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# Polychlorinated biphenyl behavior in soils amended with biosolids

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# ABSTRACT

Assessment of the mobility of polychlorinated biphenyls (PCBs) in soils, amended with biosolids at a rate of 30 Mg ha<sup>-1</sup>, was performed using an incubation process and leaching columns. The incubation process was carried out for 0, 30, and 60 d under field capacity conditions at 25 °C. The mobility of PCBs was assessed using solutions of 0.5 mol  $L^{-1}$  CaCl<sub>2</sub> and 25 mg  $L^{-1}$  linear alkylbenzenes sulfonate (LAS).

Ultrasound-assisted pressurized solvent extraction (US-PSE) was utilized for compound separation from the solid matrix. Compounds were determined by gas chromatography coupled to mass spectrometry.

The biosolids, containing a background PCB concentration about 300  $\mu$ g kg<sup>-1</sup>, were spiked with the analytes at 300 mg kg<sup>-1</sup> to obtain a clearer determination of their behavior when the biosolid was mixed with soil. In biosolid-amended soils, an increase was observed in the extractability of PCBs with increasing incubation time, which may be attributed to organic matter breakdown. The leaching column study showed that CaCl<sub>2</sub> was unable to mobilize the PCBs from the biosolid to the soil, whereas LAS mobilized these compounds within the time scale implicit in the experiment (30 d). The most mobilized congeners in the columns corresponded to those with the greatest molecular weight (hexa- and heptachlorinated), probably due to the higher hydrophobicity of these compounds. Results indicate that the presence of important concentrations of LAS in biosolids could mobilize PCBs from soil to the freatic level.

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## 1. Introduction

In Chile, the construction of water treatment plants is intended to provide treatment coverage for a significant portion of the country by 2010. The existence of new treatment plants implies improvement in water quality, with consequent sanitary and agricultural benefits; however, there is also the possible generation of huge environmental problems resulting from large-scale biosolid buildup. Biosolids contain macro- and micronutrients, microorganisms, trace metals, inorganic, and organic compounds. Organic compounds may be subdivided into harmless (from plants or animals) or toxic. The latter compounds may be polychlorinated biphenyls (PCBs), polycyclic aliphatic hydrocarbons (PAHs), surfactants such as linear alkylbenzene sulfonate (LAS), and volatile organic compounds (VOCs), among others (Eljarrat et al., 1997; Harrison et al., 2006; Cai et al., 2007; Singh and Agrawal 2008).

Biosolids may be used as both fertilizer complements and soil amendments. However, upon incorporation into the soil, organic

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contaminants may be transferred, with possible adverse effects both for the environment and for humans.

Polychlorinated biphenyls are synthetic organic compounds that exhibit high environmental persistence due to their high chemical stability, relatively low volatility, high dielectric constant, and elevated resistance to thermal decomposition. Although there are no natural sources of PCBs, they may be found in various environmental matrices such as waters, sediments, soils, and biosolids. These compounds arrive in these matrices through the production, use, disposal, spills, leaks, and fires of PCB containers.

In Chile, PCBs are found in most regions, either in use and/or stored, with a national total of 569 720 L. The large quantity of these compounds results mainly from the contribution of the mining sector (Conama, 2005).

Although most organic compounds originating from biosolid application degrade in the soil or are sorbed in the surface layer, a primary concern is that they could migrate to drinking water sources. Within this context, the mobility of PCBs, amended with biosolids at a rate of 30 Mg ha<sup>-1</sup> in agricultural soils from central Chile, was assessed through a process of incubation and use of leaching columns. The incubation process was carried out for periods of 0, 30, and 60 d under constant temperature and field capacity conditions. For the leaching column study, two leaching solutions were used to assess mobility: 0.5 M CaCl<sub>2</sub> and 25 mg L<sup>-1</sup>



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LAS, considering that biosolids contain these surfactants at high concentrations (Jacobsen et al., 2004).

To extract PCBs from soil, biosolids, and the soil-biosolid mixture matrices, there are several techniques available, such as Soxhlet extraction, ultrasound-assisted extraction, microwave-assisted extraction (López-Avila and Benedicto, 1996; Sparr-Eskilsson and Björklund, 2000), supercritical fluid extraction (Nilsson et al., 2002), and pressurized solvent extraction (Richter et al., 1996; Camel, 2001; Kiguchi et al., 2006). The choice of technique depends mainly on extraction efficiency, recovery percentage, reproducibility, minimal use of solvent, simplicity, and ease of use. Accordingly, the US-PSE technique was selected in this study because of its demonstrated high efficiency and low use of solvents in PCB extraction from biosolids (Rocco et al., 2008).

#### 2. Materials and methods

#### 2.1. Soil samples

Four soils were sampled from agricultural land area near Santiago in central Chile. Composed surface (0-10 cm) soil samples were drawn. At each site, five replicates were obtained from each corner and center of the plot using a stainless-steel hand auger with plastic liner. Samples were mixed, reduced, and processed. All samples were air-dried and sieved through a 2-mm mesh-size polyethylene sieve. Concerning soil classification, all soils were Mollisol and the order identified as El Maitén: S1 (Lat. S 33°19'902" Long. O 70°51'873), Coyuncaví Bajo: S2 (Lat. S 33°23'158" Long. O 71°07'1262"), Miraflores: S3 (Lat. S 33°24'418" Long. O 71°04'745"), and La Paloma: S4 (Lat. S 33° 19'909" Long. O 70°52'367"). Two samples belonged to the Lampa series (S1 and S4), while the remaining samples belonged to the Pomaire (S2) and Chorombo (S3) series, respectively. The organic matter (OM) content of these samples was 4.7%, 3.2%, 3.1%, and 1.2% for soils S1, S2, S3, and S4, respectively. Full characterization of these soils has been reported previously (Ahumada et al., 2010).

#### 2.2. Biosolid sample

A sample of anaerobically stabilized sludge or biosolid was collected from a monofill at the Santiago sewage treatment plant. Organic matter content of this sample was 42%.

#### 2.3. Procedures

#### 2.3.1. Incubation process

Biosolid and soil samples amended with the biosolid at a rate of 30 Mg ha<sup>-1</sup> were incubated under field capacity conditions at 25 °C for 0, 30, and 60 d. For the amendment, about 10 g of soil was accurately weighed and mixed with 0.120 g of biosolid spiked with 100 mg kg<sup>-1</sup> of each Aroclor: 1242, 1254, and 1260 (Supelco). The PCB homologues of mono-, di-, tri-, tetra-, penta-, hexa-, hepta-, octa-, nona-, and decachlorinated were abbreviated as CBp, DCBp, TCBp, TeCBp, PeCBp, HCBp, HeCBp, OCBp, NCBp, and DeCBp, respectively.

## 2.3.2. Mobility study by using leaching columns

To carry out the leaching column study, 2.5 cm inner diameter (ID) and 12 cm long acrylic leaching columns were used, divided into six equal 2 cm sections. The top section of the column contained 1.5 g of biosolid spiked with 100 mg kg<sup>-1</sup> of each Aroclor mixture (1242, 1254, and 1260). Each of the other four sections contained 12 g of soil, and the bottom section was filled with

quartz sand and glass wood (Fig. 1). The experiment was performed at ambient temperature and pressure.

For the mobility assessment of the PCBs present in soil, 0.5 M  $CaCl_2$  was used to simulate the ionic strength of the soil solution, and 25 mg  $L^{-1}$  LAS was used to simulate the effect of this surfactant, which could be supplied by constant application of biosolids to soil.

Leaching in each column was studied for 30 d with the daily addition of 2–3 mL of the respective solutions.

#### 2.3.3. Extraction process

Analyte extraction from the different matrices (biosolid and soil–biosolid mixture) was carried out by utilizing the methodology of US-PSE previously described by our group (Rocco et al., 2008). A 500 mg sample was used; the flow rate of 1:1 (v/v) dichloromethane/acetone (Merck) was kept at  $2 \text{ mL min}^{-1}$ , with 1900 ± 100 psi working pressure and 30 min extraction time with ultrasound. Finally, the extract was collected and stored in glass tubes at  $4 \,^{\circ}$ C until the next step.

The extract was then dried by passing it through a 10 cm long  $\times$  15 mm ID glass column filled with anhydrous Na<sub>2</sub>SO<sub>4</sub>. Finally, the column was rinsed with 10 mL of chromatographic grade acetone.

After drying, the extract was concentrated by a Kuderna-Danish system at 80  $^\circ$ C to a final volume of 1 mL.

# 2.3.4. Clean up

The following treatment was necessary only for the biosolid extracts. The extract was subjected to a change of solvent by adding 20 mL *n*-hexane and concentrating to 1 mL in a Kuderna-Danish evaporator at 90 °C and following the 3630C EPA method (US EPA, 1996). Finally, the extract was concentrated under N<sub>2</sub> flow to a volume of 1 mL and stored in vials for further analysis by GC-MS.

#### 2.3.5. Chromatographic determination

A mixture of phenanthrene-d10/crysene-d12 (Supelco) was added to the extract as an internal standard (Supelco). A 95 XP GC–MS MAT instrument was used with a 30 m  $\times$  0.32 mm ID  $\times$  0.25  $\mu m$  internal phase HP5 column. In order to determine



Fig. 1. Leaching column diagram.

the different PCB homologues, mass spectrometry in SIR mode (Rocco et al., 2008) was used.

Operating parameters for chromatographic determination were: carrier gas, helium at 1 mL min<sup>-1</sup>; injection temperature, 250 °C; injection volume, 1 µL (splitless). The temperature program was 80 °C to 280 °C (5 min) at 10 °C min<sup>-1</sup>. The MS transfer line was held at 280 °C and quantifications were based on calibration with standard analytes using mass spectrometric parameters.

#### 3. Results and discussion

To extract PCBs from soil, the US-PSE technique was selected because of its demonstrated high efficiency and low use of solvents (Rocco et al., 2008). This technique was first applied to determine the PCB concentrations in the original biosolid and in biosolids spiked with 100 mg kg<sup>-1</sup> of each Aroclor. The resulting values are shown in Table 1. It was found that HCBp and HeCBp homologues were predominant in the original biosolid sample. However, as expected, the spiked biosolid showed all the congeners present in the Aroclor standards.

Since background concentrations of PCBs in the biosolid were too low to study the effect of the possible mobility of these compounds, this matrix was spiked with 100 mg kg<sup>-1</sup> of each Aroclor mixture (Aroclor 1242, 1254 and 1260) to a final total concentration of 300 mg kg<sup>-1</sup> of PCBs in the biosolid. It is important to establish that each Aroclor mixture contained approximately 90 PCB congeners (Frame et al., 1996).

Concerning incubation time (Table 1), there was an apparent increasing trend in PCB extractability with increasing incubation

#### Table 1 Concentrations of PCB homologues in the biosolid at different incubation times.

Analyte	Concentration of PCBs in original biosolid (mg kg <sup>-1</sup> )			Concentration of PCBs in spiked biosolid (mg kg <sup>-1</sup> )		
	0 d	30 d	60 d	0 d	30 d	60 d
DCBp TCBp TeCBp PeCBp HCBp HeCBp	$\begin{array}{c} ND \\ ND \\ ND \\ 0.12 \pm 0.01 \\ 0.101 \pm 0.004 \end{array}$	$\begin{array}{c} ND \\ ND \\ ND \\ ND \\ 0.19 \pm 0.02 \\ 0.13 \pm 0.02 \end{array}$	$\begin{array}{c} ND \\ ND \\ ND \\ ND \\ 0.19 \pm 0.01 \\ 0.14 \pm 0.03 \end{array}$	$15 \pm 241 \pm 348 \pm 467 \pm 261 \pm 136 \pm 4$	$15 \pm 2 41 \pm 6 49 \pm 4 69 \pm 11 63 \pm 1 36 \pm 3$	$16 \pm 243 \pm 252 \pm 472.8 \pm 0.366 \pm 538 \pm 3$
Total	$0.22 \pm 0.01$	$0.32\pm0.03$	$0.33\pm0.03$	$267 \pm 7$	$273\pm13$	287 ± 8

Mean of three independent determinations. ND, not detected.

#### Table 2

Concentrations of PCB homologues in the soils at different incubation times.

time, which is not significant according to Tukey HSD (Honestly Significant Difference) test (p < 0.05).

We should note that the total amount of these compounds obtained was always lower than 300 mg kg $^{-1}$ . This result is because the CBp, OCBp, and NCBp homologues, which accounted for 3.30% of the total concentration, were not considered for quantification due to their low concentration level.

# 3.1. Effect of incubation time on PCB extractability

No PCBs were detected in the soils being studied. After the 30 Mg ha<sup>-1</sup> amendment, these compounds were determined at different incubation times. According to the quantity of PCBs added to the biosolid, the concentration in the soil-biosolid system should be 1.5 mg kg<sup>-1</sup> total PCBs. As can be observed in Table 2, extractability of these compounds increased with increasing incubation time, with a significant increase being observed in S4, which exhibits a lower OM content (1.2%).

In general, PCB extractability was higher after incubation due to organic matter breakdown. In a previous study (Retamal et al., in press), this biosolid-soil system was subjected to respirometry, confirming OM breakdown by release of 3500 mg de CO<sub>2</sub> kg<sup>-1</sup> of solid in 10 d.

Concerning PCB homologues, PeCBp turned out to be the most abundant in all the soils. Extractability of these congeners decreased with increasing incubation time, except for S4. This result could be attributed to the compound's biochemical breakdown by microorganisms supplied by the soil. In addition, the decrease in congener extractability could be the result of pentachlorinated congeners being the most abundantly used and breaking down to those of lower molecular weight, which could explain the concomitant increase in tetra-, tri-, and dichlorinated congeners with increasing incubation time (Borja et al., 2005; Field and Sierra-Alvarez, 2008; Mandalakis et al., 2008).

In general, after incubation, the highest extraction of PCBs was observed in S4 (Table 2). The higher extractability of these compounds in this soil could be attributed to its lower OM content.

# 3.2. Study of PCB mobility by leaching columns

As demonstrated earlier, organic matter breakdown is critical to PCB extractability. In this sense, for the studies of mobility in leaching columns, two soils were selected, S1 and S4, which differ

Time (day)	Concentration <sup>a</sup> of PCBs (mg kg <sup>-1</sup> )						
	DCBp	ТСВр	ТеСВр	РеСВр	НСВр	НеСВр	Total
Soil 1 0 30 60	0.057 ± 0.003CDE 0.0678 ± 0.0004DEF 0.08 ± 0.02FG	0.1245 ± 0.0001AB 0.157 ± 0.002BCD 0.202 ± 0.008EF	0.20 ± 0.04A 0.245 ± 0.005ABC 0.27 ± 0.02C	0.41 ± 0.04EF 0.34 ± 0.03CD 0.21 ± 0.04A	0.226 ± 0.007BC 0.30 ± 0.02CDEF 0.36 ± 0.03F	0.11 ± 0.03A 0.129 ± 0.009ABC 0.137 ± 0.002ABC	1.124 ± 0.07AB 1.24 ± 0.04ABC 1.26 ± 0.06ABCD
Soil 2 0 30 60	0.037 ± 0.004AB 0.07 ± 0.01DEF 0.087 ± 0.008G	0.160 ± 0.007CD 0.19 ± 0.05DEF 0.216 ± 0.007F	0.241 ± 0.001ABC 0.265 ± 0.007C 0.270 ± 0.007C	0.37 ± 0.02CDE 0.28 ± 0.02B 0.211 ± 0.007A	0.32 ± 0.01DEF 0.340 ± 0.003EF 0.35 ± 0.01EF	0.14 ± 0.02ABC 0.142 ± 0.006ABC 0.155 ± 0.006C	1.26 ± 0.03ABCD 1.28 ± 0.05CD 1.29 ± 0.02CD
Soil 3 0 30 60	0.0318 ± 0.0002A 0.042 ± 0.006ABC 0.07 ± 0.01EFG	0.285 ± 0.008G 0.211 ± 0.001F 0.11 ± 0.02A	0.199 ± 0.008A 0.261 ± 0.01BC 0.28 ± 0.07C	0.50 ± 0.01G 0.48 ± 0.04G 0.46 ± 0.03FG	0.134 ± 0.008A 0.2 ± 0.1AB 0.26 ± 0.04CD	0.11 ± 0.01AB 0.14 ± 0.02ABC 0.21 ± 0.03D	1.26 ± 0.02ABCD 1.3 ± 0.1CD 1.4 ± 0.1CD
Soil 4 0 30 60	0.054 ± 0.001BCD 0.068 ± 0.001DEF 0.077 ± 0.003FG	0.1246 ± 0.0005AB 0.147 ± 0.002BC 0.170 ± 0.003CDE	0.21 ± 0.01AB 0.23 ± 0.02ABC 0.243 ± 0.003ABC	0.32 ± 0.01BC 0.3610 ± 0.0004CDE 0.384 ± 0.005DE	0.28 ± 0.02CDE 0.327 ± 0.001DEF 0.370 ± 0.004F	0.137 ± 0.003ABC 0.144 ± 0.004BC 0.16 ± 0.01C	1.12 ± 0.02A 1.28 ± 0.02BCD 1.40 ± 0.02D

<sup>a</sup> Values followed by the same letter in each column show no significant differences for p < 0.05, according to Tukey HSD test.

mainly in organic matter content (Ahumada et al., 2010). Several methods are available to measure the leaching potential of chemicals in soil under controlled laboratory conditions. An intact soil core is the alternative method to assess mobility mechanisms considering macropore flow (Karathanasis et al., 2005). It is known that preferential flow into the macropore enhances the leaching of analytes through soil. However, hand-packed soil columns prepared after drying and sieving is also useful to compare the mobility of contaminants under different experimental conditions (Tucker et al., 1975; Till et al., 1994; Futch and Singh, 1999; Xu et al., 2002; OECD, 2004; Conrad et al., 2006). We select this last alternative by using calcium chloride and LAS solutions as leaching agents.

Calcium chloride was utilized to simulate the soil solution. Under these conditions, at the time scale at which the experiment was carried out (30 d), PCBs were not detected in any of the column sections, thus we concluded that if they reached the soil in any way, under normal conditions, these compounds would not be mobilized. This result was predictable given the high hydrophobicity of PCBs, leading to retention in the hydrophobic organic matter of the soil-biosolid system. Hence, this result may be considered the basis for further studies and is consequent with the antecedent that pure water cannot mobilize PCBs in soil columns (Tucker et al., 1975).

LAS are found at high concentrations in biosolids (Jacobsen et al., 2004), and they are soluble in water. In the biosolid used in this study, the concentration of LAS was  $5.02 \pm 0.49$  g kg<sup>-1</sup>, determined by solvent extraction of the biosolid and GC–MS (Field et al., 1992). The use of a LAS solution as a leaching solution would simulate repeated biosolid incorporation to soils. Mobilization was observed in the column, but it was different in the selected soils. In the S1 column, with the highest OM content, PCBs were present in the second and third sections of the column. PCB congeners detected in these sections are shown in Table 3.

On the other hand, in the S4 column, PCBs were detected as far as the fifth section of the leaching column, as seen in Table 4. Light-

#### Table 3

Concentrations of PCB homologues found in each section of the leaching column for S1 soil, leaching solution: 25 mg  $L^{-1}$  LAS.

Analyte	Concentration of PCBs (mg kg <sup>-1</sup> )				
	2nd section 3rd section		4th section	5th section	
DCBp	$0.016 \pm 0.001$	ND	ND	ND	
ТСВр	$0.0160 \pm 0.0002$	ND	ND	ND	
TeCBp	0.11 ± 0.03	0.091 ± 0.006	ND	ND	
РеСВр	$0.25 \pm 0.03$	$0.205 \pm 0.005$	ND	ND	
НСВр	$0.01 \pm 0.01$	$0.008 \pm 0.003$	ND	ND	
HeCBp	$0.2 \pm 0.1$	$0.05 \pm 0.01$	ND	ND	
Total	$0.6 \pm 0.1$	$0.350 \pm 0.01$	ND	ND	

Mean of three independent determinations. ND, not detected.

#### Table 4

Concentrations of PCB homologues found in each section of the leaching column for S4 soil; leaching solution:  $25 \text{ mg L}^{-1}$  LAS.

Analyte	Concentration of PCBs (mg kg <sup>-1</sup> )				
	2nd section	3rd section	4th section	5th section	
DCBp	0.038 ± 0.006	ND	ND	ND	
ТСВр	0.051 ± 0.07	$0.031 \pm 0.004$	ND	ND	
TeCBp	0.312 ± 0.01	$0.10 \pm 0.01$	ND	ND	
PeCBp	0.559 ± 0.01	$0.407 \pm 0.002$	ND	ND	
НСВр	0.213 ± 0.02	$0.16 \pm 0.01$	$0.06 \pm 0.008$	ND	
HeCBp	$0.39 \pm 0.02$	$0.138 \pm 0.003$	$0.136 \pm 0.001$	$0.10 \pm 0.02$	
Total	$1.57 \pm 0.07$	$0.83 \pm 0.02$	$0.198 \pm 0.008$	0. 10 ± 0.02	

Mean of three independent determinations. ND, not detected.

er compounds were retained in the first sections, and only the heavier compounds, such as HCBp and HeCBp, reached the fourth, with only HeCBp reaching the fifth section. This would confirm the idea that OM plays an essential role in the retention of these compounds. That is, PCBs would be less mobile in soils with a higher OM content. On the other hand, it has been established (Pignatello, 1998; Bogan and Trbovic, 2003; Pan et al., 2006) that higher molecular weight organic compounds diffuse very slowly towards humine, and they are mostly retained in humic and fulvic acids of the matrix. On the contrary, lighter compounds diffuse more readily towards humine, being preferably retained in the matrix. In this context, the LAS present in the leaching solution would interact more readily with heavier compounds, carrying them to the lower sections of the column.

#### 4. Conclusions

The presence of PCBs was detected in the biosolid sample at mean concentrations of  $300 \ \mu g \ kg^{-1}$  and with a predominance of HCBp and HeCBp congeners.

In all the soils amended with spiked biosolids, PCB extraction increased with increasing incubation time, which could be due to organic matter breakdown.

The LAS solution was able to mobilize PCBs through the leaching column, meaning that these detergents present at high concentrations in biosolids could carry PCBs through the different soil strata. It was determined that higher molecular weight compounds showed greater mobility in this medium.

Monitoring the presence of PCBs in biosolids must be constant. Given that these compounds were detected in this matrix, their existence may cause adverse environmental effects.

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