An overview of potential ongoing sources of polychlorinated naphthalenes (PCNs) to the marine environment of the North East Atlantic (OSPAR) area

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[This information was originally presented, along with proposed actions for OSPAR, as Annex 1 to paper HSC 04/6/26-E(L) to the Hazardous Substances Committee of the OSPAR Commission (for the Protection of the Marine Environment of the North East Atlantic) in April 2004]

Summary

In 2003, the Hazardous Substances Committee (HSC) of the OSPAR Commission reviewed the status of OSPAR "Priority Chemicals with No Production and Use Interest" (HSC 03/3/16). Among these substances were the polychlorinated naphthalenes (PCNs). In this context, it is important to note that absence of production and use interest must not be taken to imply that there are no significant ongoing sources of these chemicals to the marine environment.

A review of available information identified five potential sources of PCNs to the wider environment worthy of further investigation:-

- diffuse losses from old equipment/products in which PCNs were deliberately incorporated
- diffuse losses of PCNs present as contaminants in PCB formulations
- discharges and losses from the chloralkali sector, at least in historical terms
- ongoing/periodic illegal sales and use of PCN stockpiles
- ongoing formation and release of PCNs, among other persistent chlorinated pollutants, as unintentional by-products of high temperature industrial processes including incineration

Given i) the nature of the hazards presented by PCNs, ii) the specific concerns PCNs already present within the marine environment, including contamination of human food resources, and iii) the great uncertainty and data gaps which still surround existing information regarding sources and environmental fates of these substances, further work to determine the relevance and scale of these potential sources of PCNs, and to identify possible control measures, is urgently required.

1. Introduction: history of PCN production and use

1.1 Polychlorinated naphthalenes (PCNs) were first synthesised in the early part of the 19th Century, though their main commercial production and use was not realised until the first half of the 20th Century (Falandysz *et al*. 1996a). No definitive values exist for total quantities produced, though various estimates put the likely total in the region of 150 000 tonnes (Jarnberg *et al.* 1997, Falandysz 1998), manufactured primarily in the period from the 1920s to the 1950s. Many commercial formulations were available, all comprising a complex mixture of various of the total theoretical number of 75 possible congeners.

1.2 PCNs were used in a diverse array of industrial applications, including as transformer and capacitor fluids (predating the PCBs), as flame retardants, plastic and rubber additives, oil additives, fungicides, sealants and in finishes for certain textiles and papers (Marti & Ventura 1997). Other identified uses include dye carrying agents, use in the electroplating industry and as calibration solvents for refractive index determinations (Lundgren *et al.* 2003). Falandysz (1998) provides a more detailed list of recorded historic uses.

1.3 Deliberate production and use of PCNs ceased in the US (one of the main producers) in 1977, but continued for several years more in certain parts of Europe. PCNs have also long been recognised as low-level but significant contaminants in commercial PCB formulations (Falandysz 1998, Yamashita *et al.* 2000), thought to be the predominant source of PCNs to the environment in some regions.

2. PCNs as widespread environmental contaminants

2.1 It has been acknowledged for some time that PCNs, in common with many other persistent organochlorine chemical groups, have become widely distributed through various environmental compartments, including in marine systems. Until the mid 1990s, however, limitations to analytical determination of these contaminants was a barrier to more detailed description of their environmental distribution and fate, and hence to the identification of likely sources and their relative contributions (Falandysz *et al.* 1996a). Since the development of reliable congener-specific analytical techniques, however, the occurrence of PCNs as contaminants in air, water, soil, sediments and biota has been increasingly well described.

2.2 Much of the work in the early to mid 1990s focused on the Baltic Sea and its catchment, especially the Gulf of Bothnia to the north and the Gdansk Basin to the south. In the Gdansk region, for example, between 44 and 48 different PCN congeners were reported in sediments and biota (Falandysz *et al.*1996a), including in fish and seabirds (Falandysz *et al.* 1996b, 1998). In the case of flounder (*Platichthys flesus*, Falandysz *et al.* 1997a) and the black cormorant (*Falacrocorax carbo sinsensis*, Falandysz *et al.* 1997b), there were indications of biomagnification of some of the more toxic PCN congeners when compared to levels in shellfish. Further north, in the Gulf of Bothnia and in the Baltic proper, Jarnberg *et al.* (1997) described the presence of PCNs in marine sediments and fish from various locations, as well as identifying significant point sources of PCNs to certain Swedish lakes. More recent work by Lundgren *et al*. (2003) has suggested that a consistent congener pattern of PCN contamination exists across much of the Baltic Sea, although absolute concentrations vary and there are some important point sources to coastal regions. A useful review of many of the early studies in the Baltic is provided by Falandysz (1998). Based on measurements of settling particulate matter (SPM) made in the early 1990s, Lundgren *et al.* (2003) estimate total loads of PCNs to the Gulf of Bothnia to be in the order of 91 kg/y.

2.3 PCNs have also been recognised as significant contaminants, related to both point and diffuse sources, in other regions, although the total number of studies available and, hence, the geographical range they represent remain markedly limited when compared to data sets for contaminants such as PCBs and PCDD/Fs. Eljarrat *et al.* (1999) recorded PCNs in sediments from the Venice lagoon, potentially arising from a number of industrial point sources in the region. Both Kannan *et al.* (1998, 1999) and Brack *et al.* (2003) have described localised contamination of sediments downstream from specific industrial sources, namely old chloralkali facilities, in the USA and in the Bitterfeld region of Germany respectively (see below).

2.4 Moreover, in addition to these "hotspots" of contamination, PCNs are capable of undergoing long-range transport to remote regions. Their presence as contaminants in air from urban areas or downwind of industrial facilities has been recognised for some time (Dörr *et al.* 1996, Harner & Bidleman 1997), in patterns reflected also in soil contamination (Krauss & Wilcke 2003). However, it is only more recently that PCNs have been described amongst other persistent organic contaminants in top predators from both Arctic (Helm *et al.* 2002) and Antarctic (Corsolini *et al.* 2002) regions, where transfer from mother to eggs of both skuas and penguins have been recorded. Very recently, Lichota *et al.* (2004) have reported the presence of PCN congeners in the tissues of threatened high altitude herbivores (marmots) on Vancouver Island.

2.5 PCNs are commonly recognised to be lipophillic in nature, explaining their presence and even accumulation in the tissues of a variety of marine and freshwater organisms. They also tend to associate with sediments and other settling particulate matter, especially in the case of the more heavily chlorinated congeners. Nevertheless, they are capable also of significant contamination of (and presumably, therefore, transport through) water, as demonstrated by the findings of up to 79 ug/l (ppb) total PCNs in the aquifer of the Llobregat River, near Barcelona (Marti & Ventura 1997). As far as we have been able to establish, no similar studies exist for other parts of Europe.

2.6 Similarly, few of the available data sets provide reliable indications of trends in PCN inputs and concentrations. One of the few available studies, that using sediment profiles collected from Esthwaite water (in the Lake District, UK) indicates a subsurface peak of PCN contamination corresponding to the period of major manufacture and use in the first half of the 20th Century (Gevao *et al.* 2000). A similar trend was reported by Horii *et al.* (2002) for Lake Kitaura in Japan. Levels in marine biota also show some evidence of decline (Falandysz 1998), as do levels in human tissues from parts of Europe (Weistrand *et al.*1997, Weistrand & Noren 1998, Lunden & Noren 1998).

2.7 Given the reported phase-out of production and use of PCNs almost two decades ago, and the parallel phase-out of PCB use, such declining trends come as no surprise. Indeed, they are mirrored by declines in PCBs themselves, as well as of other persistent organic pollutants now subject to strict controls. Nevertheless, while these trends give some cause for optimism, they must not be taken as an indication that there are no outstanding releases to the environment. Rather it is important that attention is given to a number of identified potential ongoing sources of PCNs and their possible significance in contributing to current environmental levels (both in absolute concentrations and contributions to dioxin-like toxicity).

2.8 In short, whereas it is reasonable to conclude that there are no major commercial interests in the production and use of PCNs in the OSPAR (North East Atlantic) region, several point and diffuse sources have been identified in recent years which could have significance for environmental PCN levels within the OSPAR region and which, therefore, deserve further investigation. Although a substantial proportion of existing levels and fluxes may arise simply as a legacy of past contamination, and as such is not amenable to further action to control releases, it is important that OSPAR recognise and evaluate the significance of those potential ongoing sources which may well be amenable to further controls.

2.9 From a review of the available literature, five such potential sources can be identified; these are discussed in turn in sections 3 to 7 below.

3. Losses from old products/equipment containing PCNs

3.1 As noted above, until the late 1970s or early 80s, PCNs were used for a wide range of industrial and consumer-related applications (Falandysz 1998). While much of the equipment and material in which they were used may since have entered the waste stream, the scale and diversity of reported historic uses makes any judgement regarding quantities of PCNs still in products in use highly uncertain, based on the very limited existing knowledge. It is known that some PCN-containing electrical equipment was still in use in Europe as late as the start of the 1990s (Weistrand *et al.* 1992), though it is not possible to determine whether this reflected more general usage or, indeed, whether the situation is substantially different now. Furthermore, there is some evidence to suggest that PCNs may be capable of leaching from wastes already in landfills (Jarnberg *et al.* 1997), though the overall significance of this process to current environmental levels is simply not known.

3.2 Evaporative losses of PCNs from materials and equipment in use is a widely recognised phenomenon (see e.g. Yamashita *et al.* 2003) such that, if such articles were still in significant usage, diffuse losses to atmosphere and other media would be expected. Indeed, congener profiles of PCNs measurable in urban air samples and to some extent in settling particulate matter (SPM) in the aquatic environment still resemble those of commonly used commercial formulations (Helm & Bidleman 2003, Ishaq *et al.* 2003). Although some authors suggest that these levels arise primarily from environmental cycling of the existing contaminant load (Dörr *et al.* 1996), the possibility of contributions from equipment and/or materials in continued use cannot be ruled out.

4. Losses as contaminants in PCB formulations

4.1 PCNs generally represent a minor component of commercial PCB mixtures (up to 870 ppm in those Aroclor and Clophen formulations analysed, with a median of 67 ppm, Falandysz 1998), though levels in Delor, a product formulated in the Czech Republic, are thought to be relatively high (82-445 ppm, Taniyasu *et al.* 2003). Nevertheless, given the high tonnage of PCBs produced worldwide (estimated at 1 500 000 tonnes), the total quantity of PCNs theoretically available for release to the environment stands at tens if not hundreds of tonnes (Falandysz 1998). Yamashita *et al.* (2000) estimated around 169 tonnes of PCNs could be contained within PCB formulations worldwide. Based on the analyses of Taniyasu *et al.* (2003), as much as 3.68 tonnes of this could arise from the Delor formulation alone.

4.2 Although concerns regarding possible ongoing releases of PCNs from PCB formulations, held either in existing equipment or in waste deposits, are largely theoretical, the existence of a second PCN maximum in undisturbed sediments from Esthwaite Water in the UK corresponding to the peak in production and use of PCBs attests to the feasibility of this mechanism and the need, at least, for further investigation of its significance.

5. Discharges and losses from the chloralkali sector

5.1 The chloralkali industry, at least in the form of certain specific facilities, has been recognised as a substantial point source of PCN contamination for many years. PCN contamination has been recorded in the vicinity of chloralkali plants in Sweden (Jarnberg *et al.* 1997), Germany (Brack *et al.* 2003) and the USA (Kanan *et al.* 1998), with maximum

levels in contaminated sediments reaching 23 ppm. Jarnberg *et al.* (1997) recorded levels of 150 ppb in graphite electrode sludge arising directly from the Swedish facility.

5.2 There appears to be some disagreement regarding the specific origins of PCNs arising in sludge wastes from the choralkali sector. Some authors suggest historic uses as solvents or "binders" in the manufacture of the graphite electrodes themselves are responsible, while other suggest that *de novo* synthesis at the graphite surface during electrolysis may also contribute (Kanan *et al.* 1998).

5.3 It is not known whether any of the existing or decommissioned chloralkali plants within the OSPAR area have been or remain potential sources of PCNs to local environmental compartments and further afield, though this is clearly an issue deserving further attention.

6. Possible ongoing illegal sale and use of PCNs

6.1 Recent reports from Japan have suggested that PCN formulations may have been imported very recently from suppliers in North America and Europe as recently as the late 1990s (Yamashita *et al.* 2003, Falandysz 2003). Two separate incidents have been the subject of investigation in Japan, involving documented imports from Canada and the UK respectively. It is not known whether these reported illegal activities represent isolated incidents or hint at more widespread illegal trade and use of PCN stockpiles. Nevertheless, given the potential scale of inputs which could result, it is vital that this is investigated further.

7. Generation of PCNs as unintentional by-products of thermal processes

7.1 It has been recognised for some time that PCNs are found among the many chlorinated by-products generated by incineration and various other high temperature processes (see e.g. Oehme *et al.* 1987). Studies from the mid 1990s, conducted with the benefit of improved analytical methodologies, confirmed these earlier finding and clearly identified municipal solid waste (MSW) incineration (Imagawa & Yamashita 1994, Sakai *et al.* 1996, Abad *et al.* 1997, Schneider *et al*. 1998), copper ore roasting (Thiesen *et al.* 1993) and smelting of aluminium scrap (Aittola *et al.* 1994) as significant industrial sources of PCNs *via* exhaust gases and fly ash. Falandysz (1998) provides a useful overview of these early studies of combustion sources and estimates that somewhere in the order of 1-10 tonnes of PCNs may have been generated by thermal processes of this nature during the last Century.

7.2 More recent studies by Iino *et al.* (1999, 2001), Imagawa & Lee (2001) and Weber *et al.* (2001) have further elucidated the mechanisms which give rise to PCNs in combustion processes. The developing consensus is that PCNs are predominantly formed through post-combustion chlorination of naphthalene and other PAHs, especially on the active surfaces of fly ash particles. In addition, one recent laboratory-based study has indicated the potential significance of the chlorinated plastic PVC (polyvinyl chloride), a common component of municipal and hospital waste streams directed to incineration, as a source of PCN formation (Wang *et al.* 2003).

7.3 Although the extent to which emissions from thermal processes have contributed to overall PCN levels in the environment is still in question (see e.g. Dörr *et al.* 1996), a number of pieces of evidence do point to the importance of such sources, particularly in relation to

distributions of some of the more bioaccumulative and toxic PCN congeners. For example, thermal processes are the only known source of 2,3,6,7-substituted congeners (Jarnberg *et al.* 1999). The tetra-CN congener 44, found in black cormorants (Falandysz & Rappe 1997), and penta-CN congener 54, found in white-tailed sea eagles (*Haliaeetus albicilla*, Falandysz *et al.* 1996b), are thought to arise from combustion sources alone. Although perhaps only a minor component of total PCN concentrations in the wider environment, they are thought to be among the more toxic and bioaccumulative of the CN congeners.

7.4 In addition, while congener profiles for urban air show similarities to those of commercial PCN formulations, at more rural locations, airborne profiles suggest proportionately greater contributions from combustion sources (Krauss & Wilcke 2003). Helm & Bidleman (2003) estimate that around half the levels of PCNs determined in the vicinity of specific industrial sources could arise from thermal processes.

7.5 In short, although it is almost certain that overall environmental burdens of PCNs will have arisen (and may still be arising) from historic manufacture and use of PCN and PCB formulations, formation as by-products of thermal processes undoubtedly also contribute to this burden. The extent of this contribution on a global basis, and more specifically within the OSPAR region, is currently unknown but is certainly worthy of further investigation.

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