Short-term Changes in the PCB Concentrations of Rural UK Air

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Abstract

Daily (24 hour) air samples were collected on 161 occasions during March-October and December 1994 at a rural site in northwest England, during which time it received air masses from a variety or directions. ∑PCB concentrations only ranged over a factor of 7, between 54-375 pg m^{-3}. Generally, air masses that originated to the north of the UK contained lower concentrations than those originating to the south or west; masses which had recently passed over land also tended to contain higher PCB concentrations. Several factors combine together to influence the daily air concentrations at the site, including temperature, marine and land mass source area and presumably the frequency of deposition events. Intensive sampling of air during several days in August 1995 showed temperature-driven diurnal cycling of PCBs during a period of stable high pressure. The data from these studies are interpreted as evidence that environmental re-cycling (i.e. secondary sources) now has the dominant influence on ambient concentrations of PCBs.

Introduction

Atmospheric transport is the principal pathway for regional and global movement of semi-volatile organic compounds (SOCs). Because of the current interest in the global distribution of these compounds, studies have been undertaken to measure the ambient levels of PCBs, but few have attempted to correlate the concentrations measured with meteorological parameters and to identify the origins of the air masses which predominated at the time of sampling. These studies require a balance between the practical or financial constraints on sample numbers and the analytical detection limit constraints of the short sampling intervals required to monitor specific meteorological conditions and air masses. In other words, if the sampling interval is too long the development of meaningful correlations between meteorological parameters and air concentrations becomes difficult.

In the UK since 1992 the Toxic Organic Micro-Pollutants (TOMPS) survey has established the ambient levels of PCBs, PAHs and PCDD/Fs at urban and rural sites, by sampling every 2 weeks throughout the year. However, this sampling regime is inappropriate to demonstrate the influence of meteorological conditions or air mass origins. Consequently this study was undertaken with sampling every 24 hours from March to October 1994 and in December 1994, when different meteorological conditions and air masses were experienced. In addition, a further period of intensive air sampling was undertaken in August 1995, during which air samples were collected every 6 hours, to study the possible effect of temperature on PCB air concentrations.

Experimental Section

The sampling site is located at a meteorological station located in a rural area outside Lancaster, a town of ca. 60,000 inhabitants on the north west coast of England, approximately 5 km from the Irish Sea (54° N, 3° W). An air sampler, set to aspirate approximately 350 m^3 day^{-1} for the daily samples, trapped the particulate fraction on a Whatman glass microfibre filter and the vapour phase on a polyurethane foam plug. The GFF and PP were combined and soxhlet extracted; a range of congeners were quantified by GC-ECD (B/Z numbers 30, 18, 28, 52, 104, 40, 61/74, 66, 101, 77/110, 82/151, 149, 118, 188, 153, 105, 138, 187, 183, 128, 185, 180, 170, 198, 201 and

194/205).

Results and Discussion

Typical concentrations

Table 1 presents a summary of the data for selected PCBs. ∑PCB concentrations varied between 54 and 375 pg m^{-3}, with the lighter congeners 28 and 52 etc making major contributions to the total. Comparison with data from elsewhere in Europe and North America supports the view that this site is rural; air in UK city centres typically contains annual average ∑PCB concentrations of ~ 200-3700 pg m^{-3} (1). There is a strong marine influence on many of the air systems which are sampled at Lancaster. PCB concentrations in oceanic air have been reported by others, with the range of values detected at the site again in line with expectations based on these studies. In summary, the site can be viewed as rural with a maritime influence.

Air masses passing over the site have often spent previous days out to the west, passing over the Atlantic Ocean, Ireland and the Irish Sea. However, air masses frequently originate from other areas; during this study air masses from the north, south and east (i.e. the Arctic, northern Europe, southern Europe and the UK) were all recorded.

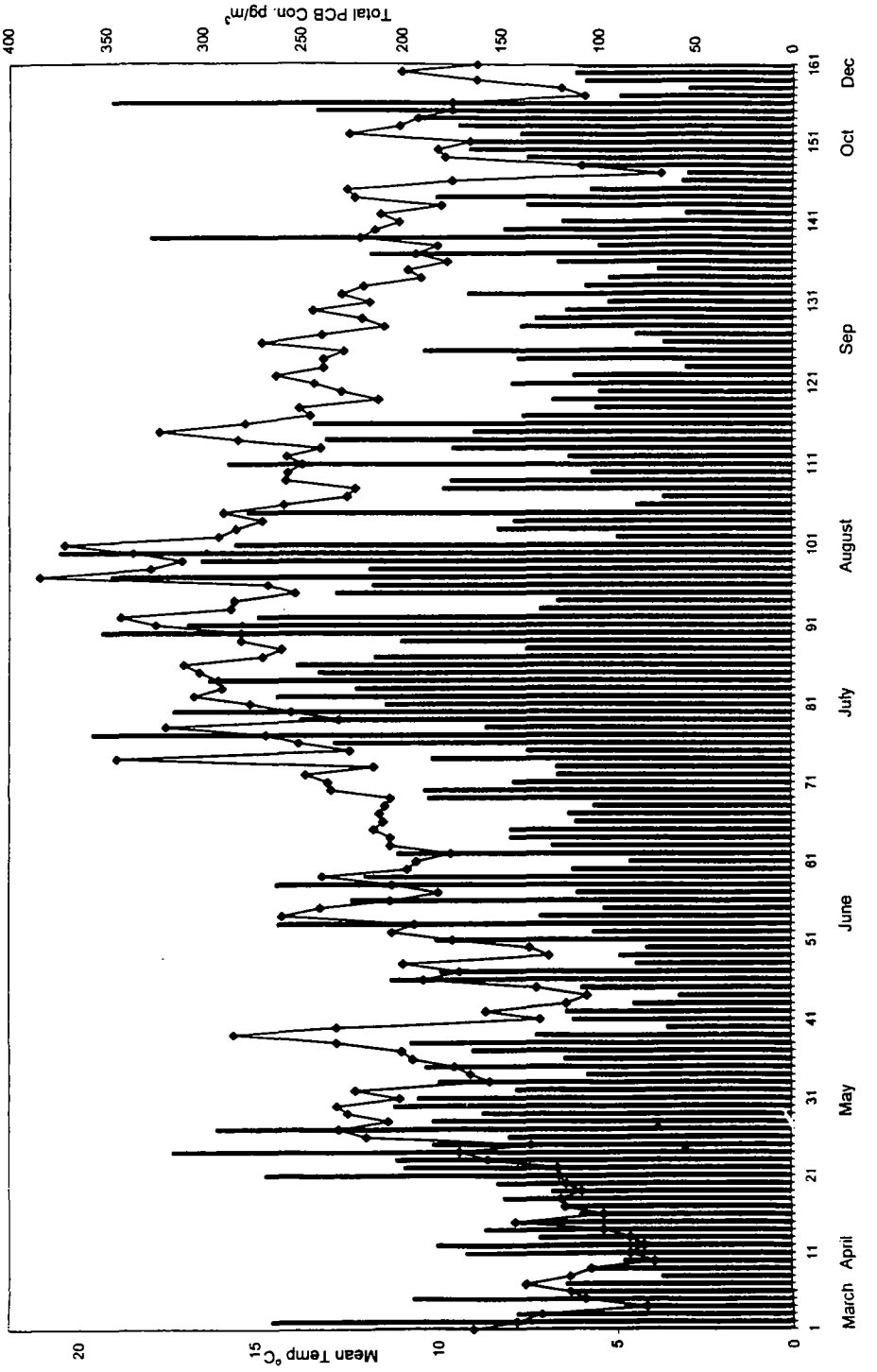
Table 1: The mean and range of selected PCB (pg m^{-3}) concentrations during the sampling period.

Compound/homolog	Mean	Maximum	Minimum
PCB-28	24.7	70.4	<10
PCB-52	18.4	59.5	<2.8
PCB-101	6.5	16.0	<2.0
PCB-153	1.7	11.7	<1.5
PCB-138	1.5	5.7	<1.2
PCB-180	11.4	41.1	<2.2
Tri-PCBs	65	180	<1.5
Tetra-PCBs	49	139	6.5
Penta-PCBs	22	66	5.8
Hexa-PCBs	6.3	28	<1.2
Hepta-PCBs	16	89	0.4
Octa-PCBs	4.1	57	<1.0
∑PCB	164	375	54

Seasonal variations

Broad seasonal variations in PCB concentrations were noted during the study, broadly corresponding to seasonal temperature changes, with summer concentrations tending to be higher than winter ones. This has been noted by other researchers. In this study the total daily concentrations remained quite stable between March and mid-June, then generally rose as the mean temperature increased until early August. Thereafter, the mean concentration and temperature decreased to levels detected before mid-July (see Figure 1). In summary, PCB concentrations were quite stable throughout the study period, ranging only by a factor of ~7 between March and October. This observation, and generally higher concentrations during warmer periods, support the view that volatilisation/environmental re-cycling has a dominant influence on the air concentrations of PCBs in rural areas; PCBs are 'old' contaminants with few new point source inputs to the environment. The generally quite uniform concentrations between air masses (see below) point to PCBs now being quite uniformily distributed through the northern temperate latitudes where they were produced and widely used in the past.

Figure 1: Daily PCB concentrations throughout the study period.



Relationships between concentrations and meteorological parameters

Meteorological data for several variables was recorded at the site, namely; mean, maximum and minimum ambient temperatures; relative humidity; rainfall; wind speed and direction; atmospheric pressure. Correlation coefficients were then determined when compound concentration was plotted against these variables.

As mentioned above, PCBs were positively correlated against air temperature; correlations were generally significant at the $P \leq 0.01$ level for individual congeners. It made no systematic difference to the strength of the correlation whether the mean, maximum or minimum daily temperature value was used. None of the other meteorological parameters regularly gave a significant correlation with compound concentrations (although several of the more volatile compounds e.g. PCB-52 were inversely correlated with windspeed). Given the lack of a clear meteorological influence on the daily concentration of PCBs and PAHs, the influence of the source and area over which a particular air mass had moved was therefore investigated.

Back trajectories and air masses

The majority of the weather systems which influence the UK originate to the west (south west-north west). On a few other occasions during the study period air was drawn directly from the north, south or east. These episodes allowed air to be sampled which had very different origins from the air usually reaching the study site, namely, masses which had originated in the Arctic, central/northern Europe and the Mediterranean. Trajectories were therefore calculated for those samples/days which had the highest and lowest concentrations and with interesting meteorological conditions. The choice focussed on the 10 highest and 10 lowest concentration events. Back trajectories were calculated based on geostrophic wind speed calculated every 6 hours from the isobars/pressure readings given on synoptic charts, assuming the geostrophic wind is parallel to the isobars. The movement of air mass parcels 4-5 days prior to their arrival at Lancaster was traced; it was assumed that air concentrations arriving at Lancaster would be a function of events/processes occurring during this time period.

Illustrative air masses from the north and south west of the UK

Figure 2 shows the air mass back trajectories derived for a number of events which had low or high air PCB concentrations at Lancaster. All the lower concentration events originated in regions to the north of the UK, at latitudes >60° north (and usually >65° north). The Figure also shows source areas of events in April and July when high PCB concentrations (200-315 pg $\sum$PCB m^{-3}) were recorded. At these times the air had moved from the west and south of the UK, starting at <60° north and usually <55° north. These 'northern low PCB' and 'south west high PCB' episodes could be separated by several months, yet the concentrations were very similar. Air from the north in August and December events, for example, showed little difference in the PCB concentrations.

Diurnal cycling

Figure 3 shows the short-term (6 hour) variations in PCB concentrations during a few days in August 1995. At this time, a stable high pressure event occurred over the UK for the first 5-6 days, followed by more turbulence (windier conditions) at the end of the study period. Under the stable air conditions there was a clear diurnal cycling of PCB concentrations, with the cyclic amplitude being greater for the lighter (e.g. 28) than the heavier (e.g. 138) congeners. Air concentrations were higher during the warm days than the cooler nights. Under the turbulent conditions the cyclicity disappeared. The high pressure event had originated to the east and north of Lancaster, thereby passing over land for sometime prior to arriving at the sample site.

The diurnal cycling of PCBs and other OCs has been noted previously by others and provides powerful evidence for the influence of re-cycling of historically emitted (i.e. volatilisation) as a major controlling factor on the contemporary ambient concentrations of PCBs.

Literature cited

1. Halsall, C. J.; Lee, R. G. M.; Coleman, P. J.; Burnett, V.; Harding-Jones, P.; Jones, K. C. *Environ. Sci. Technol.* 1995, 29, 2368-2376.

Figure 2: Air mass source areas for selected events during the study period
'Low' air concentrations (54-122 pg m^{-3}) at Lancaster associated with masses all shown originating to the north of the UK; 'high' concentrations (198-374 pg m^{-3}) associated with the source areas to the south.

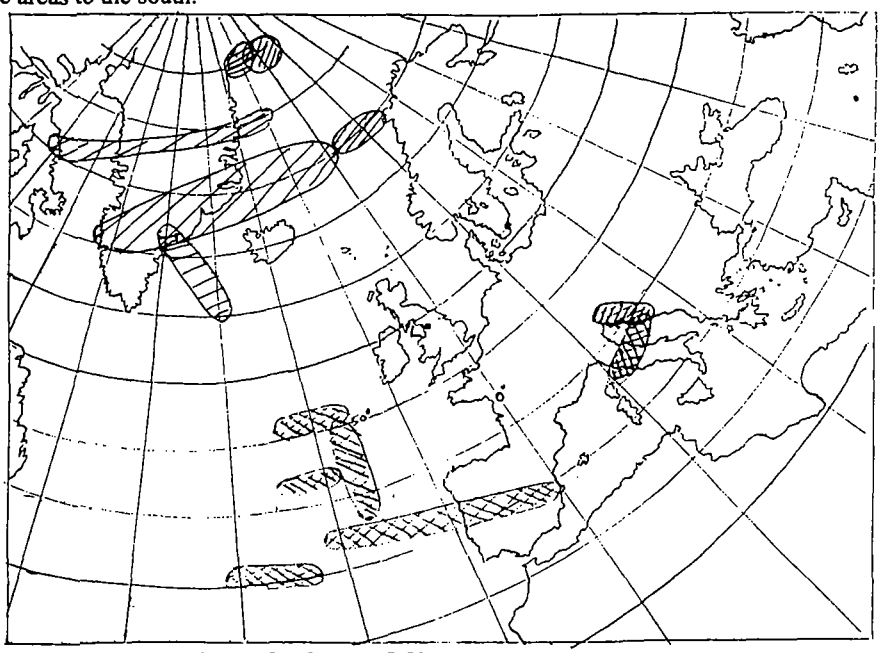


Figure 3: Concentrations of selected PCBs and the ambient temperature during an intensive sampling period in August 1995