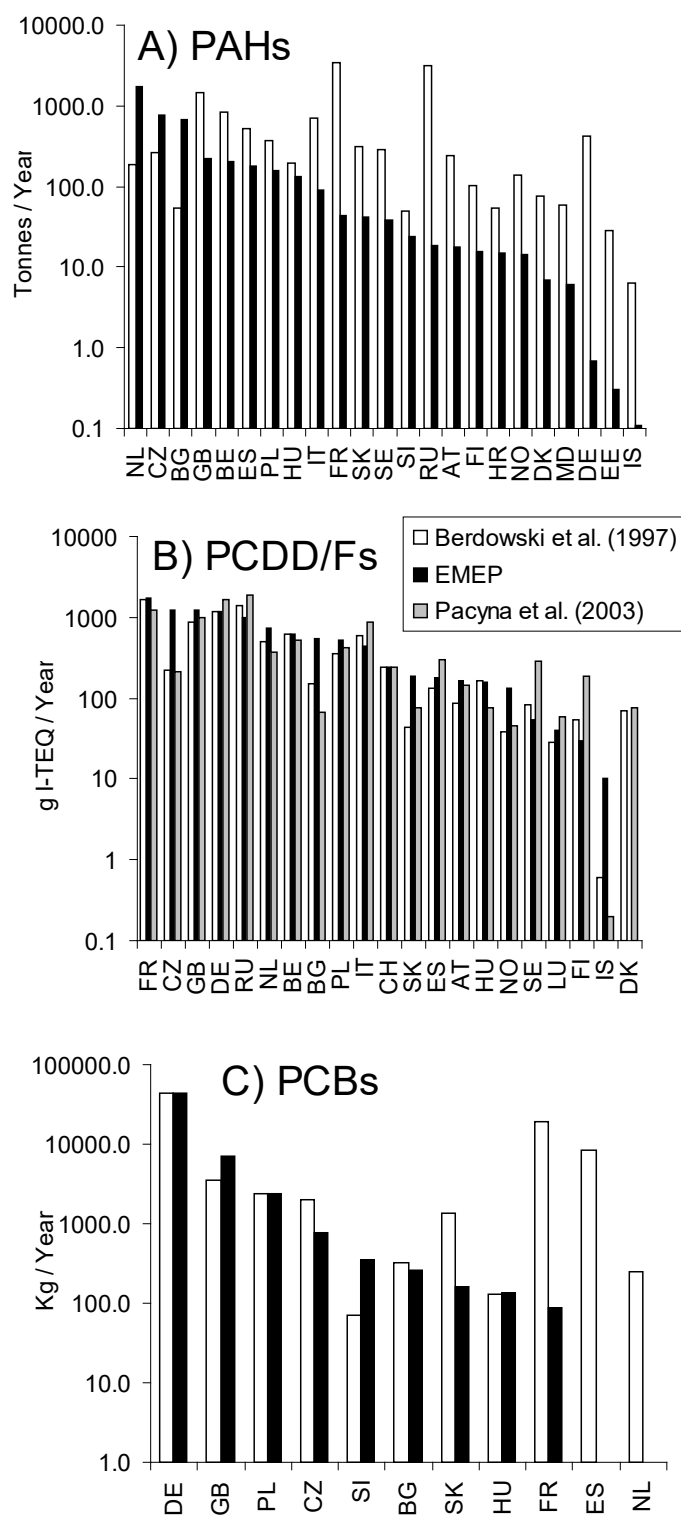


959 **Table 3.** Key Source Analysis for selected POPs in 1990 and 2003 in European countries for which official (EMEP) data for both
 960 years are available^A). The numbers in parenthesis give the relative contribution to total emissions, whilst the total
 961 emissions are presented in the 2nd last row. The last row gives the number of source categories not included, as only the
 962 top ten source categories are listed.

PAHs		PCDD/Fs		PCBs	
1990	2003	1990	2003	1990	2003
Other industrial processes (27.4)	Residential plants (30.8)	Waste incineration (30.1)	Residential plants (20.7)	Other industrial processes (86.2)	Other industrial processes (66.1)
Metal production (21.2)	Solvent & other product use ^B) (17.9)	Public electricity & heat prod. (25.1)	Waste incineration (17.9)	Metal production (6.9)	Waste incineration (10.9)
Solvent & other product use ^B) (12.7)	Other industrial processes (12.3)	Other, waste (9.9)	Public electricity & heat prod. (14.7)	Waste incineration (2.6)	Metal production (9.5)
Residential plants (8.0)	Metal production (9.3)	Metal production (9.3)	Iron and steel (10.4)		Public electricity & heat prod. (8.1)
Road transport passenger cars (5.8)	Road transport, passenger cars (4.0)	Iron and steel (8.6)	Metal production (9.8)		Iron and steel (1.7)
Field burning of agric. waste (4.2)	Non-ferrous metals (3.1)	Residential plants (3.2)	Other, manufacturing industries & construction (7.0)		
Road transport, heavy duty vehicles (3.2)	Road transport, heavy duty vehicles (2.8)	Non-ferrous metals (2.3)	Non-ferrous metals (5.4)		
Paint application (1.7)	Field burning of agric. waste (2.2)	Commercial / institutional plants (2.1)	Solvent & other product use ^B) (2.1)		
Non-ferrous metals (1.5)	Waste incineration (1.9)	Other, manufacturing industries & construction (1.6)	Other, waste (1.8)		
Solid fuel transformation (1.5)	Road transport, light duty vehicles (1.7)	Field burning of agric. waste (1.4)	Petroleum refining (1.5)		
2.4 ktonnes year ⁻¹	1.3 ktonnes year ⁻¹	4.9 Kg I-TEQ year ⁻¹	1.0 Kg I-TEQ year ⁻¹	7.2 tonnes year ⁻¹	1.5 tonnes year ⁻¹
41	41	2	5	0	0

963 A) The following countries are included in the key source analysis: PAHs: AT, BE, DK, FR, IS, NL, NO, ES, SE, GB. PCDD/Fs: AT,
 964 BE, FR, IS, NL, NO, ES, SE, GB. PCBs: FR, GB. B) Including products containing POPs.





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