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1 **Feasibility of using low density polyethylene sheets to detect atmospheric**
2 **organochlorine pesticides in Alexandria, Egypt**

3
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13
14 **Abstract**

15
16 Egypt is a major agricultural country in Africa with a known past of organochlorine pesticides
17 (OCPs) application, yet data on atmospheric levels of OCPs in Egypt is sparse. Low density
18 polyethylene (LDPE) passive samplers were therefore deployed for 3 weeks each at 11 locations
19 in July, 2010 and January, 2011 in Alexandria to screen for gas-phase OCPs. Performance
20 reference compounds were used to investigate the uptake kinetics. Field-derived sampler-air
21 partitioning coefficients (K_{PE-A}) for OCPs were significantly correlated against the compounds'
22 subcooled liquid vapour pressure ($\log P_L$): [$\log K_{PE-A} = -0.77 \pm 0.07 * \log P_L + 6.35 \pm 0.13$ ($R^2 =$
23 0.90 ; $n = 17$; $SE = 0.19$; $p < 0.001$)]. Estimated and measured OCP concentrations in Alexandria
24 agreed well (factor difference ≤ 2) indicating the feasibility of monitoring OCPs using LDPEs.
25 OCP concentrations ranged from <LOD to 168 pg/m^3 . Calculated isomeric ratios indicated
26 recent usage of chlordanes and endosulfans.

27
28 **Keywords**

29 Passive sampling; partitioning constants; Egypt; organochlorine pesticides; Sources

30

31 **Capsule Abstract**

32 LDPE passive air samplers spiked with PRCs can be used as a practical low cost technique for
33 monitoring gas phase OCPs in Egypt.

34
35 **1. Introduction**

36
37 Organochlorine pesticides (OCPs) are a group of organic pollutants with high
38 bioaccumulation (Nakata et al., 2002), toxicity (Lemaire et al., 2004; Souza et al., 2004; Tiido et
39 al., 2005; Murvoll et al., 2006), persistence and long range transportation in the environment
40 (Bailey et al. 2000; Li et al. 2007) causing them to be readily detected even in remote regions
41 and pristine areas such as the Arctic (Hung et al., 2010). Despite being banned for decades in
42 Canada, U.S. and European countries, DDTs, hexachlorocyclohexanes (HCHs), chlordanes and
43 dieldrin continue being detected in the ambient air on regional and continental scales (Van
44 Drooge et al., 2002; Jaward et al., 2004; Shen et al., 2004, 2005; Messing et al., 2013).

45 In some developing countries, however, several OCPs are still in use (Baek et al., 2013).
46 Pesticides were introduced in Egypt in 1952 and about one million metric tons of commercial
47 pesticides were used until 2003 (Mansour, 2004). In addition, illegal pesticides application
48 cannot be ignored due to the poor enforcement of environmental laws. In Egypt, 70 % of the
49 pesticides are applied on cotton crops, while the rest are applied on corn, rice, sugarcane
50 plantations, vegetables and fruits (Mansour, 2004). DDT, lindane and endrin were among the
51 pesticides used to control cotton pests. Three decades ago, the use of DDT in agriculture was
52 officially banned. Following the restriction on DDT, the use of other organochlorine pesticides
53 (e.g., aldrin, dieldrin, chlordane, heptachlor, lindane) was gradually restricted in Egypt (Abou-
54 Arab et al., 1995; Mansour, 2004).

55 Despite the fact that Egypt was a signatory of Stockholm Convention held on May, 2002,
56 and the use of OCPs was banned in Egypt, these toxic compounds are still detected in various

57 physical and biological environmental compartments in the country (EL Nemr and Abd Allah,
58 2004; Sallam and Morshedy, 2008; Barakat et al. 2012 a,b; Khairy et al., 2012; Barakat et al.,
59 2013). To our knowledge, limited studies have been carried out to assess the atmospheric levels
60 of OCPs in Egypt, possibly due to the difficulties associated with the conventional active
61 sampling techniques. According to the Stockholm Convention, parties to the Convention should
62 develop a national inventory of POPs. No information exists about the levels, sources and health
63 effects associated with the exposure to atmospheric OCPs in Egypt. Yet frequent measurements
64 of air concentrations in different locations as well as monitoring studies on various levels from
65 local point sources up to the continental scale are a matter of a great importance. Accordingly,
66 cheap techniques that accurately measure atmospheric concentrations of POPs are highly
67 desirable.

68 Passive air samplers have been proven to be a powerful monitoring technique for POPs,
69 as they are cheaper and require less labour compared to active samplers (Lohmann et al., 2001).
70 Polyurethane foam (PUF) disks (Jaward et al., 2004, 2005; Zhang et al., 2008; Devi et al., 2011)
71 and XAD-2 resin (Choi et al., 2008; Wang et al., 2010; Baek et al., 2013) are the most widely
72 used passive air samplers for monitoring OCPs worldwide. Low density polyethylene (LDPE)
73 has been successfully used as passive air samplers for PAHs (Bartkow et al., 2004; Kennedy et
74 al., 2007; Khairy and Lohmann, 2012).

75 The purpose of the current study was to investigate the possibility of using LDPE as
76 passive air samplers for OCPs in the atmospheric environment of Alexandria, Egypt.
77 Accordingly, we undertook two major 21-days sampling campaigns of 15 deployments during
78 summer (July, 2010) and winter (January, 2011) across 11 different sites. By deploying samplers

79 in different locations and in different seasons, we were able to assess the pollution levels,
80 temporal variations and the possible sources of OCPs in Alexandria using isomeric ratios.

81

82 **2. Materials and methods**

83 **2.1. Site Description**

84 Alexandria is the second-largest city of Egypt, with a total surface area of 2300 km² and a
85 population of 4.1 million, extending about 90 km along the coast of the Mediterranean Sea on the
86 northwestern side of the Nile Delta (Figure SI 1). The city's climate
87 shows Mediterranean characteristics, namely mild, variably rainy winters and hot summers that,
88 at times, can be very humid. More details about the study area can be found in Khairy and
89 Lohmann (2012). The land use map of Alexandria (Figure SI 1) indicated the existence of
90 different land uses including the agriculture, which represented ~ 27% of the governorate area
91 and occurred mainly in the south and southeastern parts (Switch, 2011).

92

93 **2.2. Preparation and Deployment of LDPEs**

94 LDPE sheets were cut from commercial sheeting (Carlisle Plastics, Inc., Minneapolis,
95 MN) with a thickness of 51 µm, yielding a 10 x 30 cm strip of ~1-2 g each. Samplers were
96 cleaned with DCM and n-hexane and spiked with four performance reference compounds
97 (PRCs) according to the method developed by Booij et al. (2002), namely 2,5-dibromobiphenyl
98 (PBB 9), 2,2',5,5'-tetrabromobiphenyl (PBB 52), 2,2',4,5',6-pentabromobiphenyl (PBB 103) and
99 octachloronaphthalene (see supplementary data, text SI 1 for more details). Once spiked, PEs
100 were strung on stainless steel wires, placed in precleaned aluminum foil packets, numbered, and
101 frozen in plastic bags until the time of deployment.

102 LDPE sheets were deployed ~ 2 meters above the ground surface in 11 different
103 locations across Alexandria city (Figure SI 1) each inside two inverted bowls providing
104 protection from direct radiation and rainfall. At each location, LDPEs were deployed for 21 days
105 in July, 2010 (summer season) and January 2011 (winter season) (site and deployment details are
106 given in Table SI 1).

107

108 **2.3. Active Air Sampling**

109 Active sampling was performed to validate LDPE as passive air samplers for OCPs (see
110 text SI 1 for more details about the preparation of the active sampling media). Samples were
111 collected using a high volume sampler at the 11 sites in July, 2010 and January 2011 (for
112 meteorological details and sampling volumes, see text SI 2 and Table SI 2). Air was first drawn
113 through a glass fiber filter (GFF) to collect the particulate bound compounds followed by a PUF
114 to retain the vapor phase compounds. Particulate bound data are not presented in the current
115 study.

116

117 **2.4. Extraction of LDPEs and PUFs**

118 LDPEs were cold extracted twice in DCM for 24 hours after spiking with 10 μL of a
119 surrogate standard mixture composed of labeled OCPs ($^{13}\text{C}_6$ -hexachlorobenzene and $^{13}\text{C}_{12}$ -p,p'-
120 DDT; 5 ng/ μL in nonane). Extracts were concentrated to ~1 mL on a rotary evaporator, solvent
121 exchanged to hexane, and concentrated to ~50 μL . Ten μL of 2,4,6-tribromobiphenyl (5 ng/ μL),
122 was added as an injection standard before analysis.

123 PUF samples were extracted using a Dionex ASE 350 (Dionex Corporation, Sunnyvale,
124 CA 94088) accelerated solvent extraction device after spiking with the surrogate standard

125 mixture. Extracts were concentrated to a final volume of ~1 mL using a rotary evaporator (after
126 solvent exchange into hexane) and passed on a glass chromatographic cleanup column packed
127 with silica gel/alumina (2:1 wt/wt) in order to remove the interfering compounds (Khairy and
128 Lohmann, 2012). The collected fraction was concentrated to a final volume of ~ 50 μ L. Finally,
129 2,4,6-tribromobiphenyl was added as the injection standard before analysis.

130

131 **2.5. Instrumental Analysis and Quality Control**

132 All samples were analyzed for OCPs with an Agilent 6890 Series GC System connected
133 to an Agilent 5973 Network Mass Selective Detector. DDTs (p,p'-DDE, o,p'-DDD, p,p'-DDD,
134 o,p'-DDT and p,p'-DDT), hexachlorocyclohexanes (HCHs: α -, β -, γ - and δ -HCH isomers),
135 chlordanes (trans- and cis-chlordane isomers, oxychlordane and trans-nonachlor), heptachlor and
136 its epoxide, aldrin, dieldrin, endosulfans (endosulfan I and II isomers and endosulfan sulfate),
137 hexachlorobenzene (HCBz), endrin, endrin aldehyde, endrin ketone and methoxychlor were
138 investigated. Separation of OCPs was accomplished with a DB-5 MS fused silica capillary
139 column (30 m \times 0.25 mm i.d., 0.25 μ m film thickness, J&W Scientific). Abbreviations of OCPs
140 and more details on the instrumental analysis are given in Table (SI 3) and text (SI 3).

141 Procedural blanks, field blanks, matrix spikes and duplicate samples (20 % of the total
142 samples) were included with each sample batch, and were carried throughout the entire analytical
143 procedure in a manner identical to the samples. OCPs were completely absent from procedural
144 and field blanks indicating negligible contamination during transport, storage and analysis.
145 Limits of detection (LODs) were determined as the concentration of analytes in a sample giving
146 a peak with a signal-to-noise (S/N) of 3 (Doong et al., 2002). (QA/QC procedures are detailed in
147 text SI 4 and Table SI 3).

148 2.6. Physico-Chemical Properties

149 Internally consistent air-water partitioning coefficients (K_{AW}) and octanol-air partitioning
150 coefficients (K_{OA}) were obtained from Schenker et al. (2005). Missing K_{OA} values were obtained
151 by correlating available K_{OA} values from Schenker et al. (2005) against values obtained from EPI
152 Suite (USEPA, 2011) (Figure SI 2). Missing K_{AW} values were calculated according to equation 1
153 (Reinhard and Drefahl, 1999):

$$154 \quad K_{AW} = \frac{H_c}{RT} \quad (1)$$

155 where H_c is Henry's law constant (atm.L.mol^{-1}), R is the gas constant (0.08206 in L.atm/mol.K)
156 and T is the absolute temperature in Kelvin. H_c for δ -HCH was obtained from Meylan and
157 Howard (2005). Values for oxychlordan, trans-nonachlor, o,p'-DDD, o,p'-DDT and endosulfan
158 sulfate were obtained from SPARC online calculator (V4.5). Values for endrin aldehyde, endrin
159 ketone and methoxychlor were obtained from Mackay et al. (2006). Internally consistent values
160 of the sub-cooled liquid vapor pressure (P_L/Pa) were obtained from Shen and Wania (2005).
161 Missing P_L values were obtained by correlating values from Shen and Wania (2005) with those
162 obtained from Mackay et al. (2006) (Figure SI 3). P_L value for trans-nonachlor was obtained
163 from Shoeib and Harner (2002). Values for endrin aldehyde, endosulfan sulfate and endrin
164 ketone were obtained from EPI Suite. Enthalpies of vaporization (ΔH_{vap} in KJ/mol) were
165 obtained from Shoeib and Harner (2002). Missing ΔH_{vap} values were obtained by correlating
166 available values against P_L (Figure SI 4). Sampler-water partitioning coefficients (K_{PE-W}) were
167 calculated according to Lohmann (2012). Values of all the physico-chemical parameters are
168 given in Table (SI 4).

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171 2.7. Estimated atmospheric concentrations based on LDPE

172 PRCs, impregnated in the polyethylene samplers before their deployment, were used to
173 gauge whether OCPs had achieved equilibrium and to adjust for disequilibrium in polyethylene
174 (C_{LDPE}) assuming that uptake and elimination rates are equivalent. (Booij et al., 2002) (Text SI
175 5). Gaseous atmospheric concentrations (C_g , ng/l) were calculated from disequilibrium- corrected
176 polyethylene concentrations (C_{LDPE}) as follows:

$$177 \quad C_g = \frac{C_{LDPE}}{(1 - e^{-k_e t}) \cdot K_{PE-A}} \quad (2)$$

178 where C_{LDPE} is the OCP concentration in the LDPE (ng/kg), k_e is the loss rate constant of PRCs
179 (day^{-1}), t is the deployment period (days) and K_{PE-A} is the sampler-air partitioning coefficient
180 (L/kg). To estimate K_{PE-A} values for OCPs, a high volume sampler was left operating for a
181 period of 15 days (5 days per week) at one of the sampling sites (site 2) during the summer
182 season (See text SI 2 for more details). Detected OCP concentrations in the LDPE and PUF
183 samples were used to calculate the K_{PE-A} values according to equation 2. Partition coefficients
184 were adjusted for temperature according to a modified form of Van't Hoff equation (equation 3)

$$185 \quad K_{PE-A}(T_2) = K_{PE-A}(T_1) \times e^{-\frac{\Delta H_{\text{vap}}}{R} \left(\frac{1}{T_2} - \frac{1}{T_1} \right)} \quad (3)$$

186 where T_2 and T_1 are the mean temperatures of the deployment and at which the partitioning
187 properties were determined (K).

Deleted: and ΔH_{vap} is the enthalpy of vaporization (J mol^{-1})

189 2.8. Uncertainty associated with K_{PE-A} calculations

190 Calculated overall uncertainty combined the uncertainty of predicted partition
191 coefficients, equilibrium adjustment of polyethylene concentrations based on the use of PRCs

192 (the fraction equilibration remaining at time of PE sampler collection), and the uncertainty
193 associated with the analysis of the OCPs (See text SI 6 for more details). K_{PE-A} values for
194 undetected OCPs (β -HCH, δ -HCH, dieldrin, endosulfan sulfate and methoxychlor) in the field
195 study were predicted from a simple linear relationship with P_L (see section 3.2, equation 5).
196 Accordingly, we used the same relative uncertainties (RU) in predicted K_{PE-As} as given for the
197 log P_L by Shen and Wania (2005). Since internally consistent P_L values did not exist for δ -HCH,
198 endosulfan sulfate and methoxychlor, they were assigned the highest uncertainty estimate of P_L
199 (500 %). The overall uncertainty (Table 1) ranged from ± 43.0 % to ± 504 % (0.20-0.80 log
200 units), with higher uncertainty values (303-504 %) for predicted K_{PE-As} compared to the field
201 measured ones (43.0-65.0 %) owing to the greater uncertainty estimates of their P_L .

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Table 1

204 3. Results and Discussion

205 3.1. Uptake of OCPs by the LDPE

206 The addition of PRCs to the samplers before deployment is useful to infer the exchange
207 rate kinetics, assuming that the elimination rates (of PRCs) and uptake rates (of native
208 compounds) are equivalent (Bartkow et al., 2004). Exchange kinetics were quantified by k_e (for
209 details, see text SI 5). Four PRCs with different log K_{PE-A} values were used in this study. The
210 least dissipated PRC in all the investigated samples was pentabromobiphenyl (PBB 103) (Table
211 SI 5). A higher elimination rate was observed for PBB 103 during the winter (30-46 %)
212 compared to the summer (25-33 %). The Kruskal-Wallis one way ANOVA statistical test
213 indicated that loss rate constants decrease with increasing the sampler-air partitioning coefficient
214 (log K_{PE-A}) (PBB 103 < OCN < PBB 52 < PBB 9) in both seasons ($H_{summer} = 39.1$, $H_{winter} = 38.6$

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215 at $\alpha = 0.05$, $p < 0.001$). Moreover, loss rate constants for each of the PRCs were significantly
216 higher in the winter compared to the summer season [Mann-Whitney rank sum test for PBB 9
217 and PBB 103: $T = 66$ and 73 respectively at $p < 0.001$; t-test for PBB 52 ($t = -5.56$ at $p < 0.001$)
218 and OCN ($t = -2.37$ at $p = 0.028$)], which was probably related to the increased wind speed
219 during the winter season (Table SI 1). This indicates that the chemical exchange between the
220 LDPE sampler and the air was limited by the air side boundary layer resistance ($k_e \propto 1/K_{PE-A}$)
221 based on the following relation:

$$222 \quad k_e = \frac{k_a \cdot A_s}{V_s \cdot K_{PE-A}} \quad (4)$$

223 where k_a is the mass transfer coefficient through the air side boundary layer and A_s and V_s are
224 the sampler surface area and volume respectively.

225 A correlation between log transformed k_e values and log K_{PE-A} of the PRCs was
226 established to extrapolate k_e for all the investigated OCPs (Tables SI 6 and 7). At the end of the
227 21 days deployment period, the average values of % equilibrium for OCPs (Figure 1) in the
228 samples indicated that OCPs with higher P_L values were approaching equilibrium (α -HCH, γ -
229 HCH, HCBz and aldrin) (> 80 % equilibrium), whereas OCPs like DDTs were still far from
230 reaching equilibrium (< 60 %). To gain more information about the accumulation patterns of
231 OCPs, three LDPEs were deployed in each season at one of the sites (site 11), and one was
232 harvested after each week of exposure period (Figure 1). The equilibration of α - and γ -HCH,
233 HCBz, aldrin, heptachlor, trans- and cis-chlordane after each week of deployment (week 2 -
234 week 1 and week 3 - week 2) started to deviate from the linear uptake after three weeks
235 deployment period in the summer season (Figure 1a), indicating that they were approaching
236 equilibrium. Contrarily, endosulfan I and II, trans-nonachlor, endrin, oxychlordane, p,p'-DDE
237 and p,p'-DDT increased linearly during the sampling period. During the winter season (Figure

238 1b), only p,p'-DDD/o,p'-DDT, o,p'-DDD and p,p'-DDT increased linearly during the sampling
239 period, whereas all the other detected OCPs were approaching equilibrium. When the
240 approached % equilibrium results of OCPs based on the PRCs at site 11 were compared to the
241 variation in the accumulated amounts of OCPs in the LDPE (ng/g PE) with time (Figure SI 6),
242 good agreement was generally observed for the predicted stage of exchange of OCPs between
243 the LDPE sampler and air in both seasons. This implies that PRCs can be used to calibrate
244 sampler/site specific mass transfer behavior, and thus accurately estimate the gaseous
245 concentrations of OCPs (and other POPs) in the atmosphere.

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246 **Figure 1**

247 **3.2. Sampler-air partitioning constants (K_{PE-A})**

248 PE-A partitioning constants were calculated directly for 17 OCPs which were detected
249 both in the passive sampler (corrected for non-equilibrium) and the PUFs at site 2 during the
250 field deployments (Table 1). To predict values for the other investigated OCPs, we examined
251 correlation between our measured K_{PE-AS} and literature K_{OA} values, which has been used
252 previously to predict K_{PE-AS} for PAHs (Bartkow et al., 2004; Khairy and Lohmann, 2012). As
253 shown in Figure (SI 5), a highly significant and strong linear correlation can be observed for
254 PAHs ($R^2 > 0.90$, $p < 0.001$), whereas for OCPs, the relation explained only 32 % of the total
255 variability in the data.

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256 We observed a significant and strong correlation when K_{PE-AS} were regressed against P_L
257 (Pa) ($p < 0.001$) (Figure 2a). The correlation explained 90.0 % of the total variability in the data.
258 The slope was insignificantly different from -1 ($p < 0.001$; $R^2 = 0.90$), implying that P_L can be
259 used as a good predictor of K_{PE-A} , and that the magnitude of K_{PE-A} for OCPs is dominated by their
260 volatilities. The derived K_{PE-A} were calculated from two 3-week field deployments and

261 constrained by our ability to safely operate active sampling equipment during the entire field
262 work. Yet the strong correlation with P_L implies that we were able to derive values that are well
263 constrained by physico-chemical properties, validating the field-derived data. Accordingly, K_{PE-A}
264 values for the non-quantified OCPs in the passive sampler were predicted (Table 2) based on the
265 derived P_L - K_{PE-A} relation (equation 5):

$$\log K_{PE-A} = -0.77 \pm 0.07 \cdot \log P_L + 6.35 \pm 0.13 \quad (5)$$

(n = 17; R² = 0.90; SE = 0.19)

271 **Table 2**

272 **Figure 2**

273 We propose that K_{PE-A} for the non-quantified OCPs in the field calibration study can be
274 best predicted from equation (5) despite of the higher degree of uncertainty for endrin, endrin
275 aldehyde, endosulfan sulfate, endrin ketone and methoxychlor (Table 1) as internally consistent
276 physico-chemical properties were missing. For further assessment, calculated K_{PE-A} values were
277 compared with K_{PE-A} values derived using sampler-water partitioning coefficients (K_{PE-W}) and
278 air-water partitioning coefficients (K_{AW}) according to equation 6:

$$K_{PE-A} = \frac{K_{PE-W}}{K_{AW}} \quad (6)$$

280 Calculated (from the field experiment) and predicted (K_{PE-W}/K_{AW}) K_{PE-A} values agreed
282 well within 94 % (factor difference < 0.4 log units) for the majority of the investigated OCPs
283 (Figure 2b). The good agreement observed for most OCPs supports the assumption that our field-
284 derived K_{PE-A} values are good approximations of their real values, despite the limited
285 deployment period in the field. Contrarily, lower degree of agreement was observed for endrin

286 aldehyde, endrin ketone, endosulfan sulfate, trans-nonachlor and oxychlorane (factor difference
287 ranged from 0.60-3.8 log units). One explanation for this lower agreement is related to the degree
288 of uncertainty associated with the partitioning coefficients (uncertainty from P_L and K_{AW} +
289 uncertainty from the K_{PE-A} and K_{PE-W} partitioning coefficients).

290

291 3.3. Spatial distribution and seasonal variations of OCPs

292

293 Estimated atmospheric OCP concentrations in Alexandria are given in Tables (3, SI 8 and

294 SI 9). Based on the average concentrations, the overall trend of OCPs showed the following

295 order: HCBz ($78 \pm 46 \text{ pg/m}^3$) > HCHs (sum of α - and γ - isomers: $67 \pm 33 \text{ pg/m}^3$) > chlordanes

296 (sum of cis- and trans- isomers, oxychlorane, trans-nonachlor, heptachlor and its epoxide: 60.5

297 $\pm 75 \text{ pg/m}^3$), aldrin ($59 \pm 48 \text{ pg/m}^3$) > endosulfans (sum of endosulfan I and II: $45 \pm 49 \text{ pg/m}^3$) >

298 DDTs (sum of o,p'-DDD, o,p'-DDT, p,p'-DDE, p,p'-DDD and p,p'-DDT: $34.5 \pm 43 \text{ pg/m}^3$) in

299 the summer season, and HCHs ($82 \pm 29 \text{ pg/m}^3$) > HCBz ($63 \pm 31 \text{ pg/m}^3$) > aldrin (30 ± 31

300 pg/m^3) > chlordanes ($19 \pm 15 \text{ pg/m}^3$) > endosulfans ($15 \pm 19 \text{ pg/m}^3$) > DDTs ($14 \pm 12 \text{ pg/m}^3$) in

301 the winter season. Endrin, endrin aldehyde (EA) and endrin ketone (EK) were only detected at

302 lower concentrations (<LOD-28 pg/m^3) in the summer season and <LOD in all the winter

303 samples. For most of the investigated OCPs, highest concentrations were always observed at

304 sites 2 and 3 (Figure SI 7). These sites were located in the northeastern part of Alexandria,

305 closest to agricultural areas (Figure SI1). In the summer season, concentrations of chlordanes,

306 aldrin, endosulfans, endrin and its metabolites and DDTs were greater at site 2 and site 3,

307 whereas only slight between sample variations were observed for HCHs. A similar trend was

308 observed for aldrin, endosulfans and p,p'-DDE in the winter season. HCBz, in contrast, displayed

309 highest concentrations (23-168 pg/m^3) in both seasons at site 2 and site 3, and other locations

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310 | characterized by high traffic and industrial activities (site 1, 4, 6 and 9). The average HCBz
311 | concentration in Alexandria was only slightly higher than its northern hemisphere average
312 | concentration (~ 50 pg/m³) (Jin et al., 2013), but much lower than measured concentrations in
313 | China (Liu et al., 2009) and Korea (Li et al., 2007) (Table SI 10). Although HCBz production
314 | has ceased in most countries, it is still being generated inadvertently as a by-product and/or
315 | impurity in several chemical processes, such as the manufacture of chlorinated solvents,
316 | chlorinated aromatics and pesticides. It is also released to the environment by incomplete
317 | combustion, and from old dumpsites (Barber et al., 2005).

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318 | **Table 3**
319 | α - and γ -HCH were the only detected HCH isomers in the atmospheric environment of
320 | Alexandria (Figure SI 7). In all the samples, α -HCH concentrations (<LOD - 98.0 pg/m³) were
321 | much higher than the γ -HCH (<LOD - 36.0 pg/m³), especially in the winter season.

322 | Aldrin was detected in 82 % of the investigated PE samples with relatively high
323 | concentrations (<LOD - 147 pg/m³) compared to HCHs and HCBz (Figure SI 7). Chlordanes
324 | were detected in the majority of the investigated samples. Based on the average concentrations,
325 | trans-chlordane showed the highest concentrations in all the samples during both seasons
326 | (summer: 19 pg/m³; winter: 9 pg/m³) followed by cis-chlordane (8.0; 4.0 pg/m³). Oxychlordane,
327 | the metabolite of chlordane, was only observed in the summer season with an average
328 | concentration of 7.0 pg/m³. Heptachlor epoxide, the metabolite of heptachlor was also observed
329 | only in the summer season (higher temperature). The detection of both metabolites only in the
330 | summer suggests that they were produced from the photodegradation of parent compounds (Park
331 | et al., 2011).

332 Endosulfan is still in use globally for agriculture and also for the control of disease
333 vectors (Pozo et al., 2011). Concentrations of endosulfans ranged from 9.0-160 pg/m³ and
334 <LOD-65 pg/m³ in the summer and winter seasons respectively. In all the samples, endosulfan I
335 concentrations were higher than endosulfan II, and endosulfan sulfate was <LOD in all the
336 samples.

337 **p,p'-DDT** was widely used in Egypt for agricultural purposes and for the control of
338 disease vectors. Although officially banned in 1988, its was recently detected in sediments of the
339 Egyptian Mediterranean Sea coast (Khairy et al., 2012) and in coastal lakes (Barakat et al.,
340 2012a, b), because of its long environmental half-life time. In the current study, concentrations of
341 DDTs were lower than those of HCHs, endosulfans, chlordanes and HCBz (Figure SI 7) ranging
342 from 4 - 168 ng/m³ and 4-29 pg/m³ in the summer and winter seasons respectively. p,p'-DDE
343 was the most frequent and abundant isomer in all the samples comprising 48-96 % of the total
344 concentrations of DDTs in Alexandria.

345 No significant seasonal variations were observed for HCBz, chlordanes (trans- and cis-
346 isomers), aldrin and DDTs (p: 0.115-0.861), whereas concentrations of γ -HCH and endosulfans
347 were significantly higher in the summer (p = 0.009-0.048), and α -HCH was significantly higher
348 in the winter (p = 0.035) (Figure SI 7).

349 HCBz concentrations in Alexandria were higher than concentrations measured in Europe
350 but lower than in Asia (Table SI 10). Chlordane concentrations were among the highest when
351 compared to concentrations in other locations worldwide using PUF passive samplers). γ -HCH
352 and endosulfans on the other hand were lower than concentrations measured in parts of Europe,
353 Canada, India, China and in the other African countries such as Ghana and South Africa.

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Comment [RL1]: Didn't you already comment on HCB above? This last paragraph should either be deleted, or you paste the sentences in the relevant sections above.

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357 3.4. Source identification of OCPs

358 Several isomeric ratios were applied (Figure SI 8) in the current study to determine the
359 source of the atmospheric OCPs (HCHs), and whether OCPs were released to the atmosphere as
360 a result of fresh application or an aged use (HCHs, chlordanes, endosulfans and DDTs).

361 In the current study, α -HCH/ γ -HCH ratio ranged from 1.3-2.9 (Figure SI 8) in the
362 summer (hot) season, and 3.2-6.7 in the winter (cold) season indicating a possible seasonal re-
363 volatilization of old lindane residues. Lower ratio values observed in the summer season could
364 be related to the increased lindane volatilization from soil as a result of elevated atmospheric
365 temperatures.

Comment [RL2]: Needs ref – maybe from Bidleman's work??

366 Technical chlordane has been widely used as agriculture pesticide and termiticide in all
367 over the world (Syed et al., 2013). Calculated ratios of trans/cis chlordane in the current study
368 ranged from 1.5-2.9 (Figure SI 8) in both seasons, indicating fresh application of technical
369 chlordane (Hinkley et al., 1990, Jantunen et al., 2000). According to the UNEP, the use of
370 chlordane is severely restricted and limited to non-agricultural uses in Egypt (UNEP, 2003).

371 Endosulfan comprises two isomers, endosulfan I and II, with a I/II ratio of about 2.33
372 (Daly et al. 2007) in the technical mixture. Endosulfan II is more reactive and so an elevated I/II
373 ratio is representative of an aged signature, whereas ratios closer to the starting technical mixture
374 suggest recent application. As shown in Figure (SI 8), calculated values in the current study were
375 around the value of the technical mixture (2.3) indicating recent application of endosulfans.

376 In this study, the ratio of p,p'-DDE/p,p'-DDT ranged from 2-106, indicating an aged
377 application of DDT in the area.

378

379

380 **3.5. Evaluation of the Predictive Ability of the LDPE Samplers**

381 Estimated gaseous OCP concentrations calculated from the LDPE passive samplers
382 corrected for non-equilibrium were compared with the gaseous concentrations actually measured
383 in the atmosphere using the high volume sampler. Results are given in Tables (SI 8 and 9).
384 Although the difference in the sampling periods between the passive (21 days) and active (2–3
385 days) sampling periods adds uncertainty to the results in Alexandria (Egypt), good agreement
386 was generally observed between estimated and measured OCP concentrations in all the
387 investigated samples (Figure 3 a & b). During the summer and winter seasons, the difference
388 ranged from a factor of 0.8 to 2.0. Additionally, slopes of both regression lines were not
389 significantly different from one (at $\alpha = 0.05$, $P < 0.001$) supporting this good agreement. The
390 significant correlations obtained here suggest that the gas-phase OCP concentrations in
391 Alexandria might not display significant short-term variations during our measurement periods.
392 This assumption is supported by the observed statistical insignificant difference ($p = 0.128$)
393 between the weekly measured gas-phase concentrations at site 2 (Figure SI 9) sampled with the
394 conventional active sampling method. Overall, LDPEs have proven to be a powerful tool for
395 predicting atmospheric OCP concentrations in the present study.

396

Figure 3

397 **4. Conclusions**

398 Our results indicated the feasibility of using LDPE as a practical and low-cost technique
399 for monitoring OCPs in Egypt, and to provide data on the spatial distribution and seasonal
400 variations of OCPs in an urban Egyptian city. Including PRCs enables the use of PE sheets as

401 passive air samplers for gas-phase OCPs in general. While our work demonstrated the
402 importance of using PRCs in verifying the equilibrium status of OCPs in PE samplers, a cheaper
403 alternative to circumvent the PRC approach is the use of LDPEs with different surface area-to-
404 volume ratios, as presented by Bartkow et al. (2004). Different passive sampling materials are
405 now popular and their utilization for monitoring organic contaminants in the atmosphere has
406 greatly increased. Future work should concentrate on comparing different passive samplers as a
407 function of deployment times, temperature and wind speed so that data derived from different
408 studies are broadly comparable, and the levels of confidence around the data are known.

409 Estimated concentrations of atmospheric OCPs in Alexandria, Egypt were within the
410 global range as measured in the global atmospheric passive sampling network (GAPS). Yet the
411 detection of OCPs in the Egyptian environment, especially those with fresh sources as indicated
412 by the isomeric ratios questions the efficacy of the official ban of most of the investigated OCPs
413 in Egypt from decades. This could encourage decision makers in Egypt (and beyond) to utilize
414 this cost-effective sampling tool for developing an extensive local air sampling network. With
415 the use of passive samplers, data can be acquired about sources and concentrations of organic
416 pollutants and to develop effective control measures.

417 **Supplementary information**

418
419
420 Supplementary data associated with this article can be found in the online version.

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426

427 **5. References**

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