DEPOSITION OF ATMOSPHERICALLY TRANSPORTED POLYCHLORINATED BIPHENYLS IN THE CANADIAN ARCTIC

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Since 1986, efforts have continued to quantify the deposition of trace organic contaminants including PCBs to the Canadian Arctic. This has resulted from an increasing concern over the potential for ecosystem and human health effects as a result of the bioaccumulation of these contaminants. PCBs are a concern as most of the 209 congeners, especially those with 5 or more chlorines, are biomagnified (Jensen et al., 1969; Reijnders, 1980; Muir et al., 1988). PCBs have been used world-wide for a variety of purposes with as much as 370 tonnes estimated to have been released into the environment (Tanabe, 1988).

It is believed that the source of these contaminants in the Arctic is primarily Eurasia. The general atmospheric circulation during the winter season, driven by the strong Siberian anticyclone, flows mainly from the Eurasian continent into the Arctic and then out over the North American continent or into major cyclonic regions in the Aleutians and off southern Greenland. In the summer, south-to north transport is replaced by a weak north-to-south transport as the Siberian high dissipates and flow into the arctic from the north Pacific and the north Atlantic is more frequent. Consequently, PCBs emitted north of the polar front in Eurasia during the winter season, may be transported quickly to the Arctic, where both wet and dry scavenging occurs. The cold, dry air minimizes wet scavenging as we know it in temperate, more humid regions. But, the stability of the air masses, and the long calm periods with ice crystal deposition interspersed with snowfalls, are likely effective scavenging mechanisms. Measurements of contaminants in snow, therefore, is a good indication of the flux of PCBs to the Arctic.

The median winter concentration of Total PCBs in snow at sites in the Canadian arctic ranging from approximately 63°N to 81°N, was 745 pg l^{-1} with a range of 20 to 1760 pg l^{-1}. The median winter deposition rate was 70 $\mu g\ m^{-2}$. The winter deposition rates for the Arctic are at least one order of magnitude less than the annual deposition rates measured in precipitation for southern Canada.

Beginning in the fall of 1990, large volume snow collectors were designed and installed at two High Arctic weather stations - Mould Bay (76°15'N, 119°16'W) and Eureka (80°00'N, 86°36'W). These collectors, with a surface area of approximately 4.5 m^2, were intended to intercept ice crystals and falling snow during calm periods but will also retain some quantity of wind blown snow due to the screening that is installed around the perimeter of the collector. Samples were collected once a week, weather conditions permitting. These samples were then stored on site until late

winter when they were returned to Resolute Bay, NWT and extracted with dichloromethane using a continuous flow extraction apparatus. The sample extracts were subsequently returned to the analytical laboratory in Burlington, Ontario for cleanup, fractionation and analyses.

Due to very low snow accumulation rates, especially at Eureka, a number of very small samples were pooled to provide a larger sample volume. This yielded only four samples for the season for Eureka and a total of nine samples for Mould Bay for the 1990/91 winter season. This experience has shown that the concentrations for most of the compounds of interest were higher in this fresh snow than expected and thus for subsequent sampling, pooling will be minimized. At both sites, triplicate, end of winter snowpack samples were collected approximately at the beginning of May for purpose of comparison with the snow collector samples. Emphasis here will be upon the Mould Bay data as it is the most complete.

While fluctuations occur, there appears to be a general increase in the concentration of the lighter congener groups (two, three and four chlorines) toward the end of the winter season at Mould Bay while the concentration of heavier congeners tend to be variable. PCB concentrations (sum of the 51 congeners) throughout the sampling season ranged from approximately 8 ng l^{-1} to more than 20 ng l^{-1}. Daily deposition rates of PCBs were more uniform at about 2 ng m^{-2} day^{-1} except for a period in the middle of January, 1991 when deposition of total PCBs increased to nearly 11 ng m^{-2} day^{-1}. This increase was primarily made up of the two, three and four chlorinated congener groups. This sample that showed the high deposition rates, represents the period of January 3 to January 8, 1991 during which there was a strong wind for several days followed by a period of calm to light winds. A total of 1.4 cm of snow fell on January 7 and 8. Assuming 100 % efficiency for the collector, this would account for only about 8 *l* of water as oppossed to the 25 *l* collected. This over-sampling of snow likely indicates a certain amount of collection of wind-blown snow during the high wind event as well as an over-collection of the fresh snow. However, the high concentrations can only be explained by the accumulation of fresh snow as wind blown snow would show concentrations similar to earlier samples which were much lower. Thus this one event is interpreted to typify a major deposition event to the Arctic.

Previous studies of selected organochlorine pesticides in arctic snow have indicated that compounds with relatively high vapor pressures tend to be released to the atmosphere from the snowpack as temperatures rise. This has been observed in a number of situations, especially for the hexachlorocyclohexanes (HCHs) (Barrie et al., in press) and has been found to be true for the Mould Bay site as well with between 20 and 30 % of HCH being lost

from the total amount accumulated in the snowpack in comparison to the total accumulated in the collector. The significance of this loss is further emphasized by the fact that the snow collector operated from late October, 1990 to April 1, 1991 whereas the snowpack sample was not collected until May, 1991, which would have allowed time for additional deposition.

In contrast to the HCHs, the PCB data for the collector compared to the snowpack, indicate that the PCB remains in the snowpack. Indeed, the total accumulation of PCB in the snowpack was 2.5 times greater than that accumulated by the collector. This apparently occurred during the month of April when the collector did not operate and supports an earlier observation that the deposition of lower chlorinated congeners was increasing toward the end of the collector sampling season. The distribution of mean concentrations for all snow collector samples for each congener group paralled the distribution for the snowpack. However, the concentrations in the collector samples for each congener group were generally 1.5 times higher than for the snowpack. This may support a revolatilization loss from the snowpack samples which has been suggested in other arctic studies to amount to about 30 % of the total deposition.

The cumulative deposition of total PCB to the collector was estimated to be 200 ng m^{-2} whereas more than 900 ng m^{-2} were deposited in the annual snowpack. The deposition difference between the two measurements was most pronounced for the di, tri, tetra and penta congener groups indicating that a substantial amount of these lower chlorinated compounds were deposited late in the winter season after the collector had been shut down.

PCBs have been shown to be between 80 and 100% in the vapor phase in urban atmospheres and in the area of the Laurentian Great Lakes (Murphy and Rzeszutko, 1977; Doskey and Andren, 1981; Eisenreich et al., 1981). Duinker and Bouchertall (1989) investigated the partitioning of several PCB congeners and concluded that while only a very small fraction of total PCB was found in aerosols, this compartment accounted for more than 99% of the PCBs in rain. More highly chlorinated congeners were particle-associated while the lower chlorinated congeners remained essentially in the vapor phase. Thus it can be speculated that for the Arctic winter, ice crystals, fresh snowfall and some unknown component of dry deposition scavenge rather effectively the full range of PCB congeners. It is likely that the lower chlorinated congeners are adsorbed to the ice crystal or snow flake whereas the higher chlorinated congeners would be bound to the particulate matter forming the nucleus. Consequently, during snow metamorphosis, it is very conceivable that the lighter congeners are more readily revolatilized from the snow pack than the heavier congeners.

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