

Spatial variability of atmospheric semivolatile organic compounds in Chile

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ABSTRACT

In an effort to characterize the spatial variability of pesticides, polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs) in the Chilean atmosphere, XAD-based passive air samplers (PAS) were deployed for 12 months (2006–2007) along three elevational gradients in Northern, Central and Southern Chile, for a total of 20 sampling sites, ranging in elevation from 10 to 4400 m and ranging over 26 degrees of latitude. Hexachlorobenzene (HCB) had largely uniform air concentrations with latitude and altitude, and is therefore used to derive sampler specific sampling rates, that account for differences in uptake kinetics due to environmental factors such as altitude and temperature. Levels of pesticides in the atmosphere of Chile are very low and only α - and γ -hexachlorocyclohexane, α -endosulfan and chlorothalonil were present in all of the samples. The concentrations of these four pesticides in air decreased from North to South, with much steeper gradients for chlorothalonil and endosulfan. Chlorothalonil levels were greatly elevated in air samplers deployed in urban locations. Endosulfan concentrations in air tend to increase with elevation. Since endosulfan is particularly susceptible to mountain cold-trapping, such gradients may reflect the re-volatilization of endosulfan that had been preferentially deposited at higher altitudes. Alternatively, increasing endosulfan concentrations with elevation could be the result of transport to higher altitude sites from the other side of Andes. Airshed analysis indeed indicates that air masses originating outside of Chile have a higher influence on the higher elevation sites, especially along the Northern transect. Concentrations of four-ring PAHs ranged from 0.3 to 1300 pg m^{-3} , with higher levels in air samplers deployed in urban areas and close to roads. PCB concentrations were low (0.7–27 pg m^{-3}), with slightly higher levels of volatile congeners in two urban sampling sites in Concepcion and Arica. PCB-52 is the only congener present in all samples, with levels that are remarkably uniform across the country.

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1. Introduction

Chile is a South American country with an extraordinary latitudinal extension (4630 km, 18 to 56°S) resulting in highly variable climate with extremely dry desert climate in the North, Mediterranean climate in the Central region, and cool and damp climate in the South. Equally staggering are the elevation changes, ranging from sea level to 6880 m over a mere 200 km distance. Similar to the latitudinal changes, vertical gradients are also steep gradients of precipitation and temperature. The Chilean atmosphere is under Pacific influence and a generally low population density suggests that local sources of organic contaminants should be relatively

minor. Chile thus is the ideal location to determine Southern hemispheric background concentrations of semivolatile organic contaminants (SVOCs). In particular, it should be a good place to study latitudinal and altitudinal gradients in organic contamination.

Relatively little information on SVOCs in the Chilean environment exists. A number of studies have reported the presence of SVOCs in Chilean fresh water and coastal ecosystems, with a focus on sediments and biota from the Biobio river basin (Focardi et al., 1996; Barra et al., 2009), and the coastal environments of Valdivia, Valparaiso and Northern Patagonia (Palma-Fleming et al., 1998a,b, 2004, 2008; Toro et al., 2004; Montory et al., 2008). Barra et al. (2001a,b) reported the concentrations of a variety of organochlorine pesticides (OCPs) in the sediments of four Chilean lakes, whereas Quiroz et al. (2005) inferred the time trend of polycyclic aromatic hydrocarbon (PAH) deposition from a sediment core. Bivalves have been used to infer latitudinal trends in the contamination of the Chilean coastal environment with polychlorinated

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biphenyls (PCBs). The International Mussel Watch programme included a number of Chilean coastal locations (Sericano et al., 1995), and noted higher concentrations of PCBs in Punta Arenas in the very South of the country compared to mussels from lower latitudes. Barra et al. (2002) and Mendoza et al. (2006) also found much higher concentrations and more volatile PCB congeners in the southernmost part of Chile when analyzing sessile marine bivalves along a latitudinal transect. This may be due to the long-range transport or local sources of PCBs (Mendoza et al., 2006). On the other hand, concentrations of PCBs and OC pesticides were higher in gull eggs from central Chile than from the southernmost part of the country (Munoz and Becker, 1999).

Some work has been reported on SVOC contamination along elevation gradients in Chile. When analyzing soil, water, and snow sampled at different altitude in the Laja River basin of South Central Chile, Barra et al. (2005) did not observe clear concentrations gradients for PAHs, PCBs or hexachlorocyclohexanes (HCHs). On the other hand, when analyzing selected SVOCs (pentachlorobenzene, hexachlorobenzene (HCB), HCHs, DDE and DDT) in mosses, sediments and soils from three altitudinal transects in the Chilean Andes at 18°S (3200–4500 m), 37°S (345–1330 m), and 45°S (10–700 m), Grimalt et al. (2004) and Borghini et al. (2005) noted higher concentrations with increasing elevation, consistent with observations in Northern hemispheric mountains (e.g. Davidson et al., 2003; Daly et al., 2007c). Both studies observed significant linear relationships between log transformed concentrations of SVOCs and air temperature, with correlation coefficients exceeding 0.5 for the more volatile compounds, such as HCHs, HCB, PCB-18 and 52. Very low concentrations of the selected SVOCs and the absence of significant local pollution sources in the Andes allowed these studies to infer that temperature is the primary factor controlling the distribution of SVOCs in remote ecosystems in the Southern Hemisphere.

Measurements of SVOCs in the Chilean atmosphere are very limited. Levels of PAHs and PCBs in urban particles sampled in Santiago and Temuco were found to be comparable to other urban areas around the world (Kavouras et al., 1999, 2001; Mandalakis and Stephanou, 2002). Only Pozo et al. (2004) have so far reported the levels of SVOC in Chilean air outside of urban areas. Deploying passive air samplers (PAS) based on polyurethane foam disks for a three month period they observed generally low concentrations of selected OCPs, PCBs and polybrominated diphenyl ethers, but endosulfan was prevalent and showed a decreasing concentration gradient from the North to the South of Chile. In this study we set out to determine annual average air concentrations of SVOCs along three altitudinal gradients, situated at three different latitudes within Chile using XAD-resin PAS. The objective was to gain insight into the atmospheric background contamination of SVOCs in the Southern hemisphere and to record and interpret concentration variability with altitude and latitude.

2. Methods

2.1. Sampling

Duplicate XAD-PAS were deployed for one year along three elevation gradients in Northern, Central and Southern Chile (Fig. 1). The 20 sampling sites, ranging in elevation from 10 to 4400 m and ranging over 26 degrees of latitude, represent a wide variety of landscapes (Fig. S1). The XAD-PAS consist of a stainless steel mesh cylinder, filled with XAD-2 resin and suspended in a stainless steel shelter with an open bottom (Wania et al., 2003). In this study, a short XAD-PAS, with half the length of the original design (Wania et al., 2003) was used. The XAD-resin PAS was prepared as described previously (Wania et al., 2003).

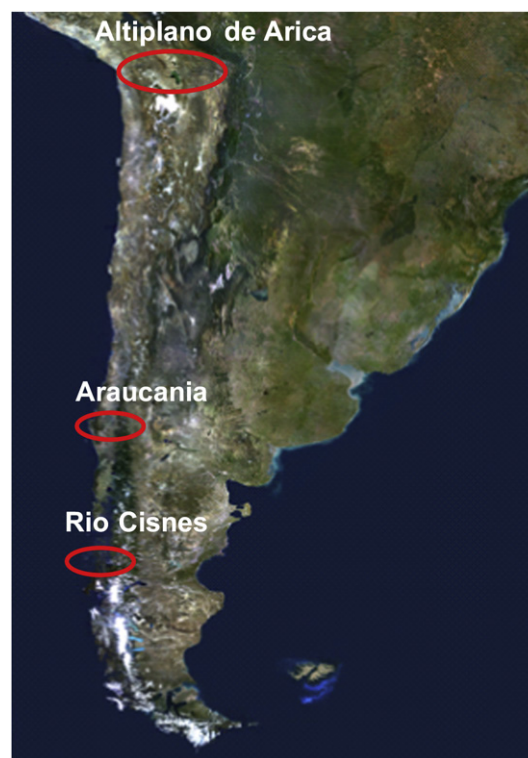


Fig. 1. Location of the three sampling transects along elevation gradients within Chile.

The transect in Northern Chile (“Altiplano de Arica”, 18°S), deployed from July 2006 to July 2007, included six samplings sites between Arica (48 m) and Lago Chungara (4400 m). The transect in Central Chile (“Araucania”, 38°S), deployed from March 2006 to March 2007, included seven sites between Lago Llu Llu (10 m) and Paso Pino Hachado (1874 m), and an additional sampling site on the campus of the University of Concepcion. The transect in Southern Chile (“Rio Cisnes”, 44°S), deployed from February 2006 to February 2007, included five sites between Puerto Cisnes (50 m) and Alto Rio Cisnes (700 m), and an additional sampling site in the vicinity of the town of Coyhaique.

2.2. Chemical analysis

After spiking with recovery standards (d_6 - γ -HCH, $^{13}C_{10}$ -HEPX, $^{13}C_{10}$ -TN, ^{13}C -Dieldrin, $^{13}C_{p,p'}$ -DDT), the XAD-resin was Soxhlet-extracted with dichloromethane for 20–22 h. The volume-reduced extracts were passed through baked sodium sulfate to remove any water residue. The final volume of the extracts was 1 ml, and 100 ng of mirex was added for volume correction. The extracts were analysed for 56 PCB congeners, legacy OCPs (HCB, *cis*-chlordane (CC), *trans*-chlordane (TC), *trans*-nonachlor (TN), heptachlor (HEPT), heptachlorepoxyde (HEPX), dieldrin, aldrin, dichlorodiphenyltrichloroethane (DDT) and its metabolites (DDE, DDD), α -HCH, γ -HCH), current use pesticides (CUPs) (α - and β -endosulfan and its metabolite endosulfan sulfate, dacthal, chlorothalonil, trifluralin, and pendimethalin) and four four-ring PAHs (benz[a]anthracene, chrysene, fluoranthene, pyrene) using an Agilent 6890 gas chromatography (GC) coupled to a 5973 mass selective detector (MSD) in single ion monitoring mode. PCBs and PAHs were determined with electron impact ionization and pesticides with negative chemical ionization. A DB-5 column (60 m, 0.25 mm i.d., 0.3 μ m film thickness) was used for OCPs and PCBs separation, whereas PAHs were separated on a DB-5MS column (60 m, 0.25 mm i.d., 0.1 μ m

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