Influence of the Pacific North American (PNA) Pattern on the Long Range Transport of Persistent Organic Pollutants (POPs) to the Canadian Arctic

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Introduction

Since the early 1970's, the usage and production of many persistent organic pollutants (POPs) have been banned or severely restricted in highly industrialized countries of Europe and North America. They are now subject to global restrictions under the Stockholm Convention on POPs. While fresh emission of these chemicals diminishes, secondary emission due to re-volatilization from historical residue, such as previously contaminated soil and aquatic surfaces, may impact atmospheric concentrations of POPs for many years to come. Under conditions of climatic variations, atmospheric circulation patterns may play an increasingly important role in the re-emission, and the subsequent movement and distribution of POPs in the environment. However, very few studies have been conducted to assess the impact of climate variations on the long-range transport of POPs. In this study, seasonally averaged air concentrations of α- and γ-hexachlorocyclohexane (α- and γ-HCH) and various polychlorinated biphenyl (PCB) congeners measured from 1993 to 1999 at the Canadian High Arctic site of Alert (82°30'N, 62°20'W) were compared over the same time period to North American surface air temperatures and indices that reflect the strength of the Pacific North American (PNA) pattern, an atmospheric circulation teleconnection pattern strongly associated with climate variations and air flow patterns in North America. The potential influence of climate variations occurring in temperate North America on the long-range transport of POPs to the Canadian Arctic is explored.

Materials and Methods

Arctic air concentrations of POPs: Under the Canadian-operated Northern Contaminants Program (NCP), weekly air samples were collected using a high volume air sampler at Alert since 1992. Sampler operation, sample extraction, cleanup and analysis have been described elsewhere.[1] Briefly, ~13000 m^3 of air was aspirated through a glass fiber filter and two polyurethane foam plugs (PUFs) to collect operationally defined particle and vapour fractions, respectively. All weekly samples were analyzed individually for PCBs, organochlorine compounds and polycyclic aromatic hydrocarbons (PAHs). Only vapour phase concentrations (pg/m^3) of α-HCH, γ-HCH and PCBs were used in this study since concentrations of these compounds in the particle-phase routinely fell below detection limits.[2,3] Compounds selected for analysis are α-HCH, γ-HCH and 9

PCB congeners which constitutes the AMAP (Arctic Monitoring and Assessment Program) subset (PCB 28, 31, 52, 101, 105, 118, 138, 153 and 180)(excluding PCB 156, since 96 % of all samples were below method detection limit for this congener).

PNA Index: Atmospheric circulation exhibits substantial variability. This variability reflects weather patterns and circulation systems that occur on time scales from a few days to several years. Atmospheric teleconnection pattern (also called "preferred mode") refers to a recurring and persistent, large-scale pattern of pressure and circulation anomalies that spans vast geographical areas. The PNA pattern is one of the most prominent modes of low-frequency variability in the Northern Hemisphere, appearing in all months except June and July. It is characterized by atmospheric flow in which the west coast of North America is out of phase with the Eastern Pacific and Southeast United States. In its positive phase, it is characterized by wave-like flow over the continent with increased temperatures and decreased storminess in the Northwest and cold temperatures in the Southeast. During the negative phase, it is characterized by a stronger zonal (east/west) flow across the US with decreased temperatures and increased precipitation in the Northwest and increased temperatures to the southeast.[4] PNA is represented by its standardized index. The monthly value of the PNA index can be obtained from the Climate Diagnostics Center of the National Oceanic and Atmospheric Administration at http:// www.cdc.noaa.gov /Teleconnections/. The PNA index is defined as: PNA = 0.25 × [Z(20N,160W) - Z(45N,165W) + Z(55N,115W) - Z(30N,85W)], where Z represents standardized 700-hpa geopotential values.[5] The occurrence of the positive and negative phases are indicated by PNA indices greater than and less than zero, respectively.

Correlation Analysis: Relationships between PNA and air concentrations of POP are assessed using annual seasonal averages in winter (December – February), spring (March – May) and Fall (September-November). Since the PNA disappears in June and July, summer is not included in the analysis. Spearman rank-order correlations were used to determine the associations of POP air concentrations at Alert with the PNA index. The Spearman rank-order correlation is a distribution-free, nonparametric measure of association based on the rank of the data values which is exact for small sample sizes and weakens the effect of outliers.[6]

Results and Discussions

No apparent correlations between air concentrations of selected compounds measured at Alert and the PNA index were found in winter and fall. However, in spring, air concentrations of γ-HCH, PCBs 28, 31, 52, 101, 153 and 180 showed statistically significant Spearman correlations with the PNA index at 90 % confidence level ($p<0.10$) (Table 1). Figure 1 shows the linear regression plots of the spring mean gas phase concentrations of γ-HCH, PCBs 101 and 153 versus the PNA index (left panel), as well as their respective time series (1993-1999) (right panel). For those compounds which showed statistically significant correlations with PNA, their air concentrations at Alert increased with increasing PNA index (indicated by positive values of r_S), i.e. towards the positive phase of the PNA. This may be explained by the pronounced differences in climate and atmospheric flow patterns presiding over North America between the positive phase (occurring in 1993, 1994 and 1998) and negative phase (occurring in 1996, 1997 and 1999) of the PNA.

During the positive PNA phase in spring, higher-than-normal surface air temperature (SAT) (or positive SAT anomalies) occurred across Canada, Alaska and the northern U.S.A. Lower-than-normal SAT (or negative SAT anomalies) were observed over roughly the same geographical area during the negative phase of PNA in spring. Since the rate of soil-air exchange is temperature dependent, with higher-than-normal SAT in North America during the positive phase of the PNA, enhanced re-volatilization of organic chemicals from reservoirs, accumulated due to past and present usage, may occur. Among the selected compounds which showed statistically significant correlations with the PNA index, γ-HCH is the only current-use chemical in North America. Lindane, which contains 99+ % of γ-HCH, is commonly used in the Canadian Prairies as a seed treatment for canola[7]. Usage and production of PCBs have been banned in North America since the 1970s. However, previously accumulated residue of PCBs can still be found in soils. Enhanced re-emission of γ-HCH and PCBs from reservoirs during the positive phase of PNA will be reflected by higher air concentrations measured downwind from the source of these substances, similar to those observed in the Canadian Arctic. Therefore, if atmospheric flow associated with the positive phase of PNA favors pole-ward transport, temperate North America is likely a major source region for these compounds to the Arctic region.

To further investigate the possibility of chemical migration from temperate North America to the Arctic, the strength of zonal flows (flows directed along latitudinal lines) at 700 hPa was examined. It was found that weaker than normal zonal flow occurred throughout the U.S.A. and southern Canada during the positive PNA phases in springs between 1993 and 1999. During the negative PNA phase, positive anomaly of the zonal wind at 700 hPa dominates roughly the same region. Weaker-than-normal zonal flow encountered during the positive PNA phases may enhance atmospheric mass exchange between polar and mid-latitude regions, favoring transport of pollutants from temperate North America to the Arctic.

Statistically significant correlations between the PNA index and the springtime air concentrations of γ-HCH and various PCB congeners measured at Alert implied that climate variations in temperate North America may result in enhanced pollutant transport to the Arctic region. Higher-than-normal SAT in temperate North America during the positive PNA phase may promote the re-emission of persistent organic pollutants, such as γ-HCH and PCBs, from previously accumulated reservoirs. These chemicals are then available for atmospheric transport under favorable conditions towards the pole.

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Table 1. Spearman rank order correlations between spring mean air concentrations of selected compounds and PNA indices

	HCHs		PCBs								
[a]	α-HCH	γ-HCH	28	31	52	101	105	118	138	153	180
r_S	0.43	**0.86**	**0.68**	**0.79**	**0.75**	**0.68**	-0.11	0.11	0.64	**0.82**	**0.82**
p	0.34	**0.01**	**0.09**	**0.04**	**0.05**	**0.09**	0.82	0.82	0.12	**0.02**	**0.02**

[a] r_S = Spearman correlation coefficient, p = Spearman p-values
Statistically significant correlations with p<0.10 are shown in bold types.

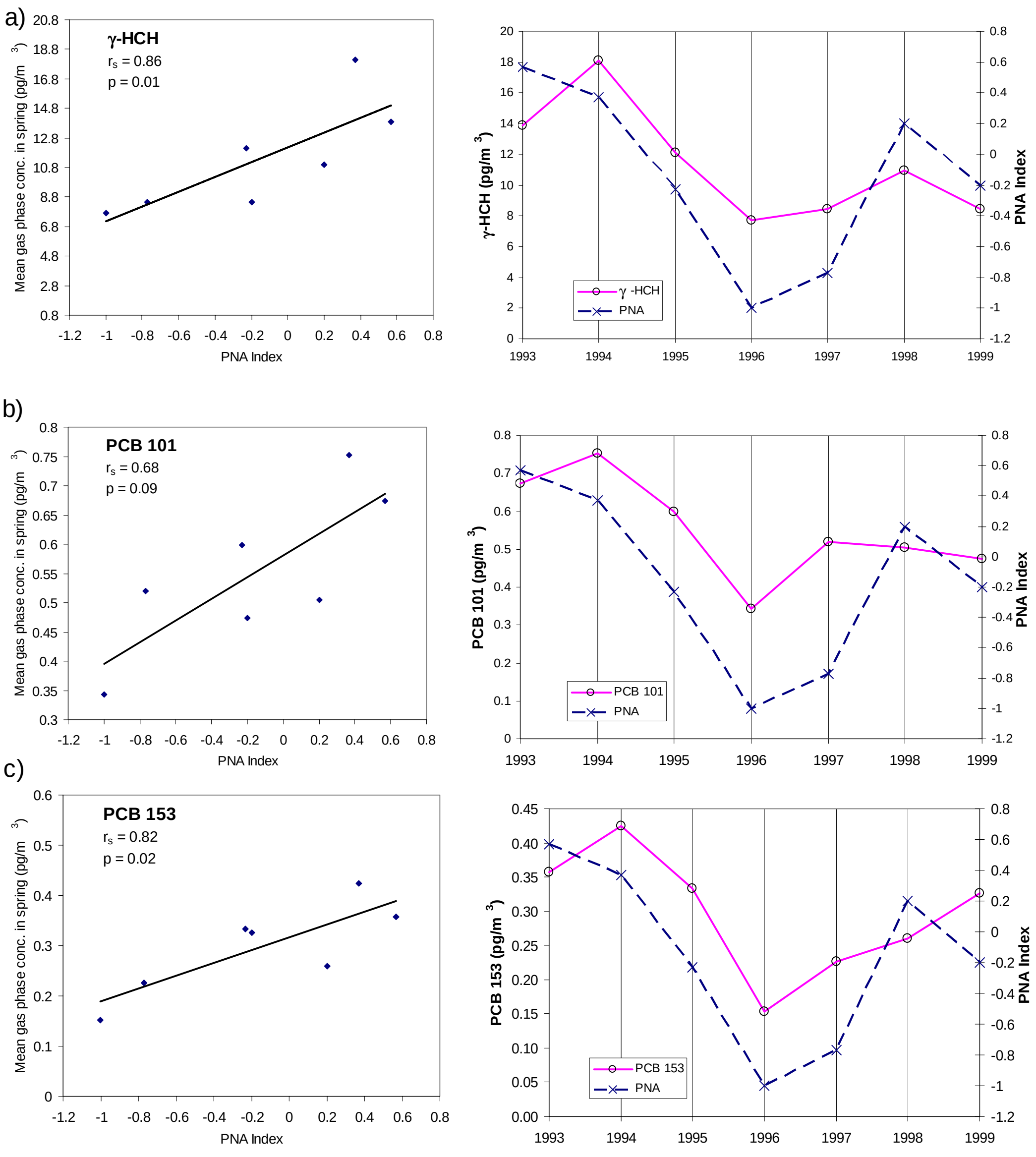

Figure 1. Association between PNA Index and springtime mean gas phase concentration of (a) γ-HCH, (b) PCB 101 and (c) PCB 153. Left panels show linear correlation plots between air concentrations and PNA indices. Spearman rank order correlation coefficients, r_S, and the p-values are also given. Right panels show time series of spring mean gas phase concentrations and the PNA index.