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# Human Health Risk Assessment of Environmental Exposure to Organochlorine Compounds in the Catalan Stretch of the Ebro River, Spain

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**Abstract** In this study, the environmental impact and human health risks associated with exposure to organochlorine compounds (OCs) through soils and tap water in the Catalan stretch of the Ebro River, Spain, were investigated. The concentrations of polychlorinated biphenyls, hexachlorocyclohexanes, as well as DDT and derivates, were determined. Relatively low levels of these pollutants were observed, with mean concentrations ranging between 0.51–315.8 µg/kg and 0.05–74.6 ng/L in soil and tap water, respectively. These values are similar to those found in a number of recent surveys over the world. In spite of the presence of a chlor-alkali plant located upstream the Ebro River, which could mean a potential source of pollution, the current levels of OCs should not mean significant additional health risks for the local population.

**Keywords** Organochlorine compounds · Soils · Tap water · Human health risk assessment
Ebro River (Catalonia Spain)

Modern industrial and agricultural practices have meant an increased use of a wide number of chemicals. These are often produced in the proximity of freshwater systems, and their breakdown products and by-products generated as part of the production process finally end up into water and/or soils. The Ebro River basin constitutes an important agricultural area in Catalonia (NE Spain). In addition to agriculture, other anthropogenic activities are also important in that area. Among these, industries and sewage treatment plants are of notable concern taking into account the potential adverse impacts on water quality and local soils (Terrado et al. 2006). The environmental contamination in the Catalan basin of the Ebro River, as well as the influence of the chlor-alkali plant located in Flix (Tarragona Province, Catalonia), have been studied by a number of investigators (Ramos et al. 1999a, b; Lacorte et al. 2006; Navarro et al. 2006). Historically, the Flix reservoir has been receiving residues dumped by a chlor-alkali plant.

It is well-known that anthropogenic activities may generate pollution in air, soil, and water. The existence of potentially toxic chemicals such as organochlorine compounds (OCs), metals, and radionuclides in water, sediments, and soils was previously reported in the Flix reservoir, and the lower part of the Ebro River (Ramos et al. 1999a, b). Humans are exposed to OCs mainly through occupational activity, environment, and dietary intake. The purpose of the present study was to assess the human health risks derived from the environmental exposure to OCs through soils and drinking water for the population living in the Catalan stretch of the Ebro River and its Delta at the Mediterranean Sea.

## Materials and Methods

In spring of 2006, surface soil samples were collected at 10 different villages/towns located in the riparian zone of the Ebro River in Catalonia. Samples consisted of a composite

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of three subsamples collected at: (a) village center, (b) recreational areas, and (c) the riverside. Sampling methodology was recently reported (Ferré-Huguet et al. 2007). On the other hand, in spring and autumn of 2006, 10 municipal tap water samples were obtained from public fountains, where the Ebro River recharges the aquifers. Drinking water samples were collected in clean amber glass bottles, according to the methods of the American Public Health Association (2005). The location of sampling points was divided into two zones: north and south, near and downstream the Flix chlor-alkali plant, respectively (Fig. 1).

In all water and soil samples, total concentrations of the following compounds were determined: α-, β-, γ- and δ-hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), pentachlorobenzene (PeCB), the environmental marker PCB-congeners No. 28, 52, 101, 118, 138, 153, and 180, $o,p'$- and $p,p'$-DDE, DDD, and DDT. For drinking water, suspended particles were retained with glass microfiber filters. Those present in the dissolved phase were concentrated by solid phase extraction (C18 disks). The concentrations of OCs in water were analyzed by GC-ECD (Agilent Technologies, HP-6890N, Palo Alto, CA, USA) (Grimalt et al. 2004). In turn, the same compounds (HCHs, HCB, PeCB, PCBs, and DDTs) were extracted from soils

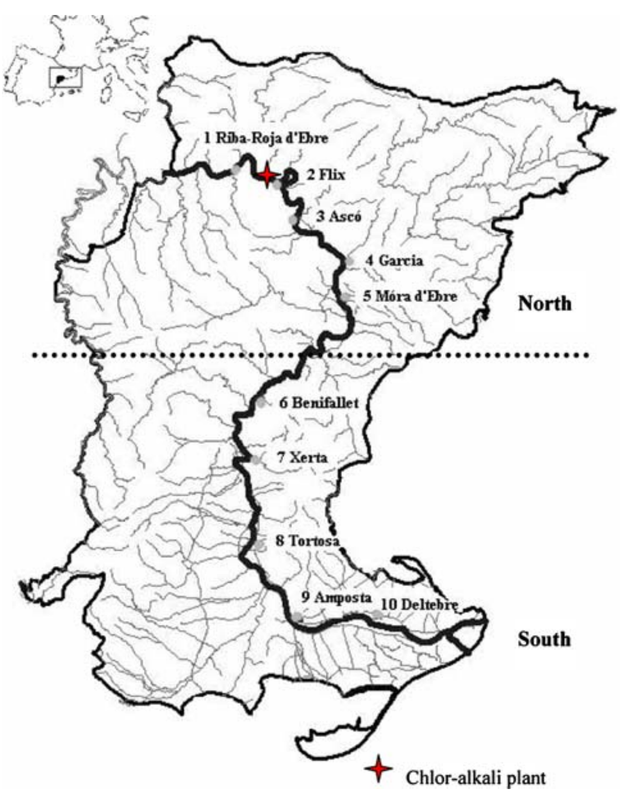


**Fig. 1** Area of sampling

and analyzed by GC-ECD. A capillary DB5 column was used for all analyses (Vilanova et al. 2001).

Human health risks potentially associated to the ingestion and dermal absorption of OCs through water and soil particles were assessed for children and adults. As data concerning OC concentrations in air are not currently available, inhalation was not considered. Monte-Carlo simulations were separately applied to calculate probabilistically carcinogenic and non-carcinogenic risks. The criteria used for calculations were taken from the US EPA (1999), as well as other recent studies (Ferré-Huguet et al. 2009). Crystal Ball 4.0© software was used for running simulations, which were based on an iteration size of 10,000. The Hazard Quotient (*HQ*) was calculated by comparing the predicted exposure with the oral or dermal reference dose for each OC. *HQ* is an estimation of the systemic toxicity potentially posed by a single chemical within a single route of exposure (non-cancer risk). The Hazard Index (*HI*) was calculated by summing the *HQ* for each exposure pathway, and normalizing according to the number of assessed pollutants. Finally, the environmental hazard index ($HI_{env}$) was calculated by summing the *HI* for each route of exposure.

The chemical-specific excess cancer incidence (ELCR), derived from the ingestion or the dermal absorption, was calculated by multiplying the daily exposure and the oral or dermal slope factor, respectively. The total pathway-specific excess cancer incidence for all chemicals within a single exposure route was calculated as the sum of ELCR to all chemicals for a single route. Finally, total excess cancer incidence posed by all chemicals over all routes was calculated as the sum of the ELCR for all evaluated routes.

## Results and Discussion

The OC concentrations in water and soil samples collected in the area under the current evaluation are summarized in Table 1. The highest mean concentrations in tap water corresponded to PCBs (0.075 µg/L), followed by DDTs and DDEs (0.045 and 0.032 µg/L, respectively). These levels were compared with those established by the Royal Decree 140/2003 for water intended for human consumption (BOE 2003) and with the safety levels established by the US EPA (2009). In any case, OC concentrations in drinking water did not exceed those threshold values. In addition, the levels of the analyzed OCs were within the safety range, being similar to those found in other Spanish areas (Eljarrat et al. 2007), as well as in rural and urban zones over the world (Na et al. 2006; Said et al. 2008; Turgut and Gokbulut 2008; Turgut et al. 2009). However, the comparison of different studies is frequently difficult because of the variability and uncertainty of water

**Table 1** Concentration of OCs in soil (µg/kg) and drinking water (ng/L) in 10 locations along the Catalan stretch of the Ebro River, Spain

| Sample | PeCB | PCBs | HCB | HCHs | DDEs | DDDs | DDTs |
|---|---|---|---|---|---|---|---|
| Soil | | | | | | | |
| 1 | 0.017 | 4.37 | 0.32 | 0.60 | 4.91 | 0.55 | 1.81 |
| 2 | 3.653 | 216.75 | 116.17 | 1.88 | 44.36 | 18.18 | 54.31 |
| 3 | 1.236 | 124.67 | 7.41 | 0.04 | 46.56 | 43.38 | 117.83 |
| 4 | 0.056 | 128.63 | 1.82 | 0.27 | 38.98 | 14.71 | 116.47 |
| 5 | 0.101 | 45.74 | 3.93 | 1.53 | 58.23 | 25.03 | 62.37 |
| 6 | 0.002 | 3.72 | 0.09 | 0.10 | 2.15 | 1.92 | 8.30 |
| 7 | 0.029 | 9.72 | 0.06 | 12.44 | 1037.88 | 1560.64 | 2740.62 |
| 8 | 0.011 | 11.02 | 0.29 | 0.18 | 9.51 | 5.43 | 4.66 |
| 9 | 0.012 | 25.95 | 0.11 | 0.21 | 3.86 | 2.80 | 5.51 |
| 10 | 0.040 | 10.22 | 0.67 | 0.51 | 13.85 | 9.43 | 46.43 |
| *Mean* | *0.516* | *58.08* | *13.09* | *1.78* | *126.03* | *168.21* | *315.83* |
| LOD | 0.002 | 0.05 | 0.03 | 0.01 | 0.01 | 0.02 | 0.02 |
| Drinking water | | | | | | | |
| 1 | nd | 0.88 | nd | 0.03 | 0.12 | 0.05 | 0.17 |
| 2 | 0.22 | 3.36 | 0.223 | 0.03 | 0.20 | 0.22 | 0.61 |
| 3 | 0.60 | 5.63 | 0.184 | 1.73 | 14.41 | 1.39 | 4.64 |
| 4 | 0.10 | 75.61 | nd | 3.20 | 19.69 | 10.80 | 29.84 |
| 5 | 0.14 | 155.66 | 0.040 | 0.11 | 96.34 | 25.22 | 145.36 |
| 6 | 0.07 | 197.98 | nd | 2.32 | 91.32 | 25.72 | 40.65 |
| 7 | 0.08 | 214.63 | nd | 5.47 | 51.87 | 34.03 | 57.50 |
| 8 | 0.05 | 12.73 | nd | 0.07 | 5.47 | 12.75 | 50.74 |
| 9 | 0.19 | 12.04 | nd | 0.11 | 1.50 | 0.54 | 1.76 |
| 10 | 0.44 | 67.33 | nd | 0.04 | 46.56 | 43.38 | 117.83 |
| *Mean* | *0.19* | *74.58* | *0.054* | *1.31* | *32.75* | *15.41* | *44.91* |
| LOD | 0.04 | 0.03 | 0.025 | 0.03 | 0.03 | 0.03 | 0.03 |

*nd* non-detected, *LOD* limit of detection

treatment methods. In the current survey, the southern area presented higher levels of DDT, DDD, and DDE, possibly due to a repeated application of DDT in crop fields throughout the 50, 60, and 70 decades of the last century. However, only HCB concentrations were significantly higher in the northern area ($p < 0.05$). These findings are probably related to the relatively higher concentrations found in the Flix village (northern area), where a chlor-alkali plant is located.

Regarding urban soils, the highest concentrations of OCs corresponded to DDT, DDD, and DDE (316, 168, and 126 µg/kg dw, respectively). The levels of PeCB, HCB, and PCBs were significantly higher in the northern zone ($p < 0.05$), whereas those of HCHs, DDT, DDD, and DDE significantly increased in the lower part of the Ebro River ($p < 0.05$). A special "hot-spot" of pollution by DDT and degradation products was particularly detected in sample No. 7. It is well-known that DDT and its two main metabolites are highly persistent and bioaccumulative. These compounds were frequently used for the control of plagues in that agricultural area. In Spain, restrictions in DDT use were established in 1977. Notwithstanding, the use of DDT is still allowed as an intermediate in the production of dicofol in Monzón (Ebro River basin, Aragon, NE Spain) since 1987 (de la Cal et al. 2008). The concentrations of OCs in soils were compared with the levels for the remediation of potentially contaminated soils marked by the Royal Decree 9/2005 (BOE 2005), as well as the Preliminary Remediation Goals (PRGs) established for residential soils by the US EPA (2009). The current levels were lower than those marked in these guidelines.

Among the different compounds analyzed, the highest contribution to OC exposure corresponded to PCBs and DDT. In order to assess the potential health risks due to the presence of OCs in drinking water and soils, *HQ* and *HI* were calculated. Table 2 summarizes these values for children and adults. For drinking water, the individual *HQ* by water ingestion in children and adults was less than 1 for all compounds. On the other hand, the risk index by dermal absorption of soil was higher than the ingestion. In summary, the total *HI* due to OC environmental exposure through soils and water, considering the two different exposure pathways evaluated (ingestion + dermal absorption) was $0.05 \pm 0.15$ and $0.04 \pm 0.14$ for children and adults, respectively. The great variability associated with *HI* was basically due to the wide range of concentrations.

**Table 2** Distributions (mean and standard deviation) of hazard quotient (*HQ*) and hazard index (*HI*) through ingestion and dermal absorption of drinking water and urban soil for the population living in the Catalan stretch of the Ebro River, Catalonia, Spain

| | Drinking water | | Urban soils | |
|---|---|---|---|---|
| | Ingestion | Dermal | Ingestion | Dermal |
| *HQ* | | | | |
| Children (c) | | | | |
| PeCB | 1.1E-05 ± 4.5E-05 | 1.0E-05 ± 2.9E-05 | 2.6E-07 ± 1.2E-03 | 1.2E-04 ± 4.5E-04 |
| Aroclor 1016 | 1.4E-04 ± 4.2E-04 | 4.1E-04 ± 1.0E-03 | 1.1E-04 ± 7.3E-05 | 5.1E-02 ± 1.4E-01 |
| Aroclor 1254 | 2.6E-03 ± 6.4E-03 | 7.9E-03 ± 1.8E-02 | 3.7E-04 ± 8.1E-04 | 1.7E-01 ± 5.0E-01 |
| HCB | 2.9E-06 ± 1.3E-05 | 7.0E-06 ± 2.4E-05 | 7.3E-07 ± 2.5E-05 | 6.0E-03 ± 3.3E-02 |
| ∑HCHs | 1.9E-05 ± 6.8E-05 | 2.6E-06 ± 7.9E-06 | 5.3E-07 ± 1.2E-06 | – |
| ∑DDE | – | 1.1E-05 − 2.8E-05 | – | – |
| ∑DDD | – | – | – | 4.3E-03 − 2.1E-02 |
| ∑DDT | 5.9E-06 ± 2.3E-05 | 2.6E-05 ± 8.1E-05 | 6.2E-05 ± 6.0E-04 | 3.6E-02 ± 1.8E-01 |
| Adults (a) | | | | |
| PeCB | 9.9E-06 ± 4.2E-05 | 9.2E-06 ± 2.7E-05 | 2.4E-07 ± 1.1E-03 | 1.1E-04 ± 4.2E-04 |
| Aroclor 1016 | 1.3E-04 ± 3.9E-04 | 3.8E-04 ± 9.5E-04 | 9.8E-05 ± 6.8E-05 | 4.7E-02 ± 1.3E-01 |
| Aroclor 1254 | 2.4E-03 ± 6.0E-03 | 7.3E-03 ± 1.7E-02 | 3.4E-04 ± 7.5E-04 | 1.6E-01 ± 4.6E-01 |
| HCB | 2.7E-06 ± 1.2E-05 | 6.5E-06 ± 2.3E-05 | 6.8E-07 ± 2.3E-05 | 5.6E-03 ± 3.1E-02 |
| ∑HCHs | 1.8E-05 ± 6.3E-05 | 2.4E-06 ± 7.3E-06 | 4.9E-07 ± 1.1E-06 | – |
| ∑DDE | – | 1.0E-05 ± 2.6E-05 | – | – |
| ∑DDD | – | – | – | 4.0E-03 ± 1.9E-02 |
| ∑DDT | 5.4E-06 ± 2.2E-05 | 2.4E-05 ± 7.5E-05 | 5.7E-05 ± 5.6E-04 | 3.3E-02 ± 1.7E-01 |
| *HI* | | | | |
| | $HI_{ing}$ c *4.6E-04 ± 1.2E-03* | $HI_{der}$ c *1.4E-03 ± 3.3E-03* | $HI_{ing}$ c *8.9E-05 ± 4.5E-04* | $HI_{der}$ c *4.5E-02 ± 1.5E-01* |
| | $HI_{ing}$ a *4.2E-04 ± 1.1E-03* | $HI_{der}$ a *1.3E-03 ± 3.0E-03* | $HI_{ing}$ a *8.3E-05 ± 4.2E-04* | $HI_{der}$ a *4.2E-02 ± 1.4E-01* |
| | $HI_{env}$ c *4.7E-02 ± 1.5E-01* | | | |
| | $HI_{env}$ a *4.3E-02 ± 1.4E-01* | | | |

*HQ* hazard quotient, *HI* hazard index, *ing* ingestion, *der* dermal, *c* children, *a* adults, *env* environmental

Therefore, the current OC concentrations in water and soils of the Catalan basin of the Ebro River should not mean a significant non-carcinogenic risk for both adults and children.

Of the 19 target chemicals, carcinogenicity values have been reported for all OCs excepting for PeCB. The risk distribution through ingestion and dermal absorption for adults is shown in Table 3. The cancer risk of OCs exposure through water in the Ebro River basin was estimated to be about $3.8 \times 10^{-6} \pm 6.9 \times 10^{-6}$ and $2.3 \times 10^{-6} \pm 3.6 \times 10^{-6}$ through ingestion and dermal absorption, respectively. Although in order to get the maximum protection for human health, the concentration of these compounds should be obviously zero, lifetime risks of $10^{-6}$ to $10^{-4}$ are considered acceptable for carcinogens in drinking water (US EPA 1999). In turn, the cancer risk of OCs exposure was estimated to be $3.1 \times 10^{-5} \pm 3.1 \times 10^{-4}$ and $3.1 \times 10^{-5} \pm 1.2 \times 10^{-4}$ through ingestion and dermal absorption of soil, respectively. The carcinogenic risk associated with soil exposure was more relevant than that of concerning water intake and absorption. Overall, that value means a total multipathway cancer risk of $6.8 \times 10^{-5} \pm 4.4 \times 10^{-4}$. The carcinogenic risk derived from the evaluated pathways greatly exceeded the generally acceptable risk level for (individually considered) chemicals of $10^{-6}$, being slightly below $10^{-4}$. However, the uncertainty associated with these values is very high. Thus, they must be considered as preliminary values. Therefore, a wide study of the environmental levels of OCs, especially in soils, is clearly necessary. More specific values of non-cancer and cancer risks must be achieved by increasing the number of samples.

This study belongs to a wider investigation aimed at assessing the health risks derived from environmental and dietary exposure to a number of chemicals for the population living near the Ebro River in Catalonia (Ferré-Huguet et al. 2008; Nadal et al. 2008). Recently, the HI and cancer risks due to metal exposure for that population were found to be $0.42 \pm 2.59$ and $2.1 \times 10^{-4} \pm 8.8 \times 10^{-4}$, respectively (Ferré-Huguet et al. 2009). Although individually, probably these substances do not mean additional health risks, an integral exposure to a mixture of these

**Table 3** Cancer risk distribution (mean ± standard deviation) derived from the environmental exposure to OCs through drinking water and soils

| | Ingestion | Dermal absorption |
|---|---|---|
| Drinking water (dw) | | |
| PeCB | – | – |
| Aroclor 1016 | 1.5E-08 ± 2.9E-08 | 2.6E-07 ± 4.0E-07 |
| Aroclor 1254 | 2.3E-06 ± 3.7E-06 | 1.4E-06 ± 1.9E-06 |
| HCB | 7.8E-08 ± 2.1E-07 | 2.0E-07 ± 4.8E-07 |
| ∑HCHs | 1.3E-06 ± 2.8E-06 | 1.1E-07 ± 2.1E-07 |
| ∑DDE | 4.1E-08 ± 7.7E-08 | 1.6E-07 ± 2.7E-07 |
| ∑DDD | 6.4E-08 ± 1.1E-07 | 1.1E-07 ± 1.4E-07 |
| ∑DDT | 2.1E-08 ± 4.8E-08 | 9.7E-08 ± 2.1E-07 |
| Urban soils (us) | | |
| PeCB | – | – |
| Aroclor 1016 | 6.60E-06 ± 1.80E-05 | 6.60E-06 ± 1.80E-05 |
| Aroclor 1254 | 6.30E-06 ± 1.80E-05 | 6.30E-06 ± 1.80E-05 |
| HCB | 7.20E-06 ± 4.00E-05 | 7.20E-06 ± 4.00E-05 |
| ∑HCHs | 6.40E-07 ± 1.80E-05 | 6.40E-07 ± 1.80E-06 |
| ∑DDE | 2.60E-06 ± 9.40E-05 | 2.60E-06 ± 9.40E-06 |
| ∑DDD | 1.90E-06 ± 9.20E-05 | 1.90E-06 ± 9.20E-06 |
| ∑DDT | 5.70E-06 ± 2.80E-05 | 5.70E-06 ± 2.80E-05 |
| | $CR_{ing}$ dw 3.8E-06 ± 6.9E-06 | $CR_{der}$ dw 2.3E-06 ± 3.6E-06 |
| | $CR_{ing}$ us 3.1E-05 ± 3.1E-04 | $CR_{der}$ us 3.1E-05 ± 1.2E-04 |
| | $CR_{env}$ dw 6.1E-06 ± 1.1E-05 | |
| | $CR_{env}$ us 6.2E-05 ± 4.3E-04 | |
| | *ELCR 6.8E-05 ± 4.4E-04* | |

*CR* cancer risk, *ELCR* excess cancer incidence, *ing* ingestion, *der* dermal, *env* environmental

chemicals should be carried out. Potential synergisms and antagonisms among the chemical agents should be considered. In a subsequent study, the health risks of exposure to radionuclides will be also characterized.

In conclusion, the results of the present survey show that tap water and residential soils in the Catalan stretch of the Ebro River, present relatively low OC concentrations. These levels are inside the safety ranges according to the Spanish legislation. Moreover, the results from the risk assessment indicate that the current levels of OCs should not mean significant additional non-carcinogenic and carcinogenic risks for the local population.

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