Postprint of: Breivik K., Vestreng V., Rozovskaya O., Pacyna J. M., Atmospheric emissions of some POPs in Europe: a discussion of existing inventories and data needs, Environmental Science & Policy, Vol. 9, Iss. 7–8 (2006), pp. 663-674, DOI: 10.1016/j.envsci.2006.09.001

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

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Abstract

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

1. Introduction

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

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

The key objectives of this evaluation have been:

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

(*ii*) To compare and contrast policy-driven (official emission data) and research-driven emission estimates[1] in terms of major features and selected outputs.

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

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

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

2. Policy and research-oriented features

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

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

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

2.1 Policy-oriented features

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

The officially reported POP emissions under the Convention on LRTAP are submitted annually to the UN/ECE Secretariat. It is emphasised that the EMEP emission database is a dynamic database, and the values contained in this database are frequently updated and improved. The requirements for reporting of these data are given in the Emission Reporting Guidelines (UN/ECE, 2002). Emission inventories should be transparent, consistent, comparable, complete, and accurate. The comparability of emission inventories is attempted by encouraging all Parties to apply the guidance given in the EMEP/CORINAIR Atmospheric Emission Inventory Guidebook (EEA, 2005). Consistent time series of POPs sectoral and total emissions should be reported from 1990 to the latest year (i.e. emission year 2003 for 2005 submissions) annually. The emissions should be complete with respect to source categories included. More than 100 source categories are defined in the reporting templates. Gridded data for the priority POPs (PAHs, HCB, PCDD/Fs) are requested every fifth year starting in 1990. For the sake of transparency, all parties are also encouraged to submit an Informative Inventory Report (IIR) together with their emission data in which as a minimum, methodologies other than those found in the Guidebook (EEA, 2005) and re-calculations should be described and a key source analysis performed. More elaborated IIRs are welcomed, and may include e.g. information on emission factors, emission trend analysis and uncertainty of emission estimates. Since the 2003 trial review of emission data reported to the Convention of LRTAP and under the NEC (National Emission Ceiling) Directive, annual review of emission data has been undertaken and documented (e.g. Vestreng et al., 2005). The IIRs have proven to be crucial for a complete understanding of the review results. Like the other requirements listed above, the accuracy of the emission data is the responsibility of the parties. The officially reported and review emission data are published in the UN/ECE web database, WEBDAB (http://webdab.emep.int/), hosted and maintained by the EMEP Meteorological Synthesizing Centre – West (MSC-W) in Oslo. For most other inventories listed in Table 1, the information is usually compiled on an aggregated level taking advantage of aggregated international statistics. It may therefore in many cases be more difficult to take advantage of the “non-official” emission inventories in the policy-oriented context of evaluating possible control measures on a local scale (i.e. for specific emission sources within a country). Prime examples of such aggregated inventories may be the dynamic PCB and PBDE emission mass balance estimates by Breivik et al., (2002a,b) and Prevedouros et al., (2004a), respectively. Other studies seem to rely on official emission data, whenever available, thus only presenting additional emission estimates when there are spatial and/or temporal gaps in the official data (Berdowski et al., 1997; Pacyna et al., 2003; van der Gon et al., 2005).

2.2 Research-oriented features

2.2.1 Spatial coverage

For policy-oriented purposes, emission inventories are needed as a useful documentation on the level of reduction of POP emissions agreed within the LRTAP Convention. So far, only a limited number of Parties to the Aarhus POPs protocol within the LRTAP Convention have reported the POP emission data to EMEP. As previously recognised by Berdowski et al. (1997), the EMEP emission database has significant gaps in the spatial coverage in reporting of emission data for individual POPs or POP groups (Vestreng et al., 2005). As previously mentioned, reliable emission data are essential for research efforts aiming to understand and predict source-receptor relationships POPs (e.g. on a European scale). Therefore, scientists studying these issues request emission data that are complete across respective model domain. In other words, the modeller needs to make sure that there are no regions with no emission data in parts of the spatial domain of the model (if significant emissions are assumed to occur in these regions). Most of the emission data are reported on a country specific level (Table 1), while some emission inventories are further distributed on a grid system, typically a 1º by 1º grid system for the global emission estimates (Li et al., 2000; 2003, Breivik et al., 2002a) or finer resolution for some of the European emission estimates. For example, the recent European inventory by van der Gon et al. (2005) is presented on a 50 by 50 km^2 grid system.

2.2.2 Temporal coverage

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

2.2.3 Speciation

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

2.2.4 Multimedia features

Under certain circumstances and for the more volatile POPs, the multimedia feature of POP releases into the environment could be crucial for understanding atmospheric source-receptor relationships. For example, subsequent transfer of PCBs from contaminated soils and sediments to the atmosphere have been hypothesised to have significant implications for the PCB mass balance (Chiarenzelli et al., 1997). Multimedia emission inventories may thus be of particular interest for attempts to evaluate the relative impact of various contaminant pathways to polluted water bodies (e.g. Breivik and Wania, 2002), or studies aiming to evaluate the relative significance of primary anthropogenic (controllable) emissions versus secondary re-emissions from environmental reservoirs as contaminated in the past (e.g. Harrad et al. 1994; Bailey, 2001; Hung et al. 2005).

2.2.5 Additional data needs

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

2.3 Common data needs

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

3. A comparison of different inventories for the year 1990

3.1 National totals

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

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

3.1.1 PAHs

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

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[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

3.1.2 PCDD/Fs

Dioxins are among the POPs that have received a lot of attention, primarily because of their toxicity at very low concentrations. As an unintentional by-product of combustion, emissions of dioxins are also expected to take place in all countries[4]. The results of two independent estimates are compared with the EMEP data in Figure 1B. Although there is a fair agreement between these estimates for some countries, large discrepancies (more than 100%) between the EMEP data and one or both independent estimates are evident for BG, CZ, DK, FI, HU, IS, NO, SE, SK.

3.1.3 PCBs

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

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

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

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

3.2 Source categories

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

Figure 2 shows a comparison of the EMEP data against data presented by Berdowski et al., (1997) and Pacyna et al., (2003). The data are presented at the SNAP Level I for the sake of comparability[5]. Table 2 presents the relative importance of emissions from various SNAP source categories in controlling emissions in selected European countries in 1990. It can be observed that the number of countries considered in Figure 2 and Table 2 is lower than the one in Figure 1. This is because some countries only report their national total emissions to EMEP. Also given in the last column in Table 2 are the total emissions in the countries considered.

3.2.1 PAHs

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

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[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.

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

3.2.2 PCDD/Fs

A comparison of PCDD/Fs emissions data in EMEP with inventories made by Berdowski et al. (1993) and Pacyna et al. (2003) is shown in Table 2 and Figures 2 B,C. Specifically, the total emissions from selected countries are comparable, and all inventories identify waste treatment and disposal (SNAP 9) as the primary source category for dioxins on a European scale (Table 2). It should be cautioned, however, that waste incineration may not necessarily be a key source of dioxins in all European countries, as previously recognised for Belarus (Kakareka, 2002). As can be seen from Table 2, the EMEP database also identifies public power (SNAP 1) and industrial combustion (SNAP 3) as two important source categories for dioxins, while Pacyna et al., (2003) suggest that significant emissions from non-industrial combustion (SNAP 2) as well as production processes (SNAP 4) took place in the selected European countries in 1990. These differences may in part be explained by differences in SNAP allocation in individual countries with significant national emissions (Figure 2C). France may serve as an example, although the total emissions in France are suggested to be fairly similar in the reported emission inventories (Figure 1). The EMEP data report the national total emissions to be 1.77 Kg I-TEQ year^{-1}, and the three major source categories for which these emissions are attributed are SNAP 1 (47.2%), SNAP 9 (28.0%) and SNAP 3 (20.0%). Likewise, Berdowski et al., (1997) report the national total emissions in France to be 1.64 Kg I-TEQ year^{-1}, for which the key sources are SNAP 3 (54.5%), SNAP 9 (31.4%) and SNAP 2 (10.6%). Finally, Pacyna et al., (2003) report the national total emissions in France to be 1.23 Kg I-TEQ year^{-1}, with the key sources being SNAP 9 (41.8%), SNAP 2 (30.9%) and SNAP 4 (22.2%).

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

3.2.3 PCBs

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

4 Temporal changes in key source categories

It is argued that official emission data should be the preferred choice of emission information, as official estimates seem to be most suitable for evaluation of emission reductions. Table 3 presents a key source analysis for selected POPs according to the EMEP data alone. The determination of key sources is a fairly simple ranking technique, which is considered useful to identify the importance of various sources in drawing conclusions about the emission current levels and emission trends (e.g. Rypdal and Flugsrud, 2001; Rypdal, 2002). Inventory improvements may then be directed towards the key sources that have been identified, although there is a general concern if all key sources are included in official national emission inventories (see discussion above). For simplicity, only the top ten source categories are listed in Table 3 for those pollutants that have more than ten source categories contributing to 95% of the total emissions. It should be observed that the number of countries included in the comparison is rather limited. This is because we only included Parties for which data in NFR (Nomenclature for Reporting) format are available (Vestreng et al., 2005) for the years considered in this study. Table 3 includes data for both 1990 and 2003 in order to evaluate potential temporal changes of the key sources. The reported emission reductions in selected European countries from 1990 to 2003 are ~80% in the case of PCDD/Fs and PCBs, whereas a reduction of ~47% is suggested for PAHs in the same period (2nd last row of Table 3). Only a few Parties report increasing emission from 1990 to 2003 (BE, DK and IS in the case of PAHs, and FR in the case of PCBs).

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

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

5 Final remarks

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

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

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

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

The upcoming revision of the UN/ECE Emissions Reporting Guidelines and future studies on emission inventories of POPs shall recognise and address more clearly the specific features of POP emission inventories related to their accuracy and completeness[7]. This consideration is needed for both scientific and regulatory purposes of developing emission inventories of POPs.

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[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)

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

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

Acknowledgements

Financial support from EMEP is gratefully acknowledged. We would also like to thank the Parties for collecting and submitting emission data to EMEP, and three anonymous reviewers for helpful comments on this manuscript.

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Figure captions

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

Figure 2 Comparison of emission estimates from Berdowski et al., (1997) and Pacyna et al., (2003) versus EMEP data at SNAP Level 1 for the reference year 1990. A) PAHs (unit in tonnes year^{-1}), B) and C) 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.

Table 1. Simplified classification of selected emission inventories for POPs with European coverage, according to policy- and science-oriented features (*see text for discussion*). A parenthesis indicates that some 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	-	(+)

Table 2. Relative importance of emissions from various SNAP source categoriesA) in controlling emissions in selected European countriesB) in the reference year 1990 (%). The latter column gives the total emissions in the European countries considered (Units are thousand tonnes per year for PAHs, kg I-TEQ per year for PCDD/Fs and tonnes per year for 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**

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

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, BE, BG, PL, IT, CH, SK, ES, AT, NO, SE, LU, FI, IS. PCBs: GB, PL, SI, BG, SK, FR.

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

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, BE, FR, IS, NL, NO, ES, SE, GB. PCBs: FR, GB. B) Including products containing POPs.

Figure 1

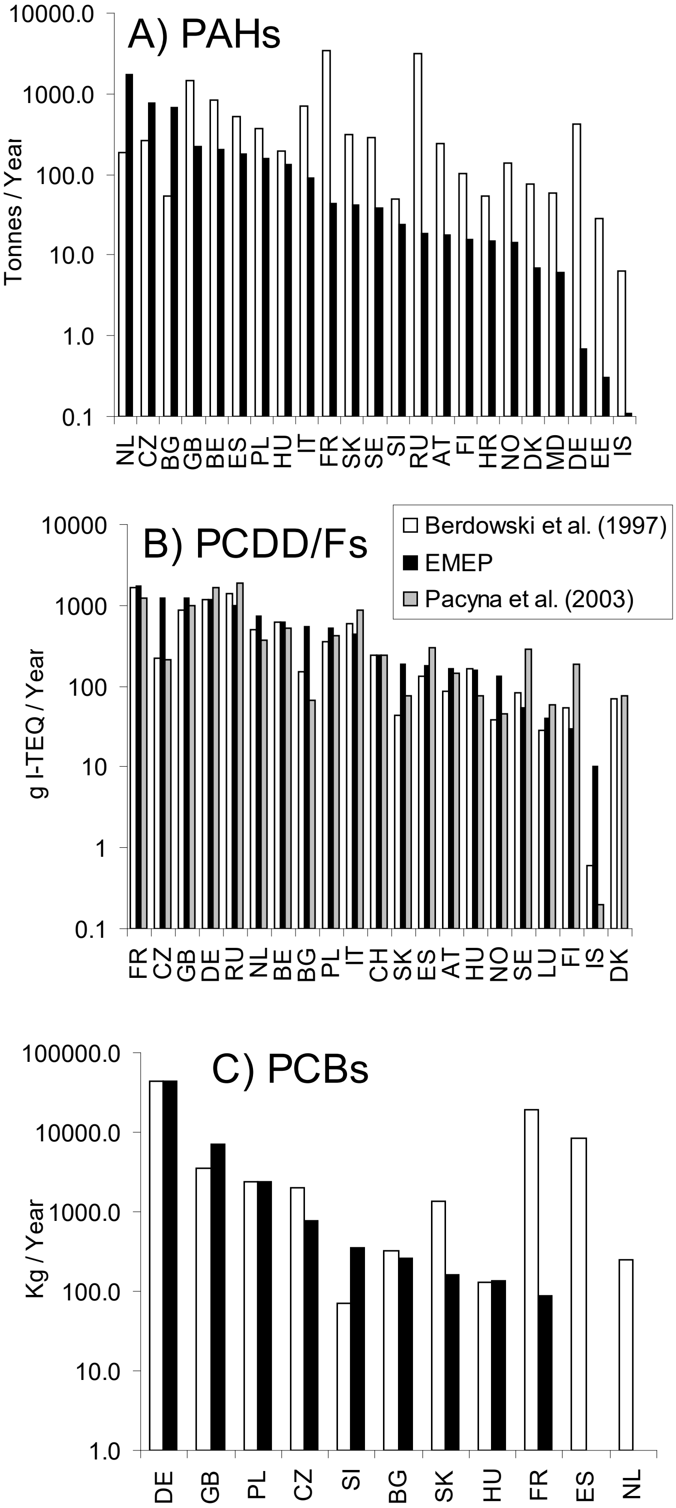
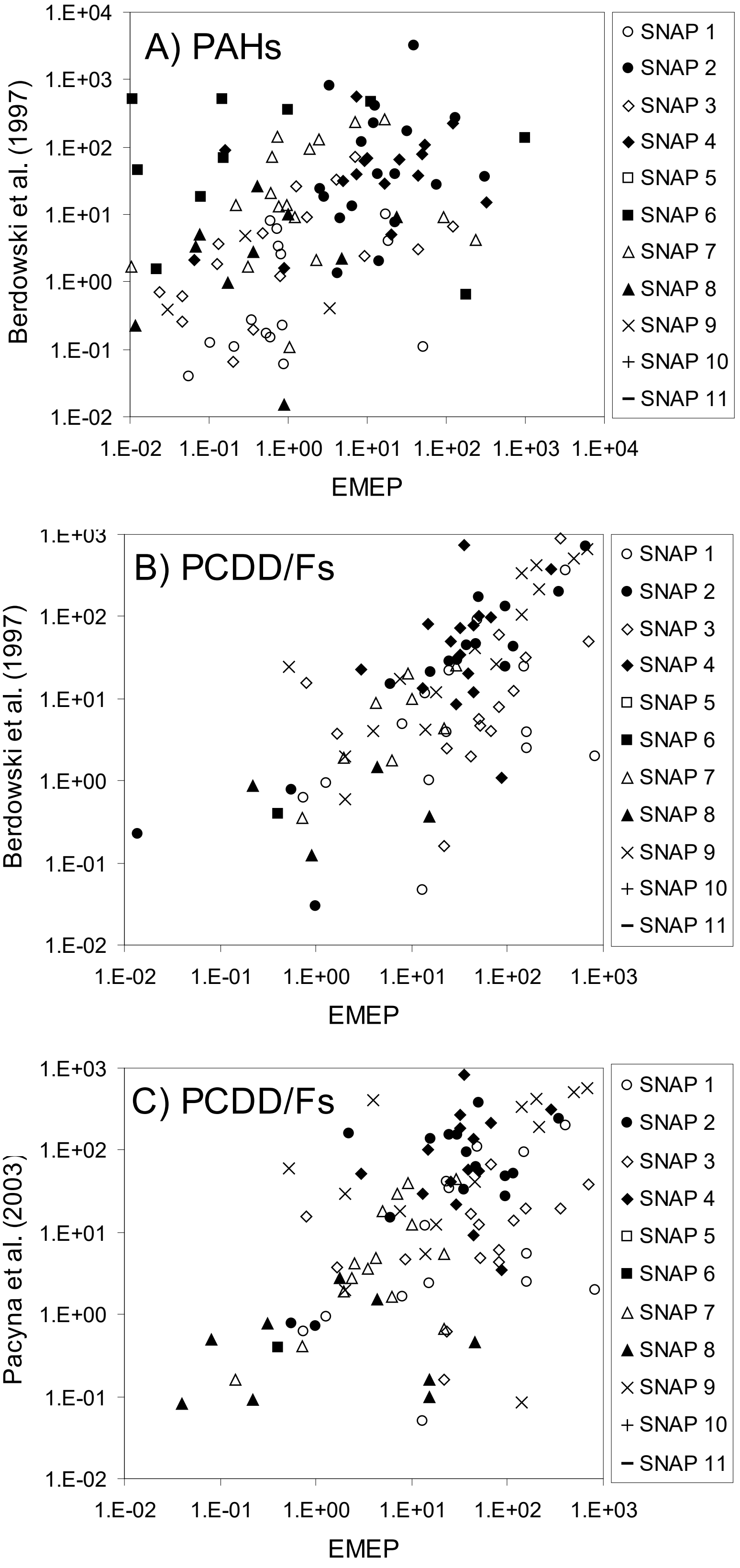

A) PAHs
Tonnes / Year
10000.0
1000.0
100.0
10.0
1.0
0.1
NL CZ BG GB BE ES PL HU IT FR SK SE SI RU AT FI HR NO DK MD DE EE IS
B) PCDD/Fs
□ Berdowski et al. (1997)
■ EMEP
▨ Pacyna et al. (2003)
g I-TEQ / Year
10000
1000
100
10
1
0.1
FR CZ GB DE RU NL BE BG PL IT CH SK ES AT HU NO SE LU FI IS DK
C) PCBs
Kg / Year
100000.0
10000.0
1000.0
100.0
10.0
1.0
DE GB PL CZ SI BG SK HU FR ES NL

Figure 2


A) PAHs
Berdowski et al. (1997)
EMEP
1.E-02
1.E-01
1.E+00
1.E+01
1.E+02
1.E+03
1.E+04
SNAP 1
SNAP 2
SNAP 3
SNAP 4
SNAP 5
SNAP 6
SNAP 7
SNAP 8
SNAP 9
SNAP 10
SNAP 11
B) PCDD/Fs
Berdowski et al. (1997)
EMEP
1.E-02
1.E-01
1.E+00
1.E+01
1.E+02
1.E+03
SNAP 1
SNAP 2
SNAP 3
SNAP 4
SNAP 5
SNAP 6
SNAP 7
SNAP 8
SNAP 9
SNAP 10
SNAP 11
C) PCDD/Fs
Pacyna et al. (2003)
EMEP
1.E-02
1.E-01
1.E+00
1.E+01
1.E+02
1.E+03
SNAP 1
SNAP 2
SNAP 3
SNAP 4
SNAP 5
SNAP 6
SNAP 7
SNAP 8
SNAP 9
SNAP 10
SNAP 11