SEASONAL TRENDS OF ATMOSPHERIC CHLORINATED PESTICIDES IN THE CENTRAL HIMALAYA

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Abstract

Passive air samplers were used to measure the seasonal concentrations of hexachlorobenzene (HCB), endosulfan I, α-hexachlorocyclohexane (α-HCH), γ-hexachlorocyclohexane (γ-HCH), p,p'-DDE and p,p'-DDT over an altitudinal transect between 2638-5605m.a.s.l in the Central Himalaya (27°44.446'-27°59.526'N 86º42.745'-86º49.790'E). There is no known usage of these chemicals in the region. Summer trends between May and October showed increasing atmospheric concentrations of all the chemicals up to a maximum of 5000m.a.s.l and then declining above that elevation. Winter trends between November and April show a decline in atmospheric concentration for all chemicals except for HCB. This indicates that the summer monsoon is a driving force in the movement of chemicals from the Indian subcontinent into the central Himalaya.

Introduction

Evidence is emerging that mountain regions sometimes have higher-than-expected concentrations and deposition rates of selected persistent organic pollutants (POPs)[1]. Earliest indications for this phenomenon came from a global survey of contaminants in vegetation, which reported the highest levels of hexachlorobenzene (HCB) in low latitudes at sites with high elevation[2]. Elevated levels of organochlorinated compounds were also reported in fish, sediments, snow and air from the Canadian Rocky Mountains[3-6]. To date, very little research has been published on POPs in the Himalaya to test if this phenomenon is observed in the highest mountain range in the world[7-9]. This mountain range is wedged in between India and China, the two most populous countries in the world. High population density often results in increased usage of pesticides and high emissions of air pollutants. In the past, the Indian subcontinent and China had experienced heavy use of organochlorine pesticides, such as hexachlorocyclohexanes (HCHs) and DDT, for agricultural purposes. It has been estimated that since its initial formulation, more than 10^8 kg of DDT have been used in India[10,11]. India banned DDT for agricultural purposes in 1989, but continues to use between 5000 to 10,000 kg y^{-1} for malaria control[11]. Technical HCH, once the most heavily used pesticide in India with an annual usage exceeding 6×10^7 kg, was banned in 1997, but lindane (γ-HCH) has been used as a replacement since that time[11]. From the 1950s until its ban in 1983, China produced and used 4×10^8 kg of DDT as well as 4.9×10^9 kg of technical HCH. Lindane and DDT are still being used in lower amounts to control certain insects[12]. Although the use of extremely persistent chemicals has decreased, they will likely persist in the environment and possibly impact the Himalayan mountain ecosystems for years to come.

The Himalaya and surrounding lowlands are linked by distinct climatic systems that operate in the region: the Southwest or summer monsoon (June-September) and the continental westerlies (December-April). Heating over the Indian subcontinent and Tibetan Plateau increases in the early summer months causing a low pressure region to develop over the land. The comparatively cool humid air from the southern hemisphere crosses the equator and is accelerated towards the low-pressure area over the continent. This causes the air mass over India to be driven into the mountain ranges in the northern part of the subcontinent and eventually into the Himalaya which normally results in significant precipitation events. In winter the Himalaya is dominated by prevailing continental westerlies[13]. The atmospheric transport and fate of pesticides in the Himalaya is expected to be influenced by this unique meteorological system. In this study we attempt to understand the movement of a variety of persistent chlorinated pesticides in the Central Himalayan atmosphere on a seasonal basis over a wide range of elevations.

Methods

Passive air samplers (PAS) as described by Wania et al.[14] were deployed in Nepal from 2638m.a.s.l to 5605m.a.s.l. during two seasons: May 2004-October 2004 and November 2004-April 2005 in order to coincide with major changes in the airmass movement over the Himalaya. Sampler tubes were packaged for deployment and retrieval in baked aluminium foil and double packaged in sealable plastic bags. Sampler tubes were sent to the laboratory of Freshwater Institute where they were stored frozen until analysis. The XAD-2 resin inside the PAS tube was spiked with deuterated endosulfan and α-HCH as well as PCB 30 surrogate compounds and allowed to soak in 100ml dichloromethane for 20 min. and then eluted with a further 250 ml of DCM from a stainless steel column. The samples were evaporated with a rotary evaporator taken into 1 ml hexane and dried with sodium sulphate, followed by cleaning up and fractionation on 1.2% deactivated Florisil. The samples were then taken into isooctane evaporated to 200µl and spiked with and aldrin performance standard. One µl of each sample was injected onto a Varian GC/ECD equipped with a 60m DB-5 capillary column and analyzed for several chlorinated pesticides.

Minimum detection limits (MDLs) defined as the average of field blanks (n=4) plus 3 times the standard deviation were determined. MDLs were found to be 0.11, 0.14, 0.13 and 0.62 pg/m^3 for HCB, α-HCH, γ-HCH and p,p'-DDE respectively. Endosulfan I and p,p'-DDT were not detected in blanks. Surrogate recoveries averaged 78%, 72% and 76% for PCB30, deuterated α-HCH and deuterated endosulfan I respectively. Deuterated surrogates were easily resolved from the native compounds by retention time on the 60m column. Samples were not recovery or blank corrected.

Results and Discussion

Figure 1 compares the seasonal trends in atmospheric concentration of HCB, endosulfan I, α-HCH, γ-HCH, p,p'-DDE and p,p'-DDT over 3000m variation in altitude in a region with no known usage of these chemicals. All the chemicals behaved similarly over the altitude range in summer with increasing concentrations up to an elevation of 5000m.a.s.l. followed by a decrease at 5600m.a.s.l. In winter all the chemicals decreased in concentration with altitude with the exception of HCB which showed an increase over the same elevation range. Summer levels of endosulfan I, α-HCH and γ-HCH in the Central Himalaya are similar to those detected in the Canadian Rocky Mountains during the snowmelt and summer seasons of 2004, while the winter levels in the Central Himalaya are much lower[15]. The proportion of α-HCH/γ-HCH appears to be higher in Asia. HCB levels are slightly higher than those found recently in Europe and lower than those detected in industrial parts of China in 2004[16,17]. One sample from 841m.a.s.l. in an agricultural region showed a p,p'-DDT concentration of 1000 pg/m^3 indicating current use. The atmospheric concentration of p,p'-DDT was found to be higher than p,p'-DDE at all elevations; both chemicals are in the lower range of concentrations reported recently in Europe[17]. All results were well above MDLs.

The results indicate that the summer monsoon is the driving force for chemical movement to the Central Himalaya. It strongly links the Indian Subcontinent with atmospheric contaminants up to very high elevations. Chemicals of lower volatility such as DDT and DDE are also moving into the high alpine regions. This is in contrast to results from the Canadian Rocky Mountains where these chemicals are not found to be enriched in high altitude areas[18]. This difference is likely due to higher temperatures in the Indian subcontinent coupled with the driving force of the summer monsoon air mass. We observe the same upper elevation limits of chemicals regardless of vapour pressure indicating an upper level atmospheric barrier in summer. Interestingly we see that snow deposition is not playing a major role most likely due to low levels of snowfall in this region of the Himalaya and the direction of the air mass. Increasing levels of HCB during winter likely indicate that this compound is found uniformly in the environment unlike the other compounds that come from a specific source namely the Indian subcontinent.

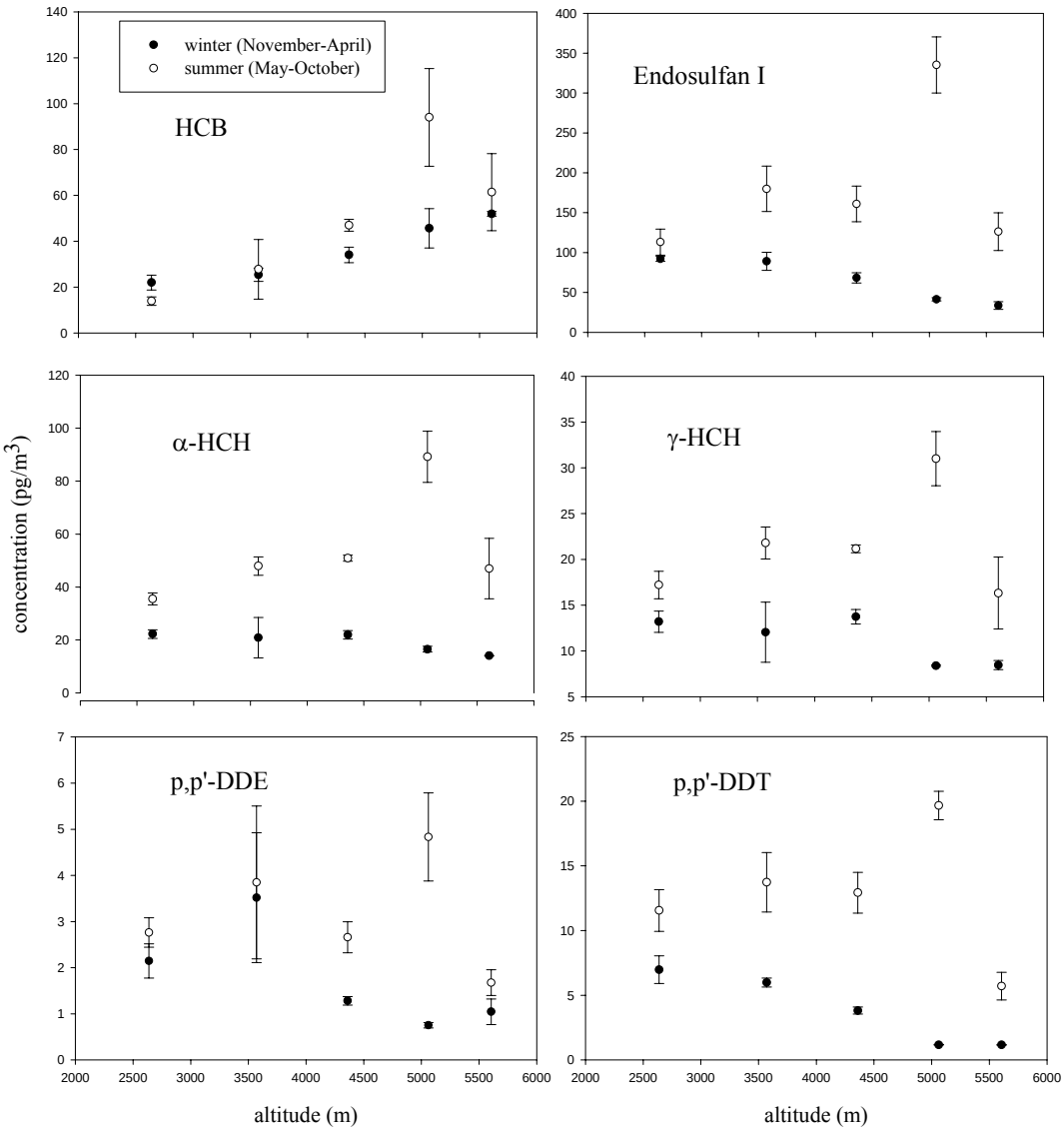

Figure 1: Atmospheric concentrations of HCB, endosulfan I, α-HCH, γ-HCH, p,p'-DDE and p,p'-DDT respectively as a function of elevation and season.

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