

DISTRIBUTION OF ORGANOCHLORINE PESTICIDES IN PINE NEEDLES OF AN OCEANIC ISLAND: THE CASE OF TENERIFE (CANARY ISLANDS, SPAIN)

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Abstract. The distribution of persistent organochlorine pesticides (HCB, HCH, DDT and its metabolites) was studied by analyzing pine needles in the oceanic island of Tenerife (Canary Islands, Spain). Tenerife is a subtropical oceanic island, characterized by the presence of a high mountain (Pico de Teide, 3,718 m a.s.l.) and by a peculiar climatic condition. The southern side is warm and sometimes during the year influenced by dry winds coming from Africa, while the northern side is cool and wet, with an almost persistent cloudiness (called 'mar de nubes'), due to winds coming from the northern Atlantic. Furthermore a developed agricultural area is present at sea level. Pine needle samples were collected on both sides of the island, from the sea level up to the upper level of pine distribution (2,100 m a.s.l.). The distribution patterns of organochlorine pesticides seem to be affected mainly by local emissions. Up to the considered altitude layer, the influence of global circulation of persistent organic pollutants seems to play a negligible role. The behavior of chemicals is consistent with their physical-chemical properties, with some influence due to climatic conditions.

Keywords: climatic characteristics, Kaw, persistent organic pollutants, temperature dependence

1. Introduction

Organochlorine (OC) pesticides are persistent organic chemicals (POPs), with marked biomagnification properties. During 1960s and 1970s they were extensively utilized in order to protect crops and to prevent health disease. Nowadays they are banned or restricted in the majority of industrialized countries, but they are still used in Africa, South Asia, Central and South America. The levels and environmental fate of OC pesticides have been extensively studied (Atlas and Giam, 1981, 1988; Tanabe *et al.*, 1982; Iwata *et al.*, 1993; Simonich and Hites, 1995; Macdonald *et al.*, 2000). Particularly, numerous studies pointed out that leaves, lichens, mosses and tree bark can bioaccumulate such substances from the troposphere and highlighted the possibility of using vegetable biomass for monitoring tropospheric contamination and for understanding the environmental cycling of OC pesticides (Reischl *et al.*, 1987; Bacci *et al.*, 1988; 1997; Villeneuve *et al.*, 1988; Ericksson *et*



al., 1989; Calamari *et al.*, 1991, 1994; Jensen *et al.*, 1992; Tremolada *et al.*, 1993; Sinkkonen *et al.*, 1995; Simonich and Hites, 1995; Wenzel *et al.*, 1997). Most OC pesticides are semivolatile organic chemicals with vapor pressures roughly ranging between 1 and $10E^{-7}$ Pa at ambient temperature (Bidleman, 1988). Because of this characteristic, the evaporation and condensation processes are basically controlled by ambient temperature. In other words, there is a tendency for these substances to evaporate from warmer regions and to condense in colder environments (Rappe, 1974). On a global scale, through long-range atmospheric transport, there is a systematic transfer of such compounds from warmer to colder regions in a process referred as 'global distillation' or 'cold condensation' (Ottar, 1981). Wania and Mackay (1993) developed the theory of 'global fractionation' that enhanced the concept of global distillation. Although the general mechanisms involved in the environmental cycling of OC pesticides are almost clear, there is still a lack of knowledge of how particular environmental conditions can influence the biogeochemical cycle of OC pesticides. For instance in a regional scale the atmospheric conditions such as air temperatures, prevailing wind directions, or the presence of natural barriers (i.e. mountains) could influence OC cycling and consequently their presence in that particular ecosystem.

In this work the case of Tenerife Island (Canary, Spain) has been studied, due to the particular interest of its orographic conformation, climate and specific geographic position.

2. Description of the Site

Tenerife is the largest island of the Canary archipelago with 2,036 km². It is pyramid shaped, with a truncated apex at 2,000 m a.s.l. (above sea level) at Las Cañadas, from which volcano Teide rises, peaking at 3,718 m, to form the highest point in the archipelago. Tenerife has a steep relief, formed by the presence of the mountain Teide, the highest mountain of Spain, which splits the island into two slopes (South and North side) that are significantly different in terms of their climatic regime. The factors influencing this differentiation are:

1. humid and cool trade winds coming from the North-East;
2. the cold marine current around the Canaries;
3. a thermic inversion layer at about 1,500 m altitude;
4. orography.

Due to the height and orientation of the mountain systems, the orography presents two opposite situations. One corresponds to the North and Northeast slopes, where trade winds help to maintain a humid climate, and the other to southern and western slopes, protected from these winds, where climate is drier and more arid.

The main environmental differentiation on Tenerife is caused by the existence of a temperature inversion at about 1,500 m, separating the lower layer of humid, cool air from a higher layer of dry warmer air. Due to this inversion the lift of air

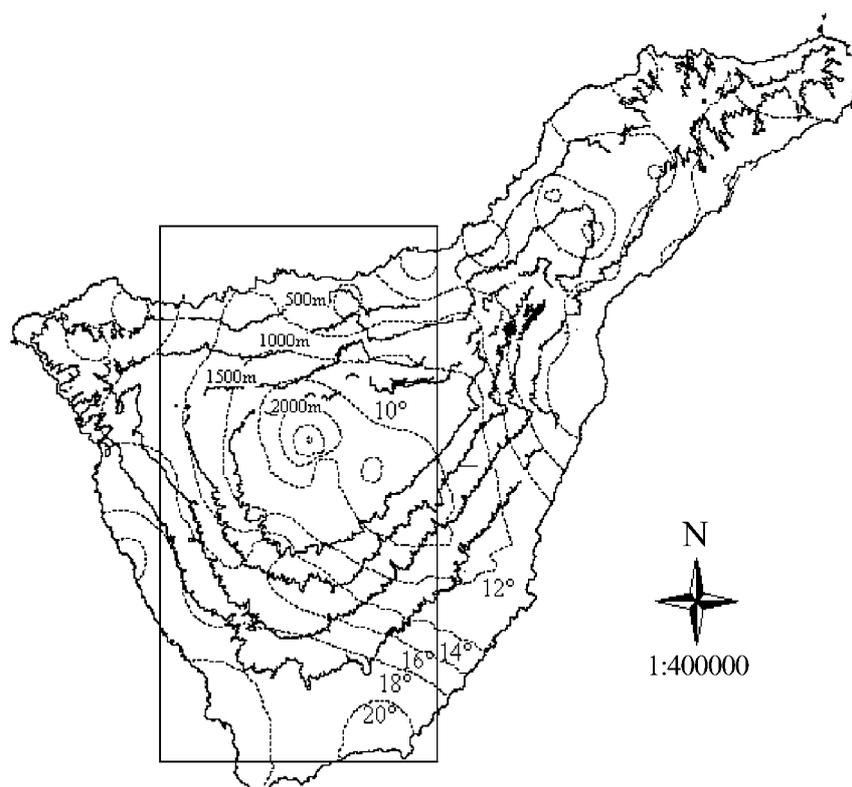


Figure 1. Schematic representation of annual mean temperature (dotted lines represent 2 °C isotherms) of Tenerife Island. The box shows the area where pine needles samples were collected.

masses carried by the trade winds is prevented and leads to the accumulation of clouds below the inversion. This phenomenon, which can be seen on the windward slope of the island, is locally known as ‘mar de nubes’ (cloud sea).

There is a strong correlation between altitude and temperature, and the north side of the island is cooler than the south side. In Figure 1, a detailed representation of annual mean temperature is reported.

Schematically, six main levels along the altitudinal profile, up to the upper pine limit, can be described as follows:

1. Matorral costero (coastal thicket): from the coastal line up to 300 m a.s.l on the windward slope, while on the leeward slope the upper limit of this coastal vegetation fringe reaches 450 m a.s.l.
2. Bosque termófilo (transition thermophilic forest): fringe located above the coastal thicket reaching 400 m a.s.l. on the windward slope and 700 m on the downward slope.
3. Monteverde (green forest): Appears only in the windward slope in a fringe from 600 to 1,200 m a.s.l. just where the northern cold humid trade winds

'alios' lift the mountain chain 'dorsal' (therefore the wettest fringe on the island due to the horizontal precipitation from the 'mar de nubes').

4. Pinar (Pine forest): This forest rings the highest points of the island (i.e. Las Cañadas and Teide) called the forest crown, with a lower limit at 1,200 m a.s.l. on the northern windward and 700 m a.s.l. on southern leeward. The upper limits reach 2,100 and 2,300 m a.s.l. respectively. During the winter there is an incidence of the 'mar de nubes' in the pine forest fringe, when these clouds reach the higher altitudes.
5. Matorral de cumbre (summit thicket): from the upper limit of the pine forest up to 2,500 m a.s.l. with more extreme conditions (daily temperature variations greater than 13 °C), there is a thicket vegetation rich in endemic species.

Furthermore, both sides of the Island (particularly the South) present developed agricultural areas. Agriculture in Tenerife can be divided into two types: the first is dynamic and more developed, with the aim of exportation, while the second is more traditional for local consumption and the domestic market.

The more developed agriculture drastically changed the landscape below 300 m a.s.l. in the past and is still ongoing today. The main impact is the building of large terraces to get flat agricultural surfaces filled with more productive soils transported from other locations, usually from higher altitudes. The use of greenhouses covering the terraces is a common practice today. The main crops grown in this fringe are bananas and tomatoes, both for exportation. Agriculture in Tenerife for domestic market or local consumption consists of three main crops: potatoes, wine grapes and fruit orchards. These crops are located mostly in a fringe between 300–800 m a.s.l.

In spite of the lack of precise data on pesticide use, DDT and, to lesser extent, lindane were used extensively for at least 30 yr.

3. Material and Methods

3.1. SAMPLING AREA

Thirty-five samples of canary pine (*Pinus canariensis* Sweet) needles were collected from sea level up to 2,100 m a.s.l. (pine altitudinal limit) on both sides of Tenerife Island during a three day sampling campaign in April 1998. *Pinus canariensis* is an endemic tree, typical of Canary islands and is the most common conifer species. In Tenerife, it forms extensive woods from 700 to 2,100 m a.s.l., but small groups or isolate trees can be found up to the sea level. Freshly fallen pine needles were collected at the soil surface under the trees. Collecting freshly fallen needles provides biological material at the end of the life cycle, with an homogeneous and enough long exposure period (for *Pinus canariensis* about 20–24 months). Each sample represents needles from an individual tree. Samples were carefully collected at the surface of the needle-bed by avoiding needles damaged

or in contact with soil. Samples were immediately wrapped in aluminum foil and closed in glass jars. At the end of the sampling day they were stored at $-20\text{ }^{\circ}\text{C}$ until analysis was performed.

3.2. CHEMICALS STUDIED AND ANALYTICAL METHOD

Pure α -, γ -hexachlorocyclohexane (HCH), hexachlorobenzene (HCB), o,p'-DDD, o,p'-DDE, o,p'-DDT and p,p'-DDD, p,p'-DDE, p,p'-DDT were purchased from Supelco, Inc.. Stock solutions were made of hexane (1 mg ml^{-1}), which were then diluted according to requirements. Tetrabromobenzene ($>99\%$ purity from Aldrich Chem.) was used as internal standard. Selected physical-chemical properties of OC pesticides, together with their temperature dependence, are reported in Table I. Due to the lack of a complete database of physical chemical characteristics, the two isomeric forms of DDT and its metabolites are supposed to have the same values for the selected characteristics.

Samples were Soxhlet extracted with n-hexane (8 h). The extract was cleaned with sulphuric acid and purified by Florisil columns and subsequently by Silica gel. The final extract was analyzed by a Perkin-Elmer 8500 gas chromatograph with a splitless injector and electron capture detector (ECD). Samples of $1\mu\text{l}$ were injected into a 25 m HP-5 column (0.32 mm i.d., $0.52\mu\text{ m}$ film thickness). The column program was: initial temperature $100\text{ }^{\circ}\text{C}$ hold 2 min, $20\text{ }^{\circ}\text{C min}^{-1}$ to $140\text{ }^{\circ}\text{C}$, $4\text{ }^{\circ}\text{C min}^{-1}$ to $200\text{ }^{\circ}\text{C}$ hold 13 min, $6\text{ }^{\circ}\text{C min}^{-1}$ to $280\text{ }^{\circ}\text{C}$ hold 14 min. Injector and detector temperatures were 250 and $350\text{ }^{\circ}\text{C}$ respectively. The procedure described has been checked for recoveries and for reproducibility. The mean recovery was 85% and the reproducibility was calculated on triplicate analysis giving an overall error of 15%. Detection limits were as follows: 0.01 ng g^{-1} d.w. for HCB and for both HCHs, 0.1 ng g^{-1} d.w. for all the DDTs.

3.3. STATISTICAL TREATMENT OF DATA

In order to evaluate the variability of OC concentration within and along the two mountainsides, Principal Component Analysis (PCA) and the statistical analysis of the means (t-test) were performed using the XLSTAT (vers. 4.2) and SPSS (SPSS x Windows vers. 3.1) software.

4. Results and Discussion

The concentrations of OC pesticides in *Pinus canariensis* needles sampled at different altitudes on both sides of the island are reported in Table II. In case of more than one sample for the same altitudes, the geometric mean is reported. DDD concentrations were generally very low, often below the detection limit; therefore these compounds were not included in the elaborations.

TABLE I

Partition coefficients (octanol/water, air/water and octanol/air) of the studied chemicals at different temperatures (a = Paschke and Schüürmann, 1998; b = Bahadur *et al.*, 1997; c = De Bruijn *et al.*, 1989; d = log K_{aw} (H/RT) is derived from Kucklick *et al.*, 1991; e = calculated from Ten uischer *et al.*, 1992; f = log K_{aw} (VP/S/RT) is calculated from Hinckley *et al.*, 1990 (VP) and Shiu *et al.*, 1990 (S); g = Harner and Mackay, 1995; h = calc. by the authors (K_{ow}/K_{aw})

	Log K _{ow}				Log K _{aw}				Log K _{oa}			
	5 °C	15 °C	25 °C	35 °C	5 °C	15 °C	25 °C	35 °C	5 °C	15 °C	25 °C	35 °C
α-HCH	3.92(a)	3.84(a)	3.79(a)	3.76(a)	-4.16(d)	-3.83(d)	-3.51(d)	-3.22(d)	8.08(h)	7.67(h)	7.30(h)	6.98(h)
γ-HCH	3.85(a)	3.77(a)	3.72(a)	3.72(a)	-4.39(d)	-4.11(d)	-3.85(d)	-3.60(d)	8.24(h)	7.88(h)	7.57(h)	7.32(h)
HCB	5.74(b)	5.06(b)	4.94(b)	4.79(b)	-2.47(e)	-2.00(e)	-1.64(e)	-1.46(e)	7.83(g)	7.34(g)	6.88(g)	6.45(g)
DDE			6.95(c)			-2.64(f)	-2.45(f)	-2.24(f)			12.4(h)	
DDD			6.21(c)			-2.86(f)	-2.61(f)	-2.36(f)			8.82(h)	
DDT			6.91(c)			-2.85(f)	-2.48(f)	-2.14(f)	11.02(g)	10.53(g)	10.07(g)	9.64(g)

TABLE II

Geometric mean of OC concentrations (ng g^{-1} dry wt.) measured in *Pinus canariensis* needles sampled at Tenerife Island (North and South sampled sites). (n.s.: not sampled)

Altitude (m.s.l.)	n	HCB	α -HCH	γ -HCH	o,p'-DDE	p,p'-DDE	o,p'-DDD	p,p'-DDD	o,p'-DDT	p,p'-DDT
SOUTH										
0-100		n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.
200-300	1	0.54	0.64	0.69	0.64	1.31	< 0.1	0.23	0.59	3.34
300-400	4	0.41	0.65	0.83	1.51	4.28	0.07	0.64	2.34	7.28
400-500	2	0.44	0.41	0.91	2.61	3.86	< 0.1	0.51	2.97	12.56
800-1,000	1	< 0.01	0.54	0.46	0.53	1.41	< 0.1	0.24	0.39	2.66
1,000-1,100	3	0.41	0.65	0.35	1.16	2.05	0.07	0.36	0.84	3.63
1,300-1,400	1	0.32	1.50	0.94	0.28	1.55	< 0.1	0.22	0.25	1.74
1,400-1,500	2	< 0.01	0.57	0.4	0.08	0.57	0.03	0.09	0.11	0.69
1,900-2,000	1	0.31	0.93	0.12	0.06	0.31	< 0.1	< 0.1	< 0.1	1.46
2,000-2,100	2	0.40	1.2	0.08	0.06	0.29	< 0.1	0.7	0.20	1.38
NORTH										
0-100	1	0.52	0.74	0.93	1.78	2.21	0.06	0.35	0.63	3.45
200-300	4	0.30	0.88	2.08	1.91	1.64	0.05	0.42	0.73	3.98
300-400	2	0.37	1.45	1.99	1.09	1.42	< 0.1	1.00	0.49	5.47
400-500		n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.
800-1,000	2	0.28	0.29	0.37	2.04	1.15	0.04	0.16	0.73	6.30
1,000-1,100	3	0.58	0.89	0.80	1.49	0.88	0.08	0.22	0.68	5.29
1,300-1,400	2	0.59	1.47	0.59	0.08	0.06	< 0.1	< 0.1	< 0.1	0.91
1,400-1,500	2	0.21	0.68	0.24	< 0.1	< 0.1	0.04	0.11	0.02	0.23
1,900-2,000	1	0.32	0.98	0.26	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.28
2,000-2,100	1	0.36	1.53	0.23	0.02	0.17	0.04	0.48	0.49	0.50

In order to investigate the variability of OC levels among all the sampled areas, a PCA was performed on the log-transformed concentrations of the analyzed compounds.

The PCA elaboration is reported in Figure 2, where the 1st and the 2nd component account for 79% of the total variation (58% axis 1 and 21% axis 2).

The dispersion range of samples in the ordination diagram reflects the altitudinal profile of the mountain. All samples above 1300 m a.s.l. fall in the left part of the graph, characterized by the prevalence of α -HCH (79% of correlation), while DDT isomers and their metabolites, which present high correlation with the first axis (95% o,p'-DDE, 92% p,p'-DDE, 90% o,p'-DDT and 89% p,p'-DDT of correlation), play a major influence on lower altitude samples (below 1300 m a.s.l.), mainly falling in the right part of the graph. In particular, among lower samples, the highest levels of DDT and DDE were measured between 300 and 500 m a.s.l. on the south

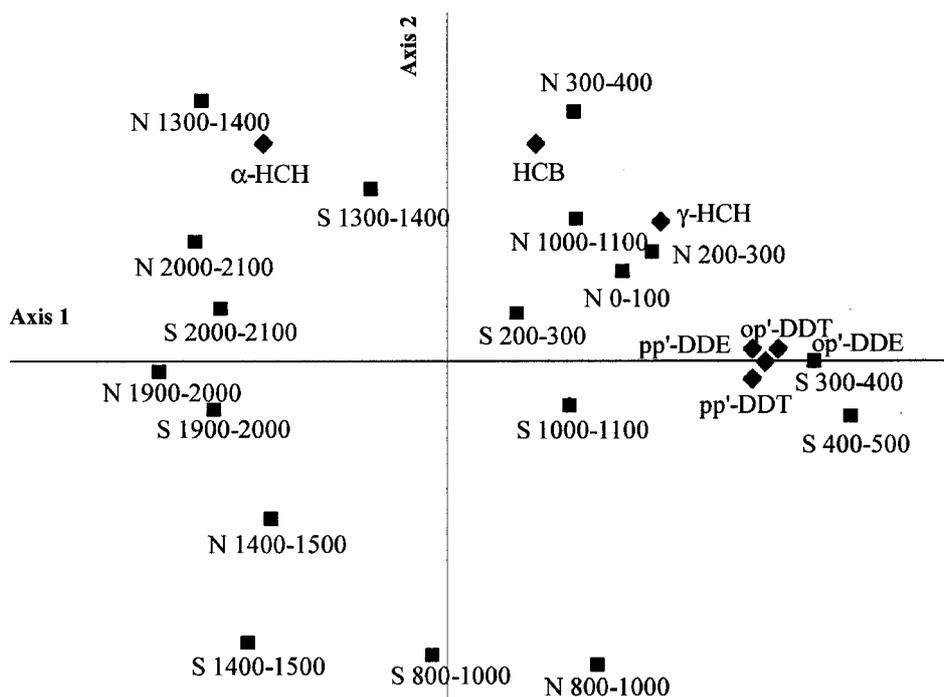


Figure 2. Principal Component Analysis of log transformed OC concentrations in Tenerife pine needles is shown by using the first two components which account for 79% of the total variation (58% axis 1; 21% axis 2). Samples are represented with a black square, while chemicals with black rhomb.

side (at the right end of axe 1), while high levels of γ -HCH (63% of correlation with the first axis) were measured on the northern side (0-400 m a.s.l.), where lindane is presently used.

Independently from the mountain orientation, samples collected between 1,100 and 2,100 m a.s.l. are closely plotted indicating a high degree of similarity. On the contrary, samples collected in the lower part of the mountain (0-1,100 m a.s.l.) are more widely scattered along both the axes, suggesting a different fingerprint of contamination between the North and South side of Tenerife.

On this basis the following comments can be made:

1. PCA indicated no differences among samples collected above the 1,100 m a.s.l. independently of the mountainside. Considering the different origin of winds which affect the northern and southern part of Teide mountain, it can be concluded that the influence of long range transport is negligible at least up to the sampled altitude;
2. the differences highlighted for the lower sample could be attributed to local use and to differences in climatic conditions as consequences of the mountainside orientation.

In order to describe the influences of local conditions (i.e. climate and usage) on the environmental distribution of OC pesticides, the results obtained for HCB, HCHs and DDTs will be briefly discussed below.

4.0.1. *Hexachlorobenzene (HCB)*

HCB concentrations, ranging from < 0.01 to 0.59 ng g^{-1} dry wt (Table II), are homogeneously distributed all over the island. These values are generally lower if compared with literature data of Europe (Calamari *et al.*, 1994, Simonich and Hites, 1995). However, these values seem to be at least one order of magnitude higher than the levels found in Africa (Tremolada *et al.*, 1993). It should be noted that HCB levels measured in Africa refer mainly to mango leaves samples, so a direct comparison could be misleading (Okenden *et al.*, 1998).

The statistical analysis (t-test $P > 0.05$) indicated no significant differences among all the sampled sites: neither according to mountainside orientation nor considering different altitudes. Its relatively low concentrations and its homogeneous distribution all over the island seems to indicate the low relevance of recent sources of emission. It should also be considered that, among all OC pesticides, HCB shows the highest volatility with a K_{aw} ranging from $3.4E^{-03}$ at $5 \text{ }^{\circ}\text{C}$ to $3.45E^{-02}$ at $35 \text{ }^{\circ}\text{C}$ (Table I). HCB shows a strong temperature dependence of K_{aw} (there is a decrease of more than one order of magnitude from 35 to $5 \text{ }^{\circ}\text{C}$). Nevertheless, even at the lowest temperatures typical of the study area, HCB volatility is still higher in comparison with DDTs at sea level, and more than one order of magnitude higher in comparison with HCHs. Therefore, it could be hypothesized that HCB reached an equilibrium of distribution on the island, at least up to the sampled altitudes.

4.0.2. *Hexachlorocyclohexanes (HCHs)*

The levels of both HCH isomers (Table II) are generally lower in comparison with those reported for Europe, where the concentrations in pine needles range between 0.09 and 19.4 ng g^{-1} dry weight for α -isomer and between 0.11 and 27 ng g^{-1} dry weight for γ (Jensen *et al.*, 1992; Reisch *et al.*, 1987; Sinkkonen *et al.*, 1995; Calamari *et al.*, 1991, 1994; Wenzel *et al.*, 1997; Okenden *et al.*, 1998).

Actually lindane is used in some places on Tenerife, mainly in the northern slope, on vegetables and tomato crops.

Both isomers generally show higher concentrations on the northern side of the island, however statistical analysis (t-test) indicates that these differences are not significant ($P > 0.05$).

Up to $400\text{--}500 \text{ m a.s.l.}$, independently of the side of the island considered, γ -HCH concentrations are higher than those measured for α -HCH. On the contrary, α -HCH is more prevalent at higher altitudes. This represents an increase of the α/γ ratio as function of the altitude.

The α/γ ratio has been widely utilized as an indicator of the use of technical mixtures or for identifying old/new contamination, frequently associated with long-range transport phenomenon (Atlas and Giam, 1988). Literature reports contrasting

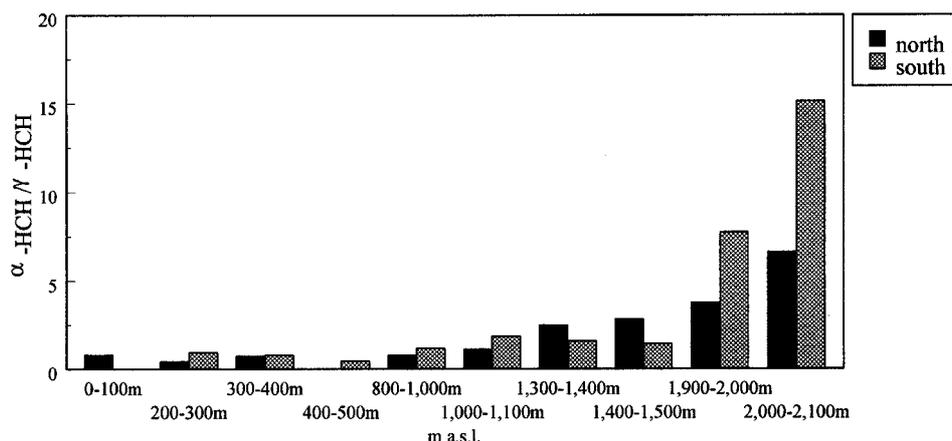


Figure 3. α -HCH/ γ -HCH ratios at different altitude in Tenerife Island.

opinions about the use of this ratio for identifying old or new contamination (Steinwardter, 1978; Oehme and Mano, 1984; Ballschmiter and Wittlinger, 1991; WHO, 1991).

The trends of α/γ ratio for both sides of the Island (Figure 3) are generally comparable and related with different altitudes. PCA suggested that a contribution of α -HCH due to the long range transport phenomenon should be excluded, then the mechanisms ruling the environmental cycling of HCHs should be identified in the local environmental conditions.

Figure 4 shows the temperature dependence of K_{aw} for both HCHs isomers; α -HCH, with a deeper slope, seems to be more influenced by temperature. In fact it is evident that K_{aw} differences between the two substances increase with temperature. Consequently the volatilization processes of such substances are also different as function of the environmental temperatures.

It may be hypothesized that, in the range of 15–35 °C, α -HCH has a higher diffusive capability, that is reflected in an increase of the α/γ ratio. This hypothesis seems to be supported by the inversion of the α/γ ratios that occurs at different altitudes (1,000–1,100 and 800–1,000 m for North and South respectively). In fact, as reported above, Tenerife Island is characterized by differences in climate, with the North characterized by lower temperature and by the presence of the ‘mar de nubes’ (800 – 1,000 m a.s.l.).

Differences in temperatures as function of the different mountainside orientation, altitude and of moisture-laden air layer (mar de nubes) could be responsible for the different distribution capability of HCHs isomers and consequently for the slowing down in the inversion of α/γ ratio on the north side of the Island.

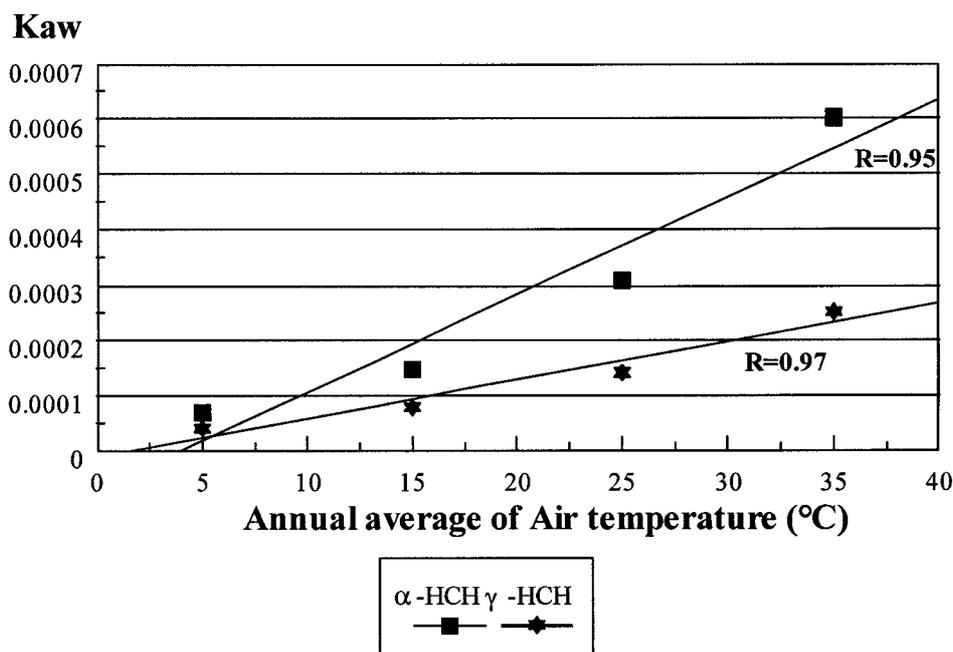


Figure 4. Kaw temperature dependence for α - and γ -HCH.

4.0.3. DDTs

In all samples p,p'-DDT and its metabolite p,p'-DDE are the most abundant OC pesticides, independently of the sides and altitude considered. The levels reached by these two compounds are generally higher by an order of magnitude in comparison with the other analyzed compounds. Particularly in the South (300–500m a.s.l.) p,p'-DDT reaches the highest concentration as the probable result of its agricultural use in banana and tomato crops. In the south of Tenerife the incidence of pests is greater than in the north due to differences in climatic and soil conditions. Nevertheless, t-test revealed ($P > 0.05$) no significant differences for both p,p'-DDT and p,p'-DDE between North and South side of Tenerife.

Figure 5 reports the altitudinal trends of contamination referred to the sum of DDTs and DDEs for each mountainside. It seems evident that the southern profile of concentration shows a sharp increase of Σ DDT and Σ DDE concentration up to 400–500 m a.s.l. followed by a quick decline in the upper part of the sampled sites, while in the north part Σ DDT and Σ DDE show more homogeneity of concentration up to 1,000–1,100 m a.s.l. and a rapid decline in the upper part. These findings suggest the influence of the mountainside orientation in ruling the environmental distribution of OC pesticides. Indeed, the warmer southern climate, seems slightly enhance distribution of such chemicals along the altitudinal profile of the mountain, whereas, on the northern side, due to a colder climate and the

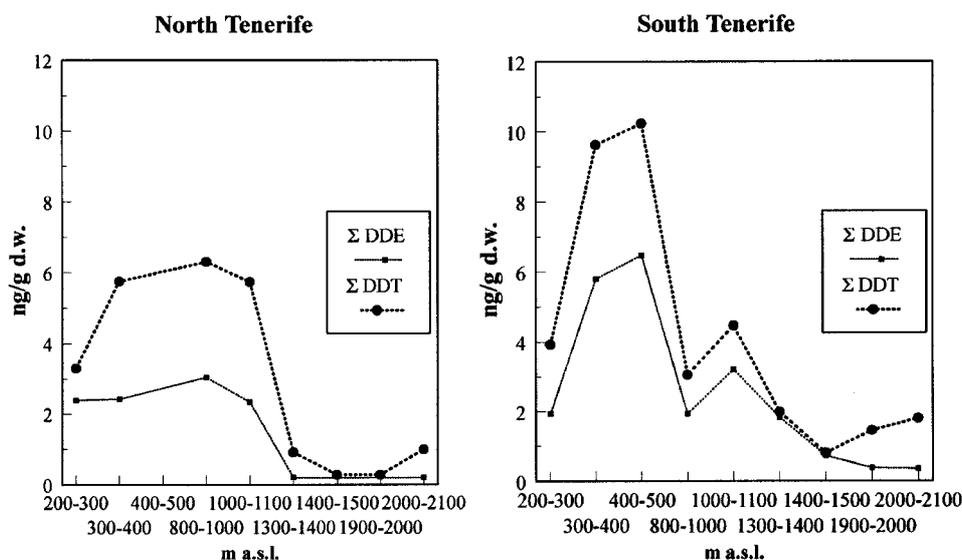


Figure 5. Levels of DDTs and DDEs at different altitudes for both sides of Tenerife Island.

presence of the 'mar de nubes', their capability to move up over 1,000–1,100 m a.s.l. seems to be reduced.

The ratios $\Sigma \text{DDE}/\Sigma \text{DDT}$, reported in Figure 6, show a different trend on the two sides. The South is characterized by an increase up to 1,300–1,400 m a.s.l., while the North shows a progressive decline with higher altitudes. The reasons for these different trends are difficult to explain; they should not be related to differences in volatilization capability because such substances show comparable K_{ow} independently from the temperature considered (Table I). A possible hypothesis could be a different DDT half-life in the South characterized by more incidence of sunlight. Higher DDT persistence on the northern side could also be associated with aerosol adsorption at the level of the 'mar de nubes'.

5. Conclusions

The distribution of organochlorine pesticides in Tenerife Island seems more affected by local contamination than by long-range transport, at least within the altitude range examined (up to 2,100 m a.s.l.). To verify if a 'cold condenser' effect on the global circulation of these chemicals may occur, as observed in tropical and subtropical mountains (Calamari *et al.*, 1991), other environmental matrices should be analyzed up to the top of the mountain.

In general, chemical behavior appears in substantially good agreement with physical-chemical and persistence properties. The high volatility of HCB may account not only for the equilibrium reached in the sampled area, but probably

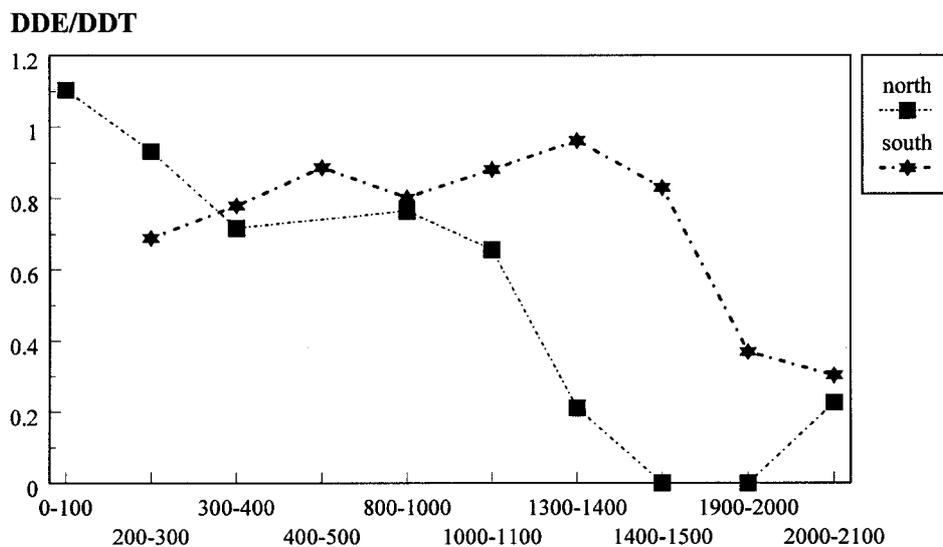


Figure 6. Trends of DDE/DDT ratio on the two mountainsides.

also for the relatively low levels measured in comparison with other comparably developed European sites. Indeed, tropical and subtropical climate may enhance emission of volatile POPs toward global circulation.

Differences between the two mountainsides, though slight and generally not statistically significant, have been observed for some distribution patterns of HCHs and DDTs. Thus, an effect of climatic differences may be hypothesized, even if not definitely demonstrated.

Finally, DDE/DDT ratio, which may be assumed as an indicator of the age of the contamination, is around or below the lower limit of the range characteristic for European countries, typically between 0.6 and 2 (Calamari *et al.*, 1994). Thus, a relatively recent use of DDT could be hypothesized.

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