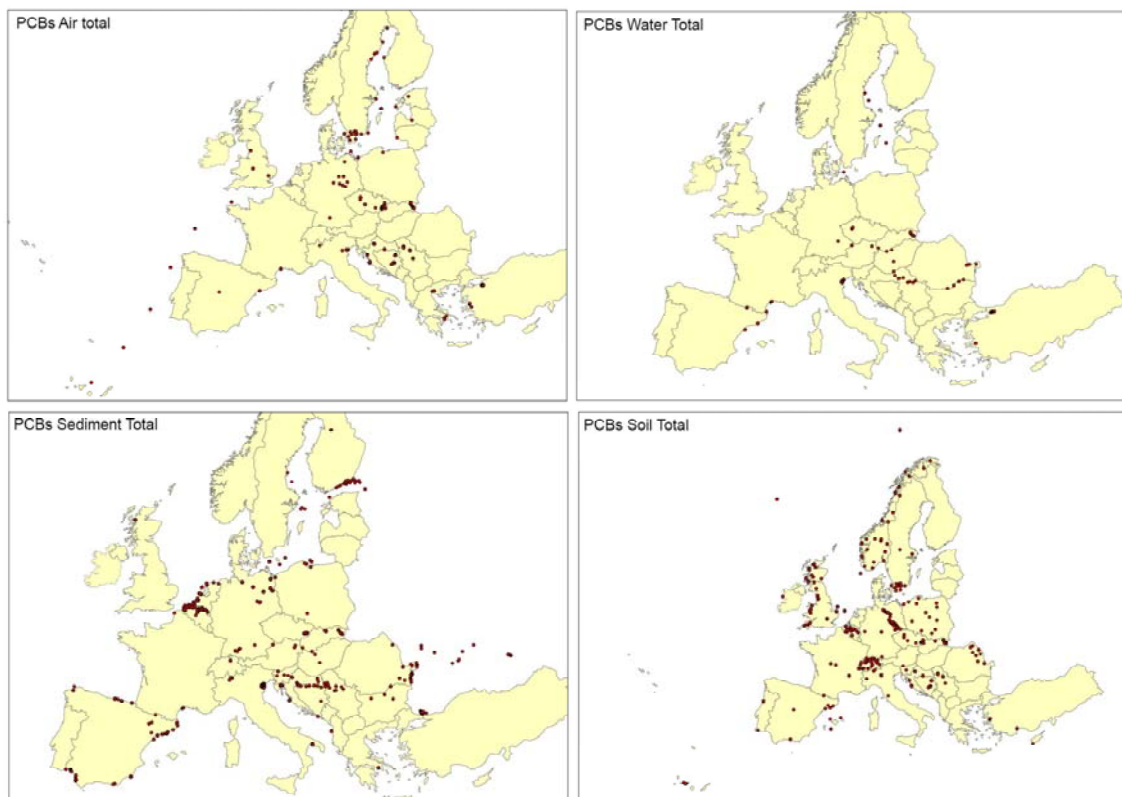




A compilation of Europe-wide databases from published measurements of PCBs, Dioxins and Furans

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1. Introduction

Chemical risk assessment always entails an evaluation of expected environmental concentrations of substances; these concentrations may be predicted using more or less sophisticated mathematical models (predicted environmental concentrations or PECs) or may be chosen on the basis of experimental observations and monitoring activities. The latter are usually relatively expensive, as they require sampling and laboratory analyses. The former are usually relatively inexpensive, especially when using simple back-of-the-envelope calculations as is current practice in the early, more generic stages of risk assessment. When using a mathematical model, however, observations are still fundamental in evaluating whether predictions are realistic and cannot be avoided; on the contrary, the more observations are available, the stronger confidence can be built in a model that proves its ability to reproduce them correctly.

Models tend to be more and more spatially explicit and to include probabilistic assessment moving from the concept of a single representative or worst case PEC, to a frequency distribution of PECs. Spatially explicit models provide site specific information and allow capturing spatial trends, but need to be tested with data in turn equally reflecting spatial variations. Only a few cases exist where spatially explicit models have been tested with observations, in particular the model capability to reproduce the spatial pattern of observed concentrations (Pistocchi et al., 2009).

The difficulties in accurately matching observations and predictions has lead to the attitude of jointly interpreting data and models, instead of relying only on an alternatively “model-” or “observation-based” approach to chemical assessment.

Despite the importance of observation data, most of the substances of potential concern for chemical risk assessment are not routinely monitored. Programmes providing routine monitoring of chemicals in Europe include some marine protection conventions (e.g. HELCOM in the Baltic: <http://www.helcom.fi/>, OSPAR in the North-East Atlantic: www.ospar.org, the Barcelona convention (MEDPOL) in the Mediterranean Sea; <http://www.unepmap.org/>), national water monitoring under the European Water Framework Directive 60/2000/EC, international conventions such as the Stockholm convention on persistent organic pollutants (POPs) (www.pops.int) and the UNECE LRTAP Protocol on POPs (<http://www.unece.org/env/lrtap/>), and the AMAP (Arctic monitoring and assessment programme: <http://www.amap.no/>). All these programmes cover partly overlapping spatial domains, with different temporal frequency and different target environmental media. Data are gradually brought to centralized repositories managed at European level by the European Environmental Agency (EEA), EMEP, or other organizations but the availability and comparability of data is still an issue.

In parallel to monitoring programmes, a wealth of observation data on POPs as well as other chemicals of concern is daily published in scientific papers in many international journals. This information is relatively cheap to obtain, and usually undergoes processes of peer reviewing before publication that ensure, in principle, a reasonable level in the quality of the data. Published papers are therefore an attractive and easily accessible source for monitoring data. On the other hand, observations published in journals are sparse, complex in interpretation or simply not intercomparable, and specific to areas of interest for individual case studies. These facts make scientific literature reporting on pollutants concentrations in the environment rather heterogeneous in terms of analytical techniques, spatial and temporal density of samples and environmental matrixes sampled, and unevenly distributed in space. As such, they can not be easily used in chemical risk assessment.

In this work, we discuss the usefulness and limitations in building compilations of published monitoring data, by describing a feasibility study on polychlorinated biphenyls (PCBs) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), for which observed concentration data were retrieved from scientific journal articles published between

2000 and 2009. These chemicals are typical POPs and are of constantly high concern for their potential adverse effects on human health and ecosystems. PCBs were mainly used by the power industry in electrical transformers, capacitors, hydraulic equipment, and as lubricants. These compounds were also added to many consumer products, like adhesives, waxes and inks. Since the mid 1970s PCBs have been banned from active use in most countries due to their toxicity and extreme persistence. However, sediments of rivers, lakes, and oceans, and soils of temperate areas, as well as marine waters are primary locations for PCB accumulation in the environment and, therefore, themselves can act as emission sources if environmental conditions change.

PCDD/Fs are formed as unintentional by-products of chemical manufacturing and incineration processes, as well as natural processes such as volcanic eruptions, and biomass burning. Emissions from incineration of industrial wastes such as metal reclamation and domestic heating (especially in central Europe) are considered as current sources of PCDD/Fs to the environment.

There are 209 PCB congeners, with a range of physical-chemical properties which is rather representative of POPs in general, and 210 PCDD/F congeners, also spanning a relatively wide range of physico-chemical properties. However, both types of substances are non-polar, with a high affinity for carbon (organic matter), low volatility and solubility. Therefore, they tend to absorb or adsorb to suspended particulate matter in air and water, to sediments, to the organic fraction of soils, and to lipids.

A database of published observed concentrations of PCBs and PCDD/Fs in air, soil sediments and water was built as described in details in the following chapters. As continental scale assessment is aimed more at the evaluation of the regional distribution of contamination, we excluded from the database those measurements taken close to known PCB or PCDD/F pollution sources of exceptional entity such as waste incinerators or contaminated sites.

After presenting and interpreting the results of the literature search, we critically examine the completeness and usability of this information, and generally speaking the usefulness of data compilations in the framework of chemical risk assessment.

2. Data acquisition method

Many local scientific studies measure PCBs and/or PCDD/Fs concentrations in the environment. Sometimes these studies are published, and the values measured or monitored are published in the reports. The method for acquiring the data consists in searching for this kind of data inside these articles, recording them in a database when they are relevant.

The articles were searched on the Scopus® website (www.scopus.com), by different combination of keywords. The keywords used were country name, “dioxin”, “pcb”, and media (“soil”, “water”, etc). They were normally combined like the following examples:

- (dioxin OR pcb) AND (soil OR water OR sediment OR air)
- (dioxin OR pcb) AND (soil OR water OR sediment OR air) AND Croatia

In the database, we also included data easily available from JRC technical reports or anyway activities performed at the JRC IES RWER Unit, for which references are provided on a case by case basis. A particularly important source of information for the Danube catchment is the so called JDS2, Joint Danube Survey 2 (<http://www.icpdr.org/jds/>) to which the JRC contributed.

The following table summarizes the different searches that were performed in Scopus, considering both dioxins and PCBs.

ISO_2DI GIT	COUNTRY_NAME	PCDD - Dioxins				PCDF - Furans				PCBs			
		So il	Ai r	Wat er	Sedimen ts	So il	Ai r	Wat er	Sedimen ts	So il	Ai r	Wat er	Sedimen ts
AL	Albania	X	X	X	X	X	X	X	X	X	X	X	X
AT	Austria	X	X	X	X	X	X	X	X	X	X	X	X
BE	Belgium	X	X	X	X	X	X	X	X	X	X	X	X
BA	Bosnia & Herzegovina	X	X	X	X	X	X	X	X	X	X	X	X
BG	Bulgaria	X	X	X	X	X	X	X	X	X	X	X	X
HR	Croatia	X	X	X	X	X	X	X	X	X	X	X	X
CY	Cyprus	X	X	X	X	X	X	X	X	X	X	X	X
CZ	Czech Republic	X	X	X	X	X	X	X	X	X	X	X	X
DK	Denmark	X	X	X	X	X	X	X	X	X	X	X	X
EE	Estonia	X				X							
FI	Finland	X				X							
FR	France	X	X	X	X	X	X	X	X				
DE	Germany	X				X							
GR	Greece	X	X	X	X	X	X	X	X	X	X	X	X
HU	Hungary	X	X	X	X	X	X	X	X	X	X	X	X
IE	Ireland	X				X							
IT	Italy	X				X							
LV	Latvia	X	X	X	X	X	X	X	X	X	X	X	X
LT	Lithuania	X	X	X	X	X	X	X	X	X	X	X	X
LU	Luxembourg	X	X	X	X	X	X	X	X	X	X	X	X
MK	FYROM	X	X	X	X	X	X	X	X	X	X	X	X
MT	Malta	X	X	X	X	X	X	X	X	X	X	X	X
ME	Montenegro	X	X	X	X	X	X	X	X	X	X	X	X
NL	Netherlands	X	X	X	X	X	X	X	X	X	X	X	X
NO	Norway	X				X							
PL	Poland	X	X	X	X	X	X	X	X	X	X	X	X
PT	Portugal	X				X							
RO	Romania	X	X	X	X	X	X	X	X	X	X	X	X
RS	Serbia	X	X	X	X	X	X	X	X	X	X	X	X
SK	Slovakia	X	X	X	X	X	X	X	X	X	X	X	X
SI	Slovenia	X	X	X	X	X	X	X	X	X	X	X	X
ES	Spain	X	X	X	X	X	X	X	X	X	X	X	X
SE	Sweden	X				X							
CH	Switzerland	X				X							
TR	Turkey	X	X	X	X	X	X	X	X	X	X	X	X
UK	United Kingdom	X				X							

Table 1: Searches performed on Scopus®.

The literature search that has been conducted was restricted by the resources available for this study. As such, it cannot claim to be a complete survey of what is available from the literature. However, it gives a good indication of what is realistically achievable for a chemical within reasonable time limits of e.g. a couple of man-months.

The database was built as a Microsoft Office Excel® file, with a single table. In this table the following data is stored for each piece of data published in the retrieved literature (each of the retrieved references usually displays more than one sampling location):

- Reference to the article the measurement is taken from
- Country of sampling
- Sampling site
- Start / end date of the study
- Media sampled
- Measurement units
- Concentrations of each PCBs or PCDD/Fs congener

- Whether the sampled site corresponds to a background value or not
- Sampling site coordinates
- Sum of the concentration of all congeners reported for each specific case

The coordinates are normally captured using Google Earth® when there are not specified in the article, from geographic references in the article (such as maps or city names). In any case, a qualitative evaluation on the reliability of the reported coordinates is performed using Google Earth ®.

3. Overall spatio-temporal and congener distribution of the samples

3.1 PCBs

The following chart shows the number of sample points obtained for PCB analysis, per media and year.

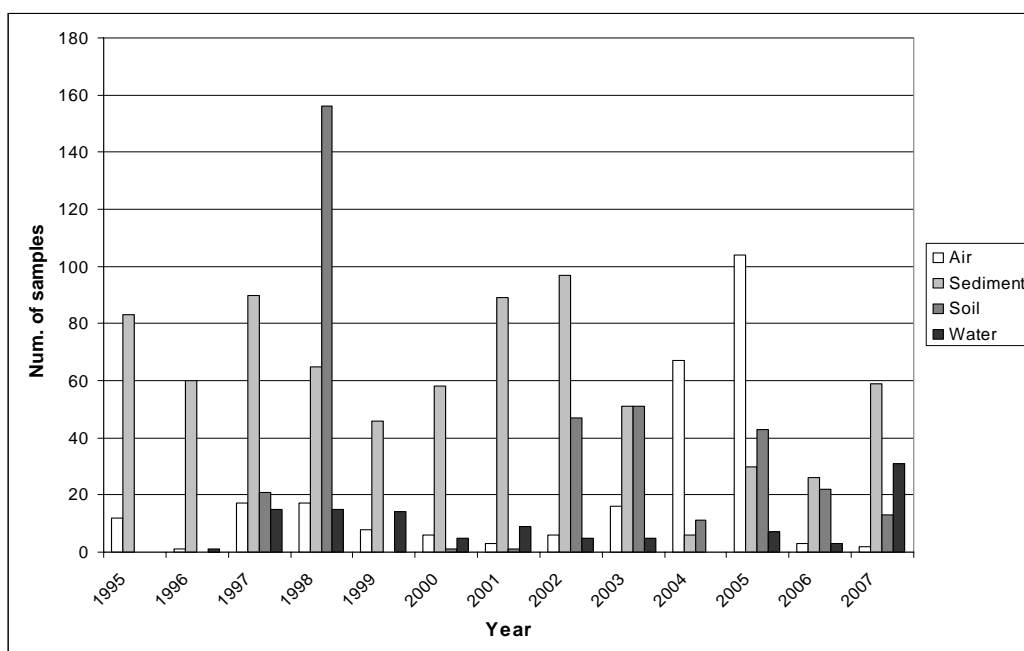


Figure 1: Number of sample points per media and year

The following diagram shows the mobile sum of the number of samples over three years, i.e. each year plus its previous and following years.

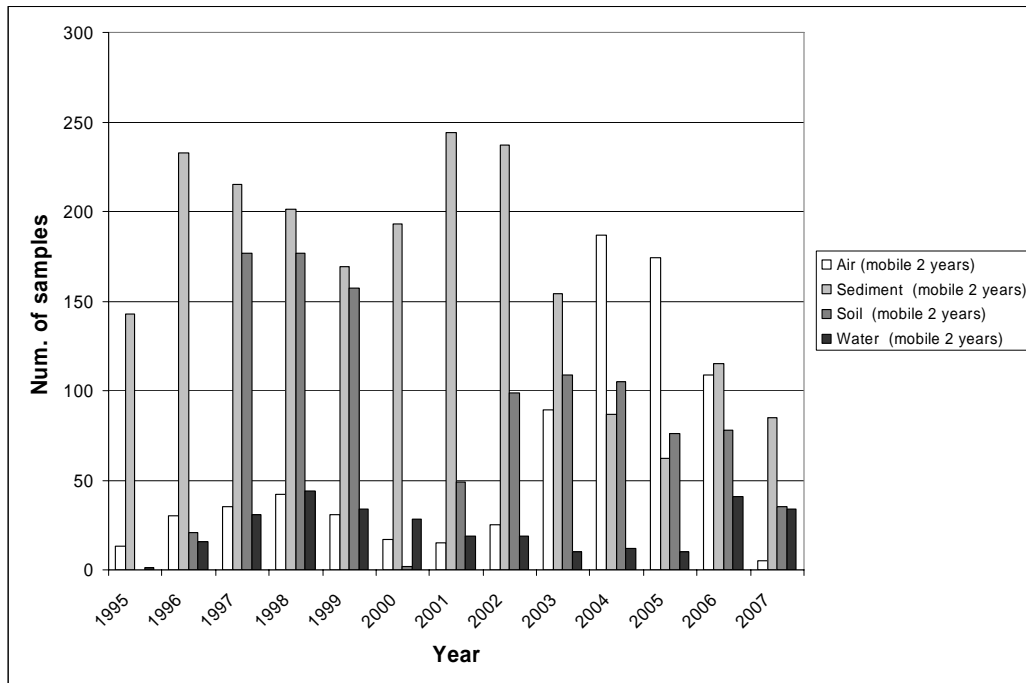


Figure 2: Mobile sum of the number of samples, per media and year

From the number of samples per year and the mobile sum charts, the following can be observed:

- the water samples are much less frequent than the number of samples that can be found for other media
- the year to year variability in number of samples is very high
- pooling together ± 1 year improves the availability of data
- the trend of number of samples in sediments decreases with the years while in air the trend observed increases, and in soil and water remains more or less equal.

The following four charts show the box and whiskers plot for each media, containing the mean concentration (\pm the standard deviation) of observed total PCBs, and their the median. Statistics are computed on the whole dataset for Europe.

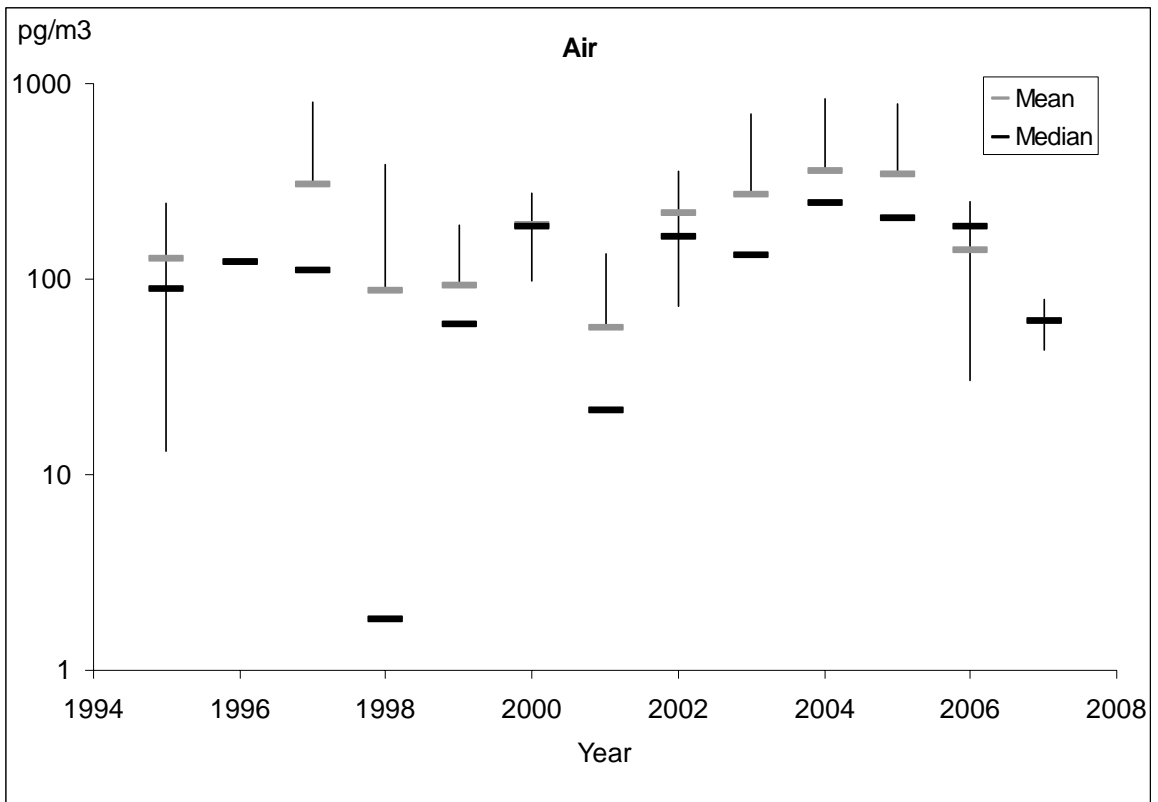


Figure 3: box and whiskers plot showing the air PCBs data distribution per year

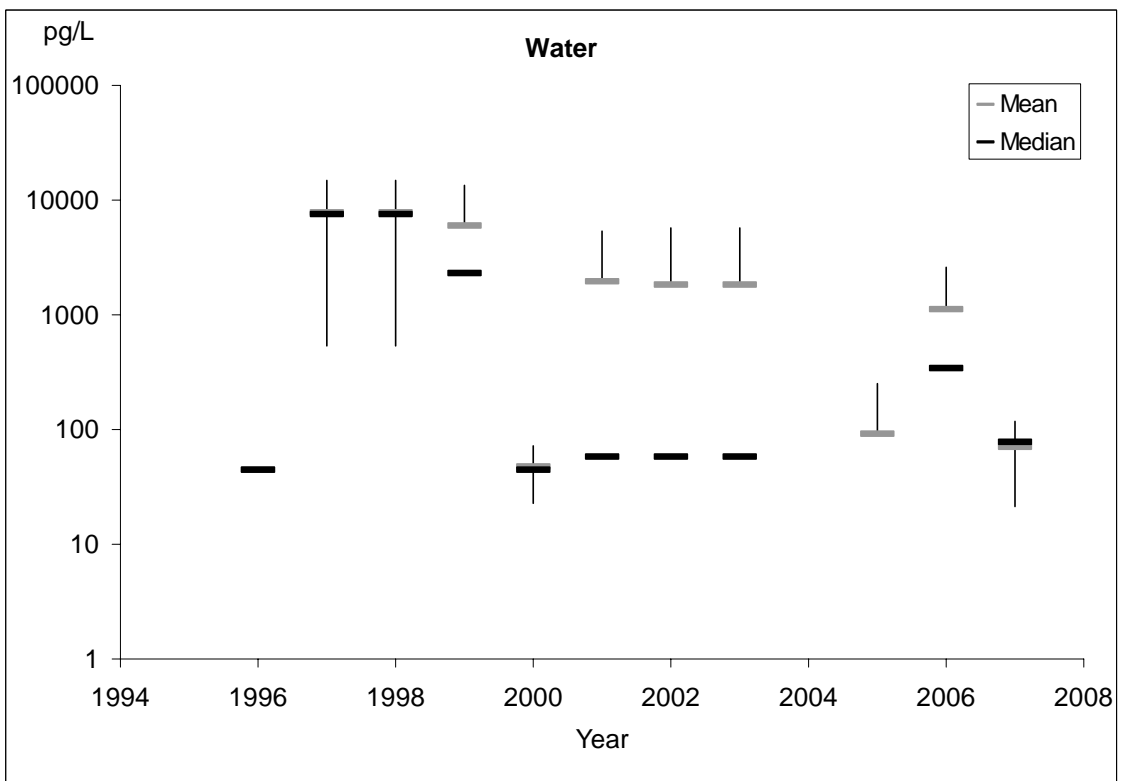


Figure 4: box and whiskers plot showing the water PCBs data distribution per year

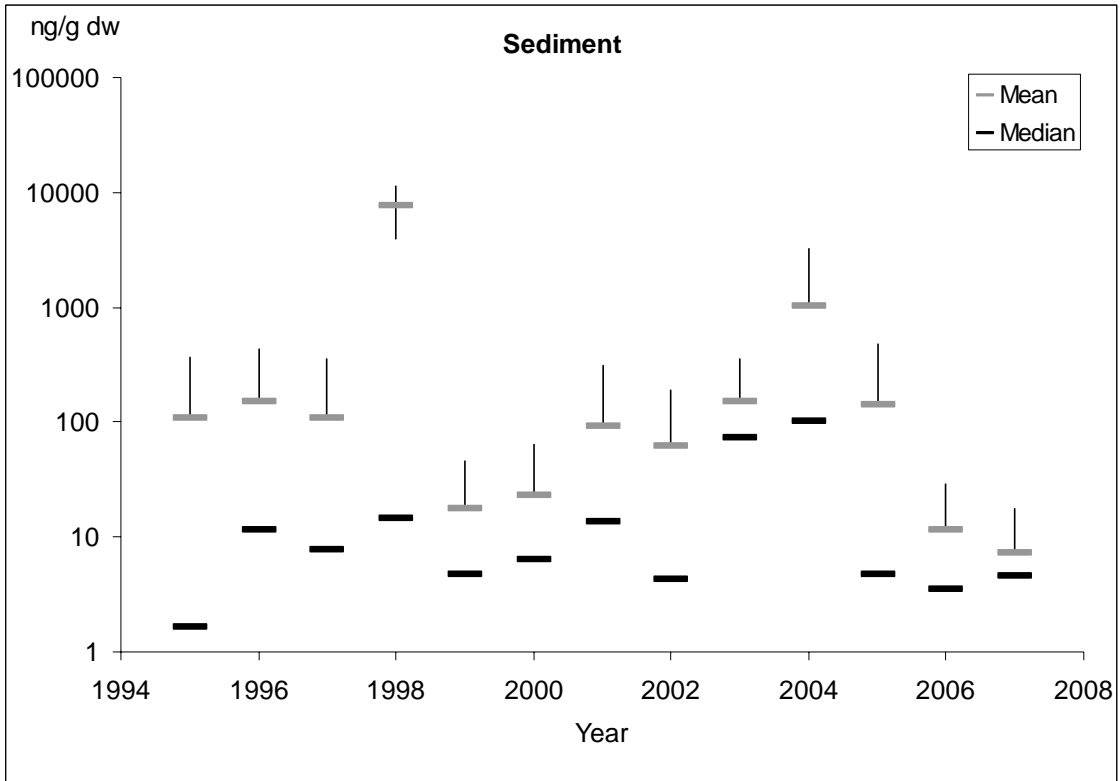


Figure 5: box and whiskers plot showing the sediment PCBs data distribution per year

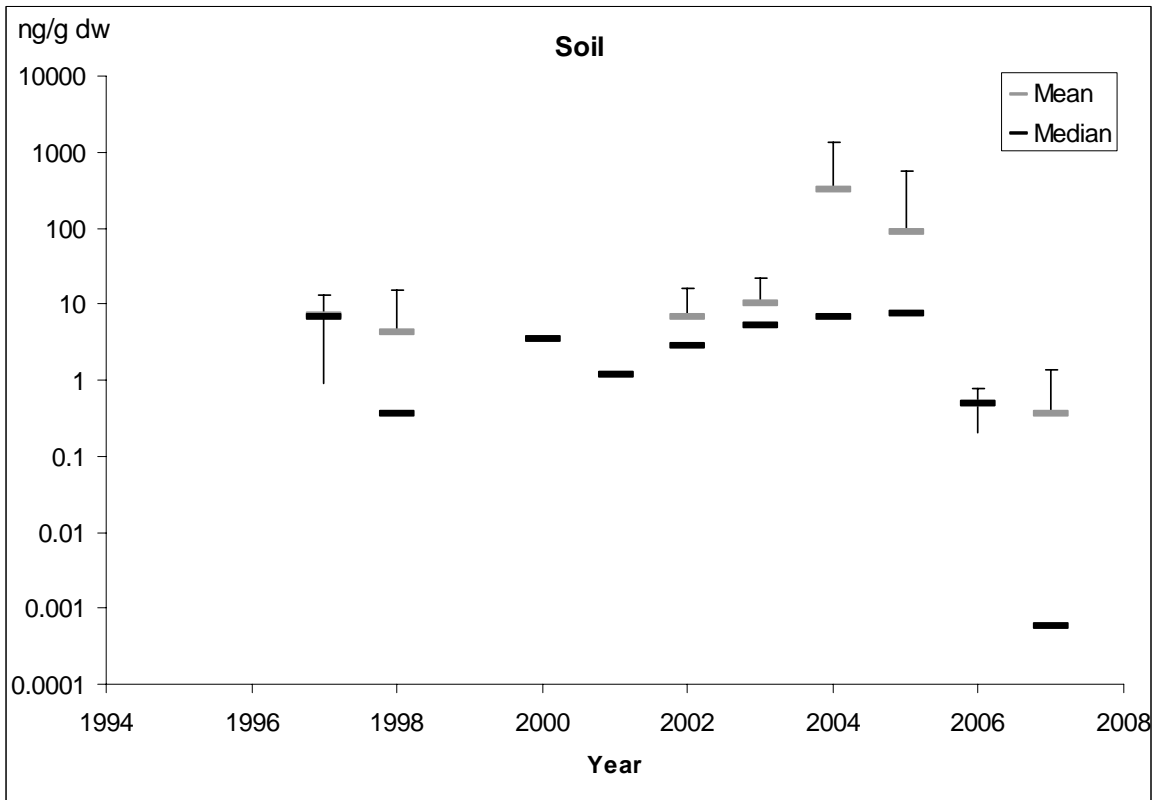


Figure 6: Diagram box showing the soil PCBs data distribution per year

The plots highlight no significant trend regarding PCBs concentration in time, in any of the media considered.

The following charts show the histograms of concentration of PCBs for each media.

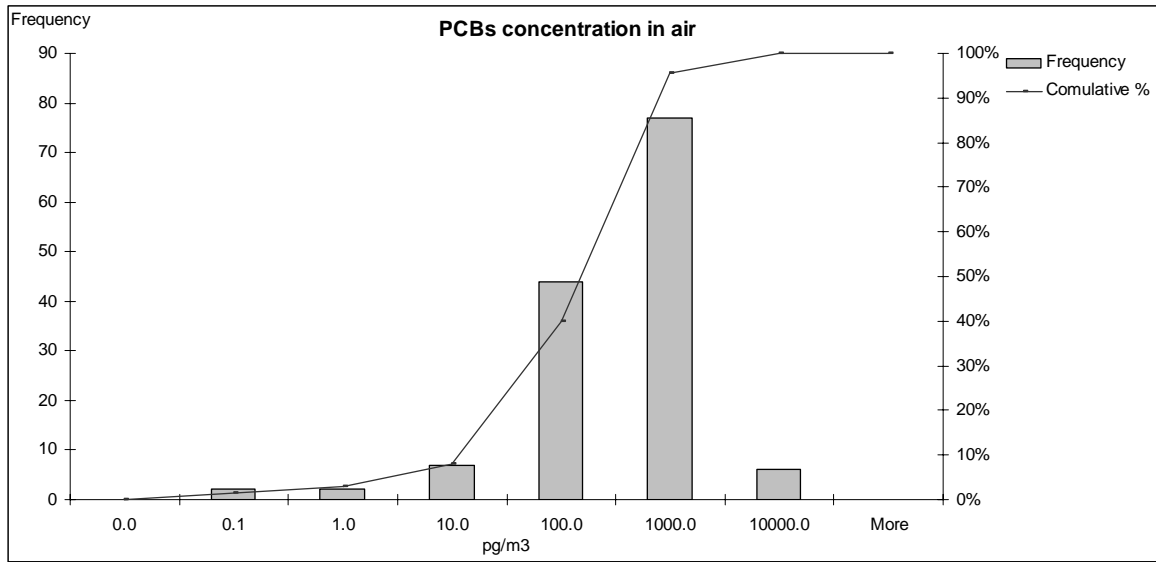


Figure 7: PCBs concentration in air

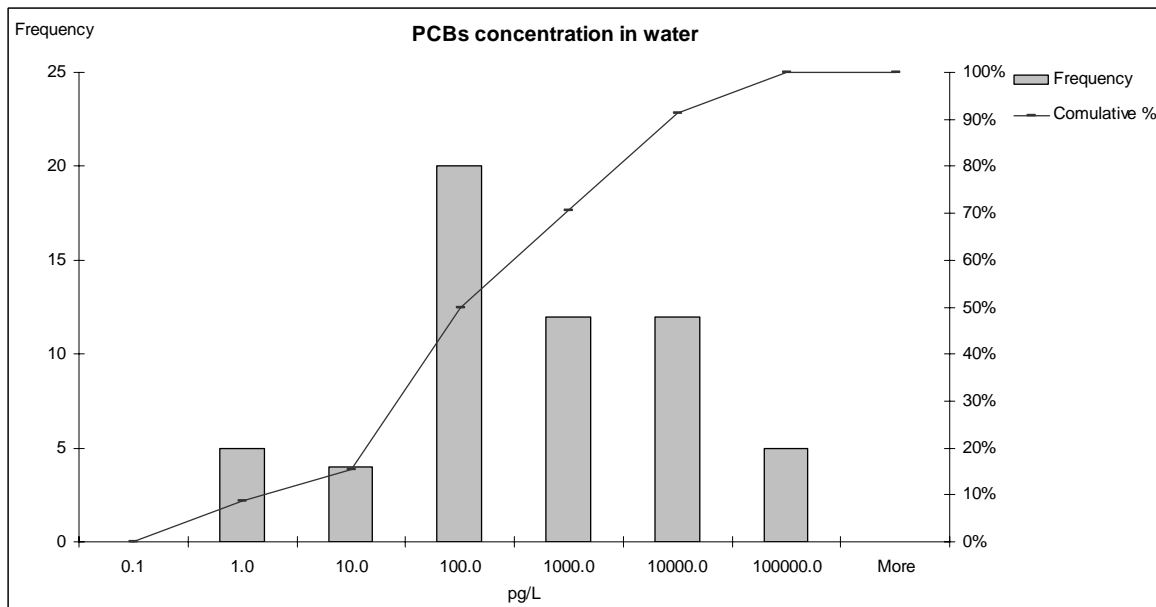


Figure 8: PCBs concentration in water

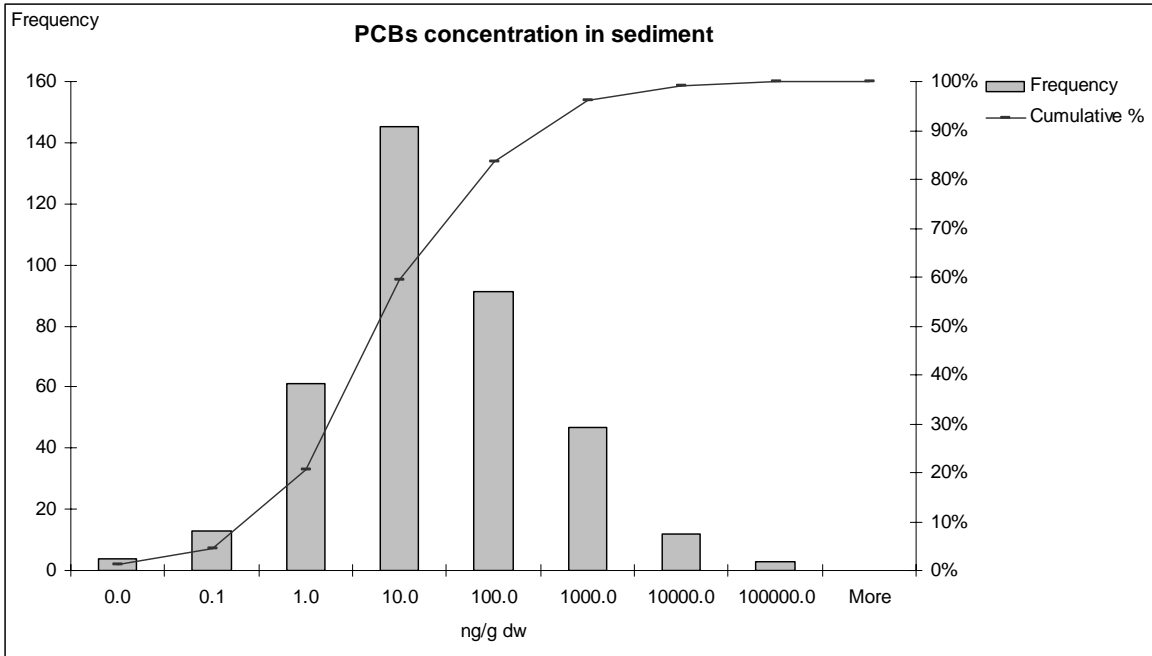


Figure 9: PCBs concentration in sediment

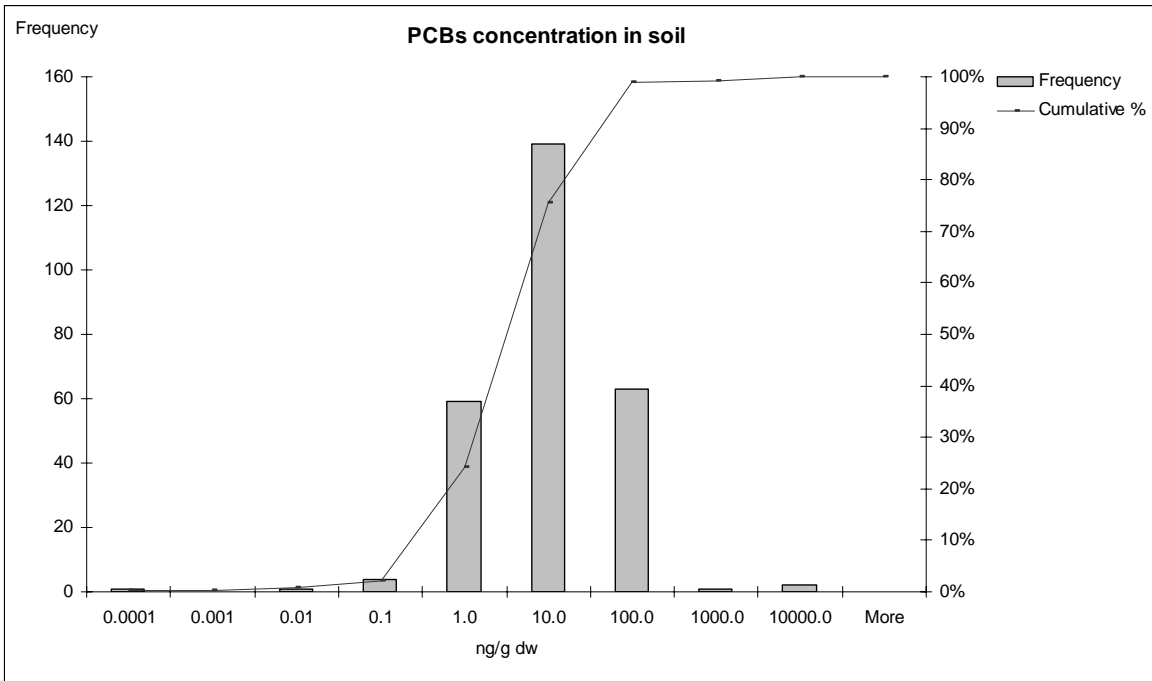


Figure 10: PCBs concentration in soil

The previous figures show outlier-like values for each media. In the case of air, the highest values belong to sample points located in urban areas. The same happens with soil: the highest values correspond to values at Zadar, Croatia (Picer, 2006). In the case of water, the highest values correspond to the study done by Telli-Karakov, in 2002, in the Izmir Bay, in Turkey. In sediments the highest values correspond to a sample point at the coast of Belgium, where there is a strong influence from a river whose catchment has a large urban and industrial burden.

The following maps show the spatial distribution of the sampling points by media.

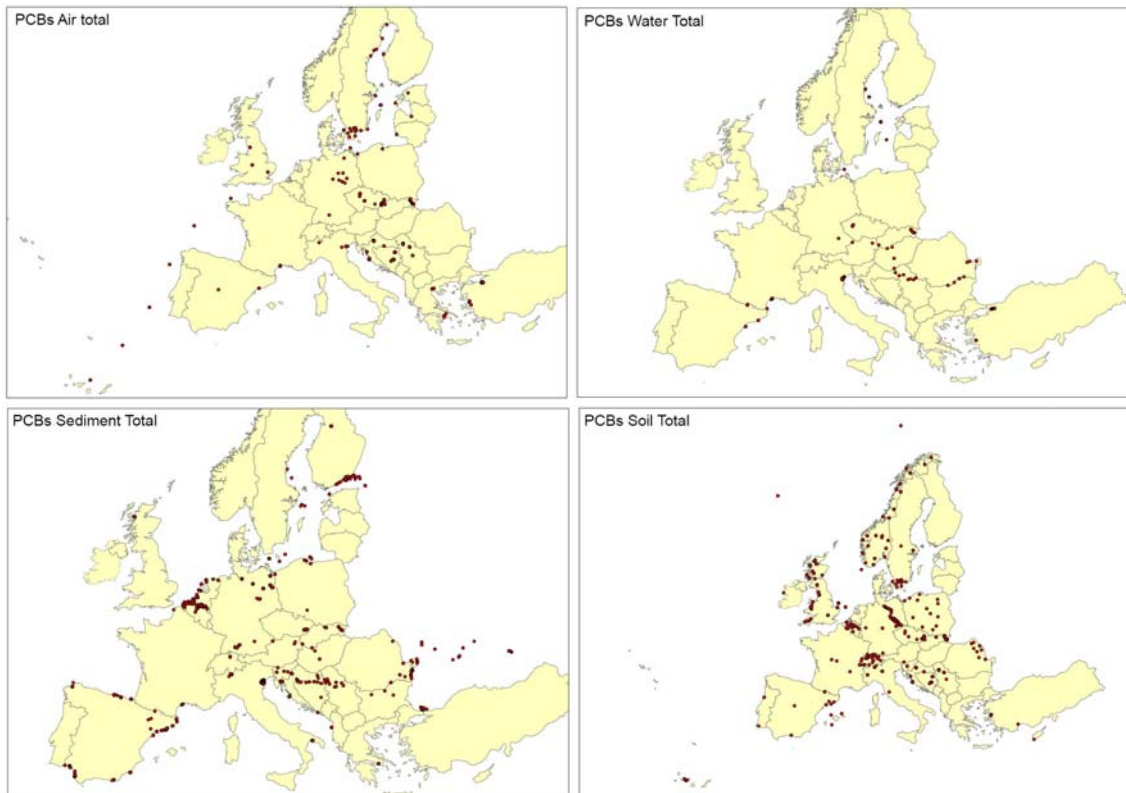


Figure 11: Maps showing the distribution of the sampling points per media

The maps show that the samples are reasonably evenly distributed in Europe in the case of soils. For air, water, and sediments, the spatial distribution seems to be more clustered into a few focus areas. Original spatial sampling campaigns (such as the above mentioned JDS2 for Danube) clearly determine focus areas in the above maps. It's also interesting to note how, for a specific time window, the studies appear to be more clustered in specific regions rather than being homogeneously distributed, as shown in annex 1 where the geographic distribution of the sample points, grouped by moving time windows of three years, is presented for further detail.

An important issue when comparing ambient levels of PCBs is the high variability on the number and type PCB congeners reported depending on the study. In addition, not all PCB congener gives the same information. For example, dioxin-like PCB (DL-PCBs) may give more toxic effects related information whereas 'indicator' PCB (except CB-118) gives an idea of the overall spatial distribution of PCBs in the environment since these 7 congeners are ubiquitous in many environmental compartments. In general, even if there are 209 different PCB congeners, most of the studies only focus on a small subset of them. Moreover, some studies don't specify the individual congeners studied. The following plot shows the percentage of studies grouped by the number of PCB congeners considered. From the graph, it is apparent that 18% of the studies does not provide details on the congeners investigated; 5% concerns the 12 "dioxin-like" PCB congeners, and 23% the 7 indicator PCBs. However, a vast majority (54%) of the studies does not refer to a standard set of congeners, which makes comparisons more difficult as mentioned before (Figure 13). Figure 14 shows the number of samples containing each PCB congener, PCB118 being the most frequent one. Among the 209 PCBs, seven of them are anyway the most frequently studied.

For the PCB 118, which is the most frequent in the PCBs data collected, Figure 12 shows the mobile sum of the number of samples for each year, ± 1 year.

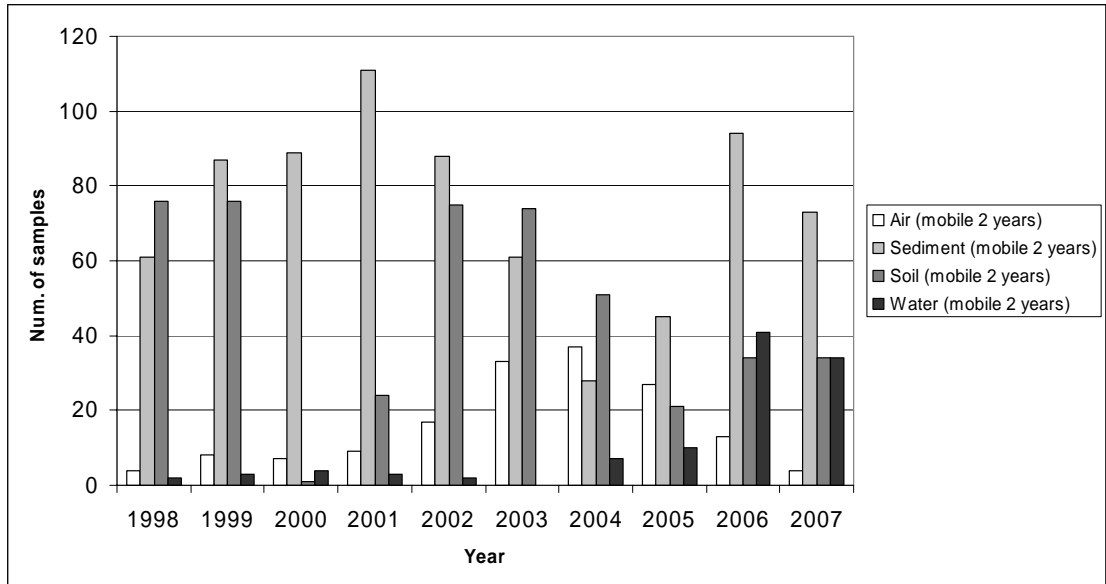


Figure 12: Mobile sum of the number of samples analysed for PCB118, per media and year

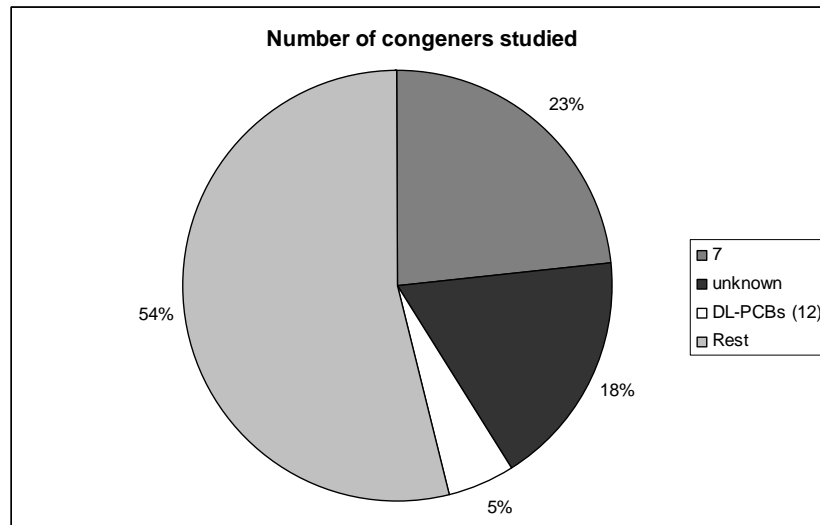


Figure 13: Groups of PCB congeners studied in each study. (DL-PCBs means “dioxin-like PCBs”)

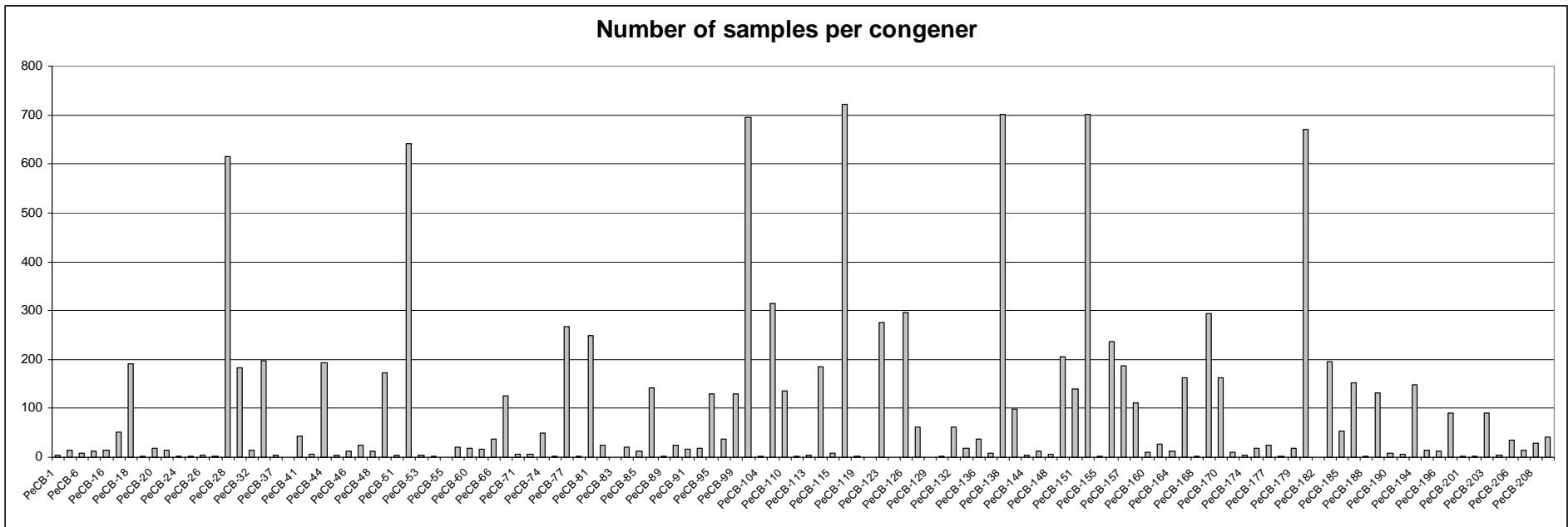


Figure 14: number of samples per PCB congener

In air and water, PCBs are analyzed sometimes just on gas (dissolved) phase, or particulate phase, and some other times on both phases. Some studies don't specify any difference among phases (so the phase is unknown), or the sum of both. The following table shows the percentages for phase distribution for air and water.

Air phases	Total
Air (gas+particle) Phase	55,33%
Air (gas phase)	41,00%
Air (part phase)	3,67%

Table 2: percentage of phase distribution in air studies

Water phases	Total
Water (Dissolved phase)	52,78%
Water (Dissolved + particle) phase	38,89%
Water (phase unknown)	8,33%

Table 3: percentage of phase distribution in water studies

3.2 PCDD/Fs

The following chart shows the number of sample points obtained, per media and year:

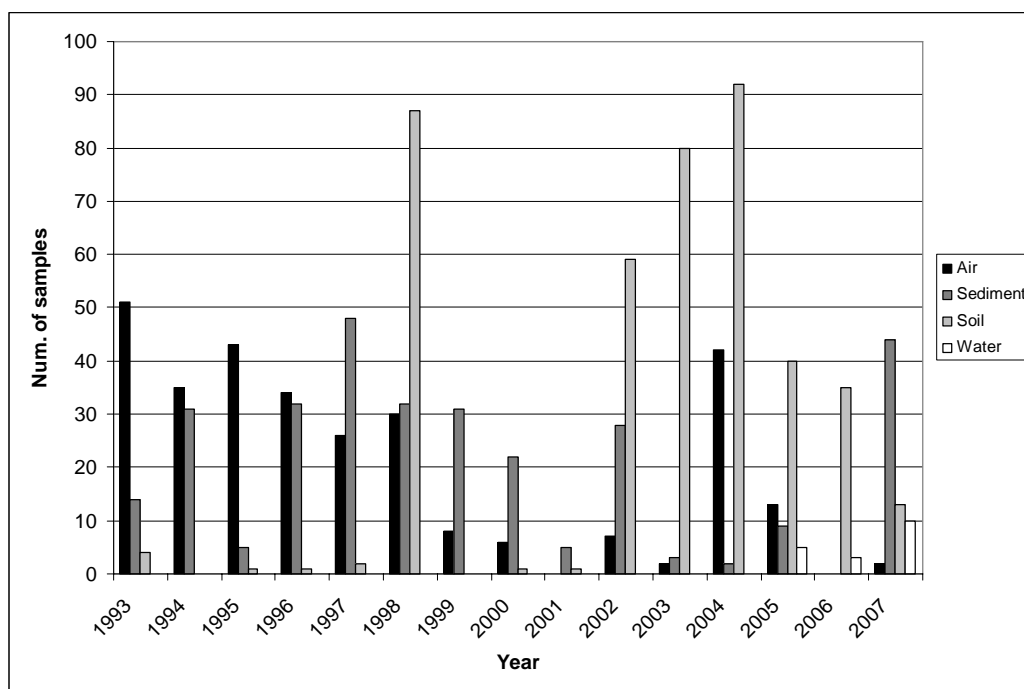


Figure 15: PCDD/Fs number of samples per year and media

The following diagram shows the mobile sum of the number of samples over three years, i.e. each year plus its previous and following years.

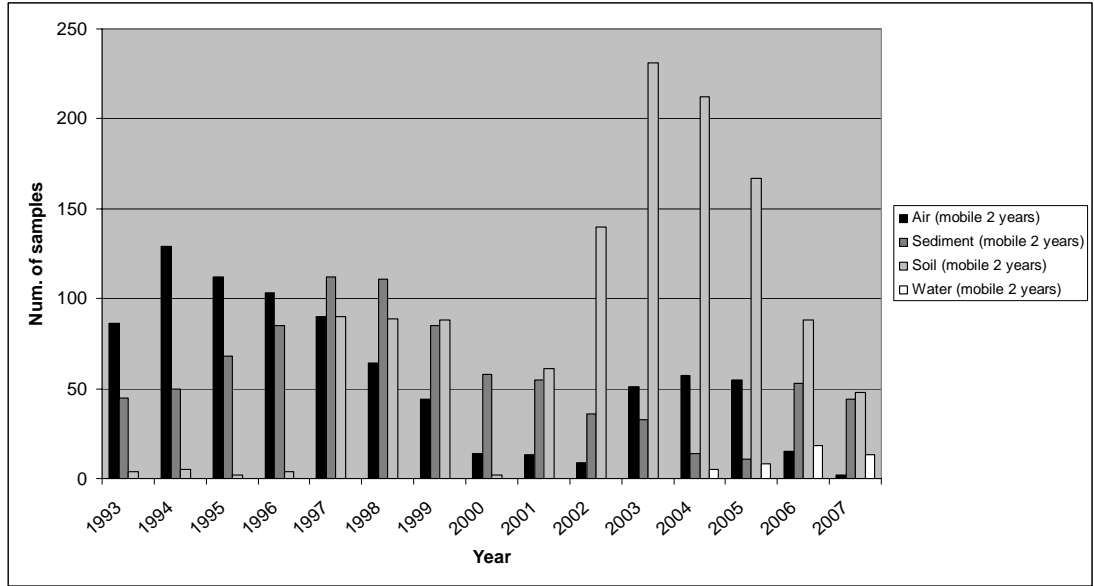


Figure 16: Mobile sum of the number of PCDD/Fs samples, per media and year

From the above charts, it can be observed that the water samples are much less frequent than the number of samples that can be found for other media; before 2004 there were no water samples and the year to year variability in the number of samples is very high; pooling together mobile time windows of three years improves the availability of data.

The following four charts show the diagram boxes for each media (containing the average concentration of total PCDD/Fs observed, the median, and +/- the standard deviation for each year).

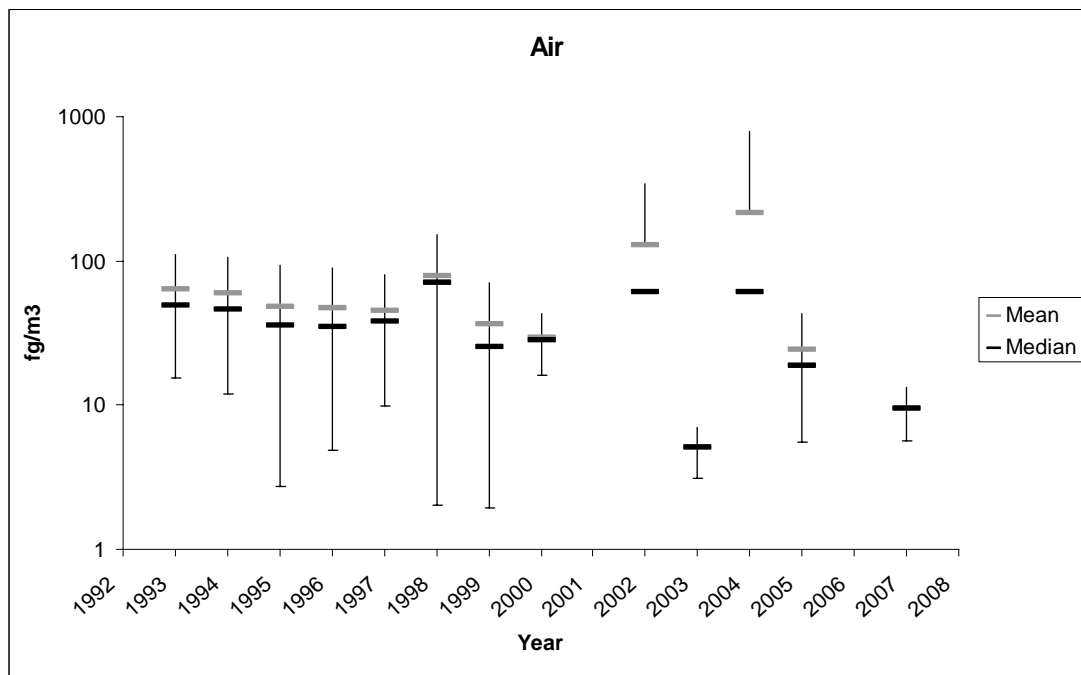


Figure 17: box and whiskers plot showing the air PCDD/Fs data distribution per year

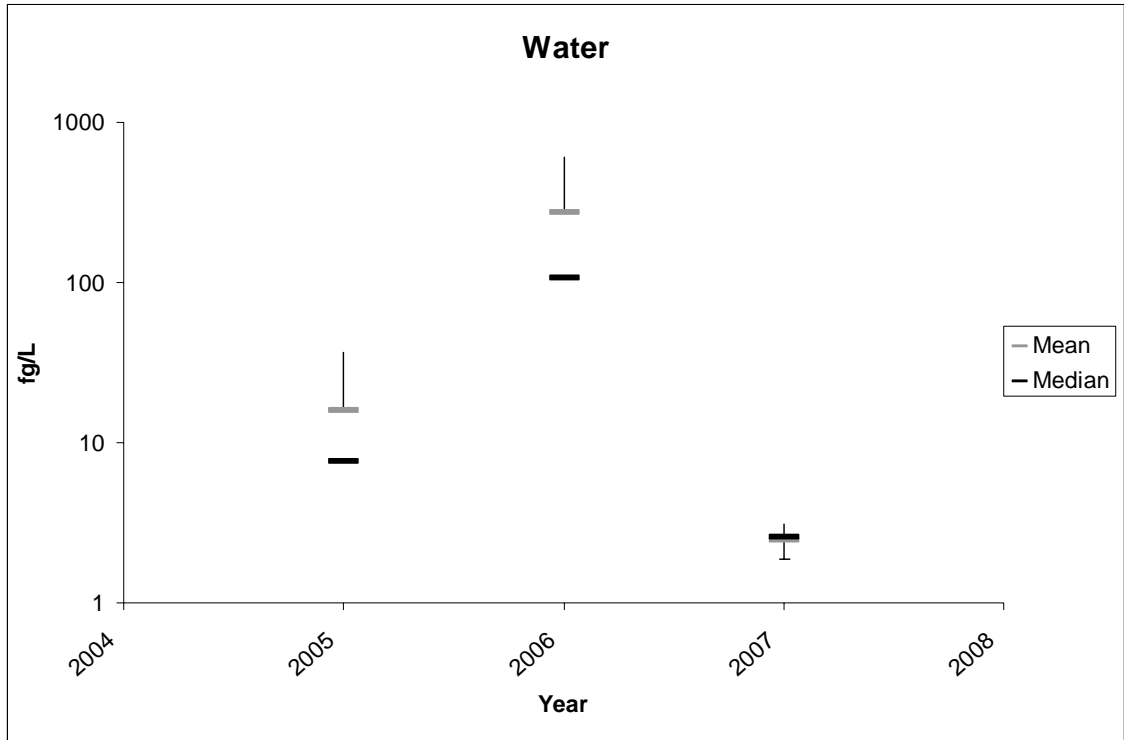


Figure 18: box and whiskers plot showing the water PCDD/Fs data distribution per year

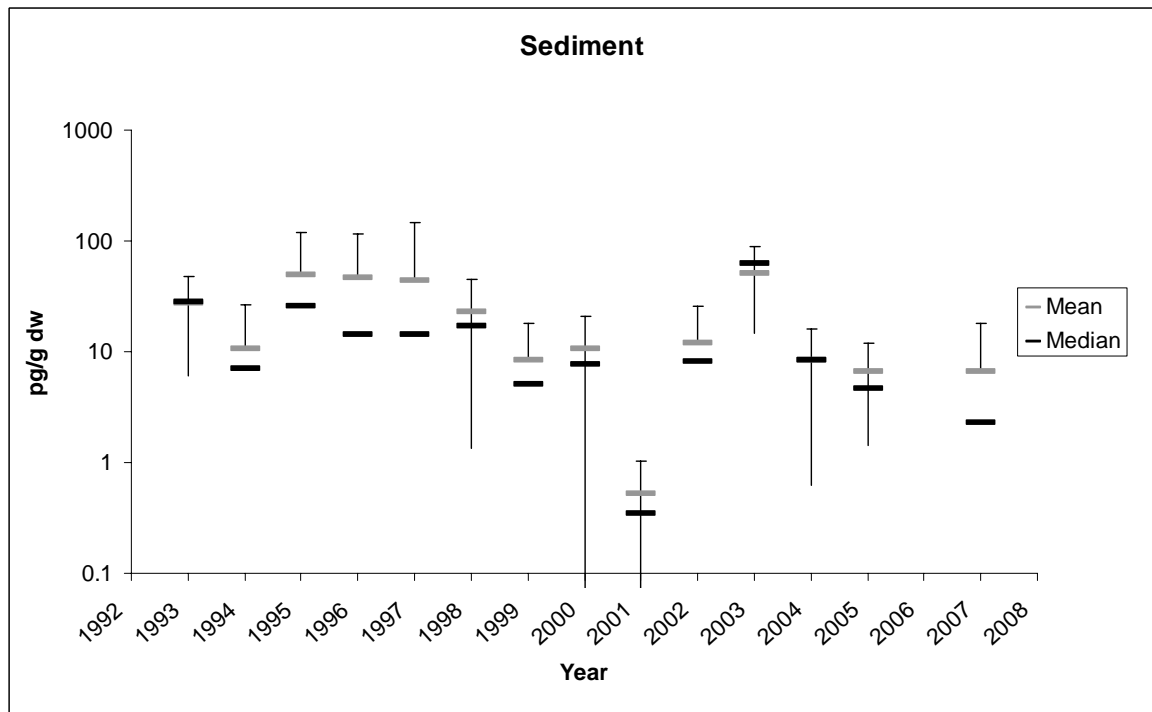


Figure 19: box and whiskers plot showing the sediment PCDD/Fs data distribution per year

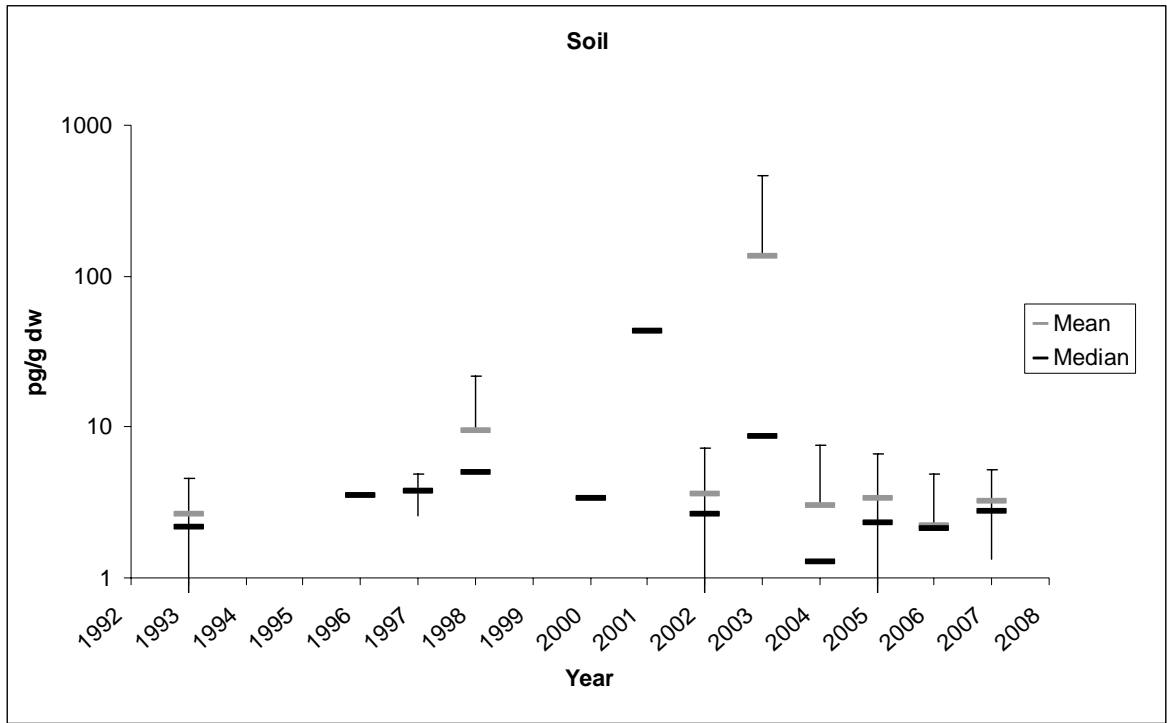


Figure 20: box and whiskers plot showing the soil PCDD/Fs data distribution per year

The box and whiskers plots don't show any clear trend for any of the four media studied.

The maps of Figure 21 show the spatial distribution of the sampling points by media. In the case of PCDD/Fs, the best geographic coverage is for soils and sediments. However, the spatial distribution of samples is always clustered in focus areas.

It's also important to stress that the number of PCDD/F congeners analysed in each case do not vary very much, all the studies only focusing on a subset of the many congeners. The following table highlights that the majority of the studies focuses on 17 congeners. When congeners are not reported, most of the times this is due to concentrations being below the level of detection.

Number congeners	Total
17	75,89%
5	23,25%
2	0,85%

Table 4: Percentage of studies, according to the number of PCDD/Fs congeners studied

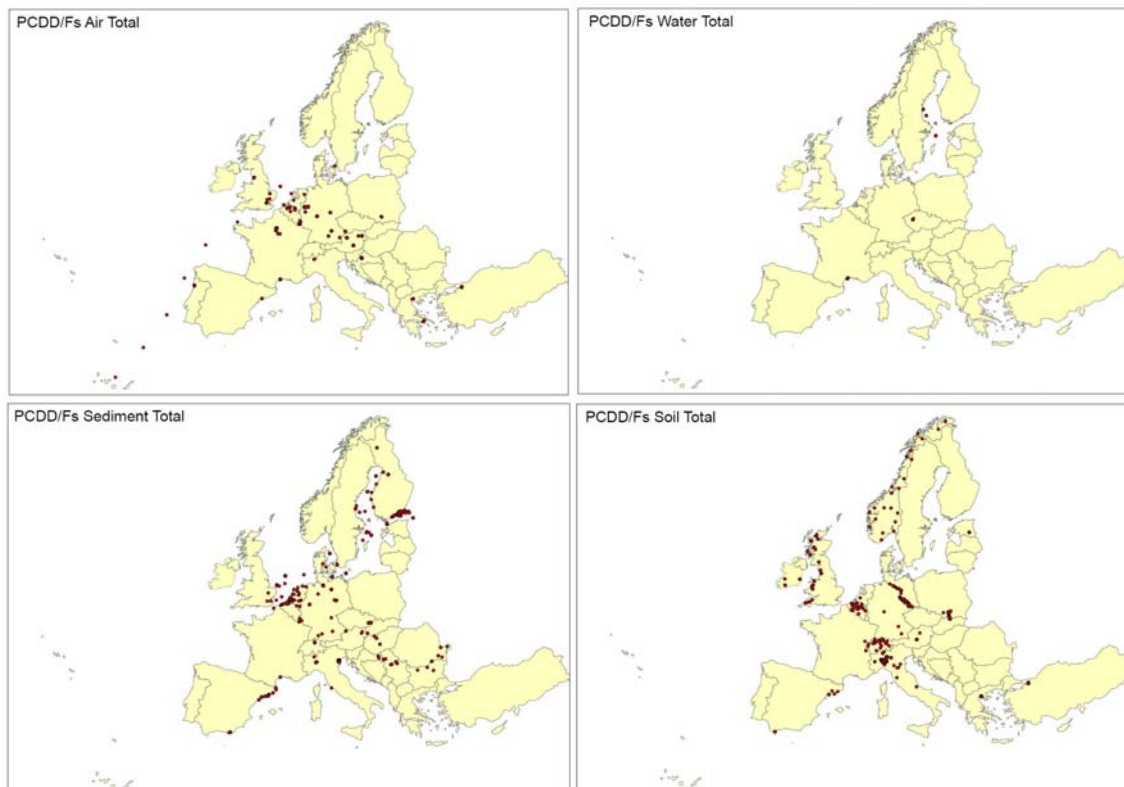


Figure 21: Maps showing the geographic distribution of the samples, per media. “Total” means that maps display the whole dataset.

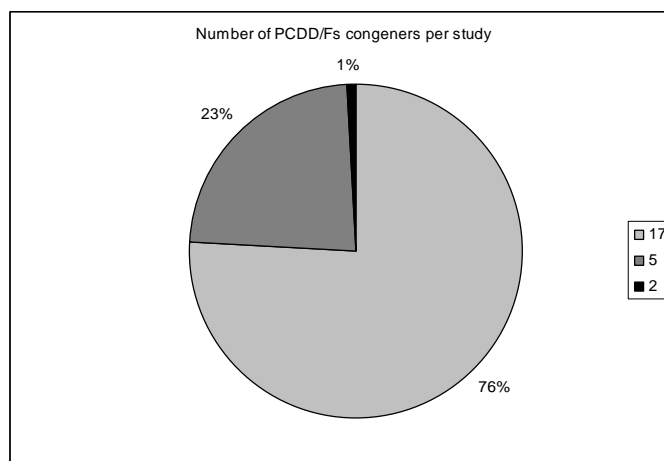


Figure 22: of studies, according to the number of PCDD/Fs congeners studied

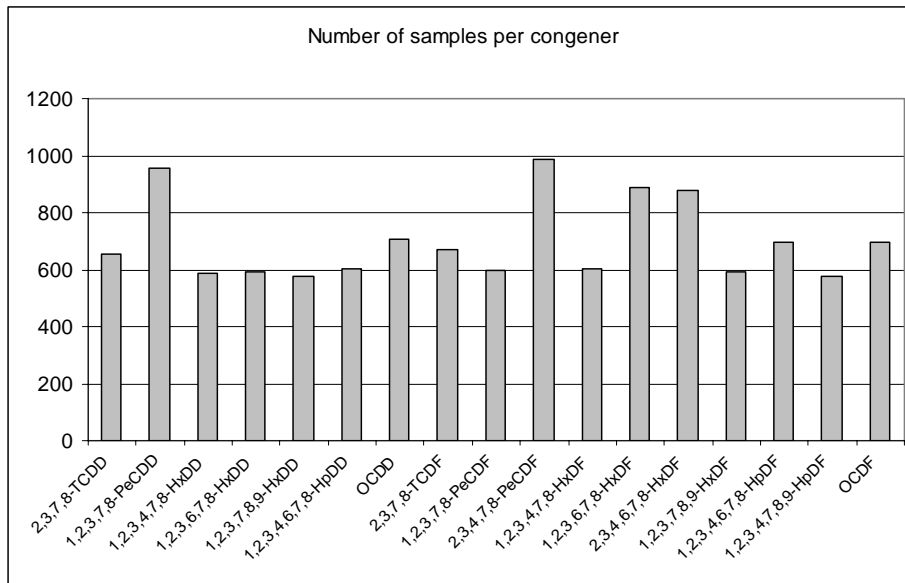


Figure 23: number of samples per PCDD/Fs congener

The majority of the studies represented the total PCDD/F concentration in terms of WHO-TEQ (65% of the studies), while the rest refer to I-TEQ (35% of the studies). What about the ones which report absolute concentrations???)

In air and water, PCDD/Fs are analyzed sometimes on gas (dissolved phase), sometimes on particulate phase. Some studies specify what the concentration in each phase is if they have studied both, some specify one phase or the other, and finally others don't specify the phases of measurement, or provide the sum of both. The following tables show the percentages of occurrence of the phase of measurement for air and water.

Air phases	Total
Air (Gas+particle) Phase	97,58%
Air (phase unknown)	1,21%
Air (part phase)	0,91%
Air (gas phase)	0,30%

Table 5: air phases where PCDD/Fs are described, distribution breakdown

Water phases	Total
Water (Dissolved phase)	72,22%
Water (Particle phase)	27,78%

Table 6: water phases where PCDD/Fs are described, distribution breakdown

4. Spatial correlation

The result of the data collection is a georeferenced database of PCBs or PCDD/Fs concentrations for different years. The first question we might be interested in answering is whether the PCBs have an overall spatial trend in Europe. In order to analyze that, geostatistical techniques should be used.

Data for each environmental medium (air, soil, water, sediments) were selected for each moving time window of one year +/- 1 year and the spatial distribution of concentrations is examined in terms of:

- the variogram cloud
- the variogram surface

- the omnidirectional variogram
- if relevant (if spatial trends are observed), the directional variogram(s) in the directions where a spatial trend is observed in the variogram surface.

For these operations, we used the popular VARIOWIN® software: <http://www-sst.unil.ch/research/variowin/>.

The variogram cloud indicate the presence of more than one statistically homogeneous population of data within the data set; the variogram surface allows detecting directions of anisotropy; the variograms capture spatial correlation and indicate whether the data are, to a certain extent, continuous in space. Further details and the basic concepts of geostatistics can be found e.g. in Clarke, 1979.

In general, all variograms that have been calculated do not indicate strong spatial correlation of the data. This was largely expected given the heterogeneity of the data and their being referred to different study locations in Europe. However, in some cases a less weak spatial correlation emerges as shown in more details in the examples below. In general, a short-range spatial correlation (i.e. a positive slope of the variogram near the origin, or “near trend”) indicates spatial continuity of the samples, while a long-range spatial correlation (i.e. a positive slope of the variogram distant from the origin, or “distant trend”) indicates continental scale gradients of concentrations. The results discussed in the examples below, and the full set of analysed variographies provided in Annex 2, are to be seen as purely explorative analyses and not to be taken as rigorous geostatistical analyses, which instead would require separating the samples in homogeneous clusters; their purpose is just to give a taste of the type of spatial correlation that may be found in such heterogeneous data set.

In the periods 1997-1999, and 2003-2005, the concentrations of PCBs in soils show some spatial correlation, as the variance grows with distance (Figure 27 and Figure 28 respectively). The variogram clouds show different sills of variance, corresponding to the different clusters of data points in Europe.

For the period 2004-2006, no spatial correlation appears (Figure 29).

With reference to the years 2004-2006, the analysis was repeated using 3 geographical subsets, corresponding to three clusters of samples which are apparent on the map (Figure 30): Belgium, Czech Republic, and the Balkans. However, also in this case spatial correlation is absent (Figure 31, Figure 32, Figure 33).

The same analysis for soil has been made for PCB congener 118, but only for the periods where this PCB congener was more frequent. It apparently shows a spatial correlation as well, especially for the period 1998-2000 (Figure 34), and less so for the other years (Figure 35, Figure 36).

For sediments, result for the period 2001-2003 shows some spatial correlation (Figure 37).

The complete set of the geostatistical analyses prepared are included in Annex 2. A summary of the trends found is provided in Table 7.

The analyses on PCDD/Fs were not conclusive, probably because of the small amount of sample points available. There were more samples for soil, but no spatial correlation was found. The results of the analyses are included in annex 2. A summary of the results is provided in Table 8.

As a benchmark to understand what one may expect in terms of spatial correlation of POPs at the European scale, variograms may be computed for natively spatially distributed datasets. We show here the examples of the passive sampler data concerning PCBs in air described in Jaward et al., 2004, and the data for soils presented in Meijer et al., 2003; these data were used in Pistocchi, 2008, to evaluate a European model of chemical fate. In the following graphs, the variograms are displayed. The passive samplers show no significant spatial correlation (the variogram is almost a pure nugget effect)¹. On the contrary, soil data show some trend in spatial correlation, which is more pronounced when referring to concentrations normalized by organic matter content. However, the trend is always rather weak with respect to the nugget.

¹ In geostatistical jargon, “nugget” is the variance at very short distance between sample points.

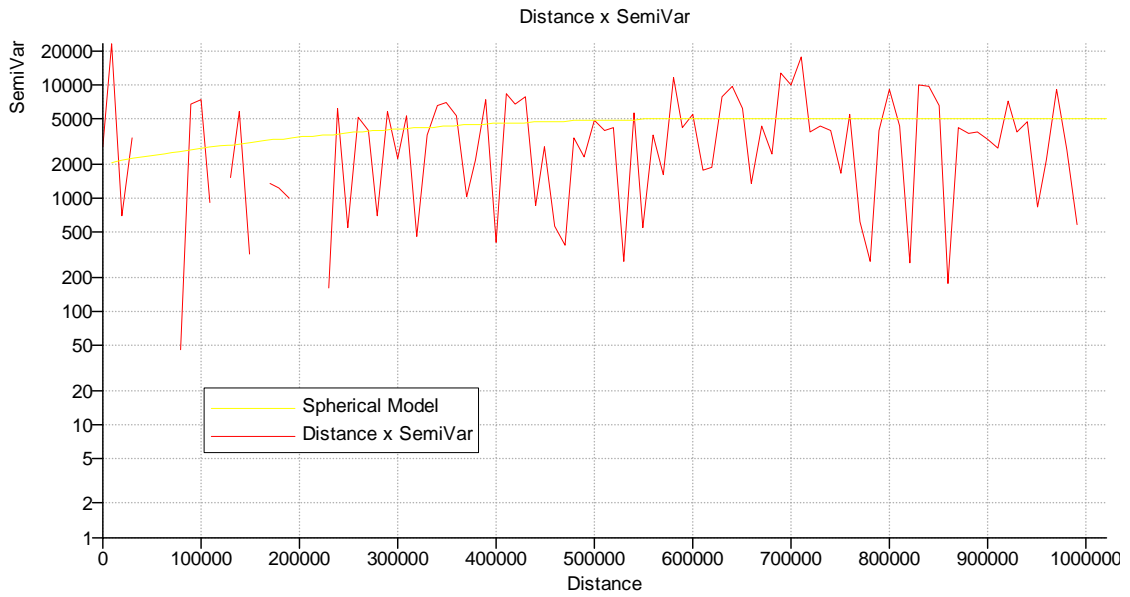


Figure 24 – variogram of the data on total PCB mass in air passive samplers from Jaward et al., 2004

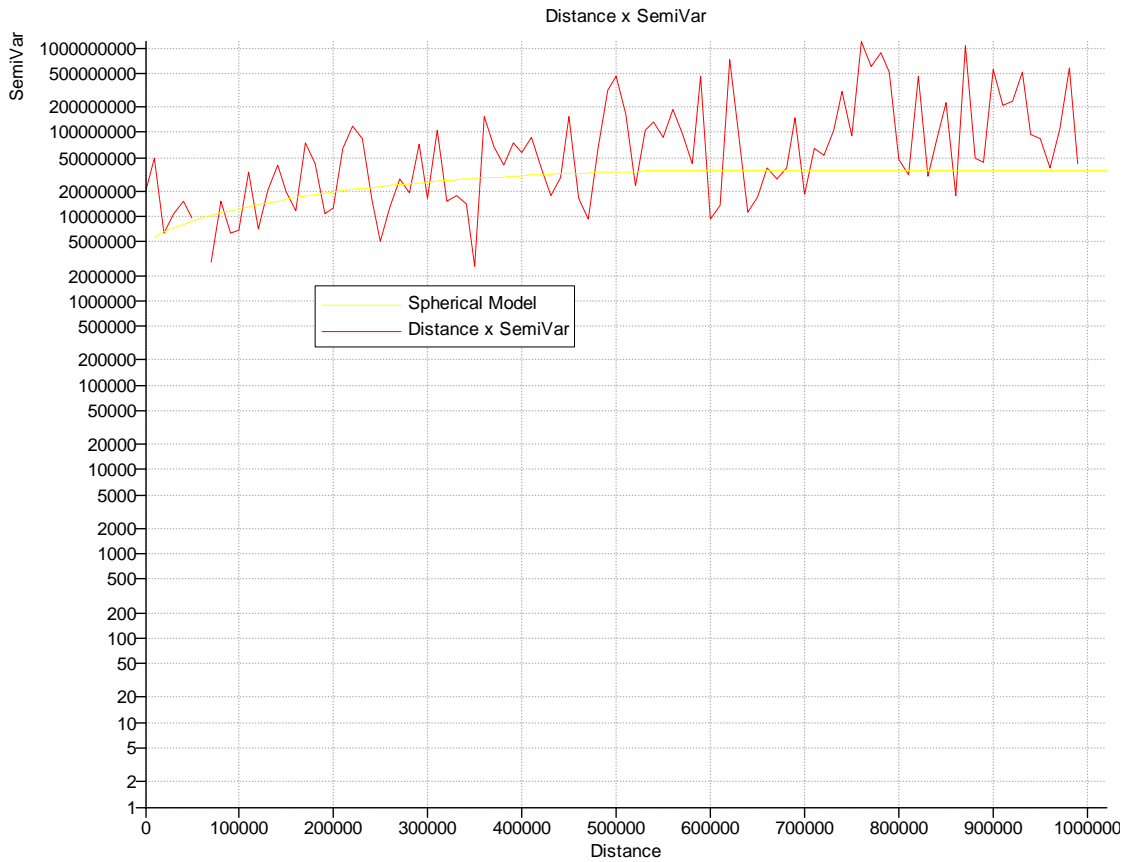


Figure 25 - variogram of the data on total PCB concentration in soil from Meijer et al., 2003

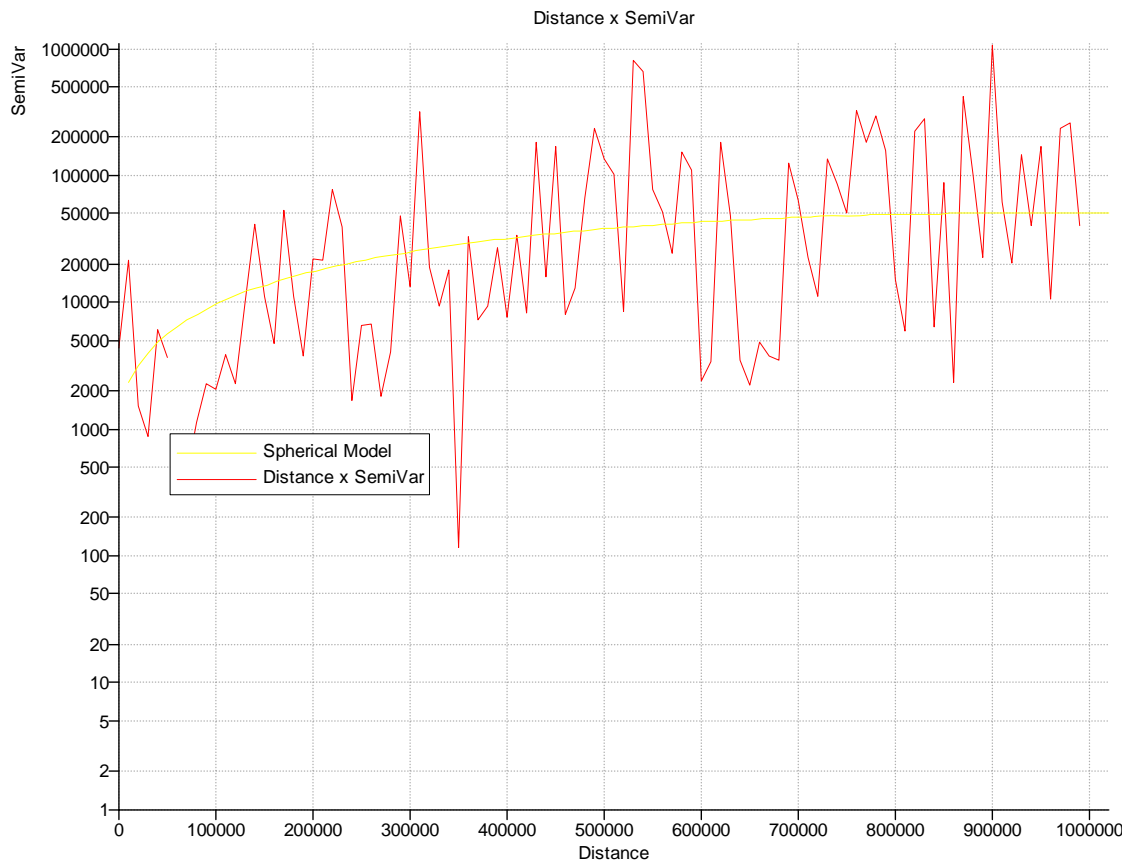


Figure 26 - variogram of the data on total PCB concentration in soil from Meijer et al., 2003; concentration normalized by organic matter content of soils.

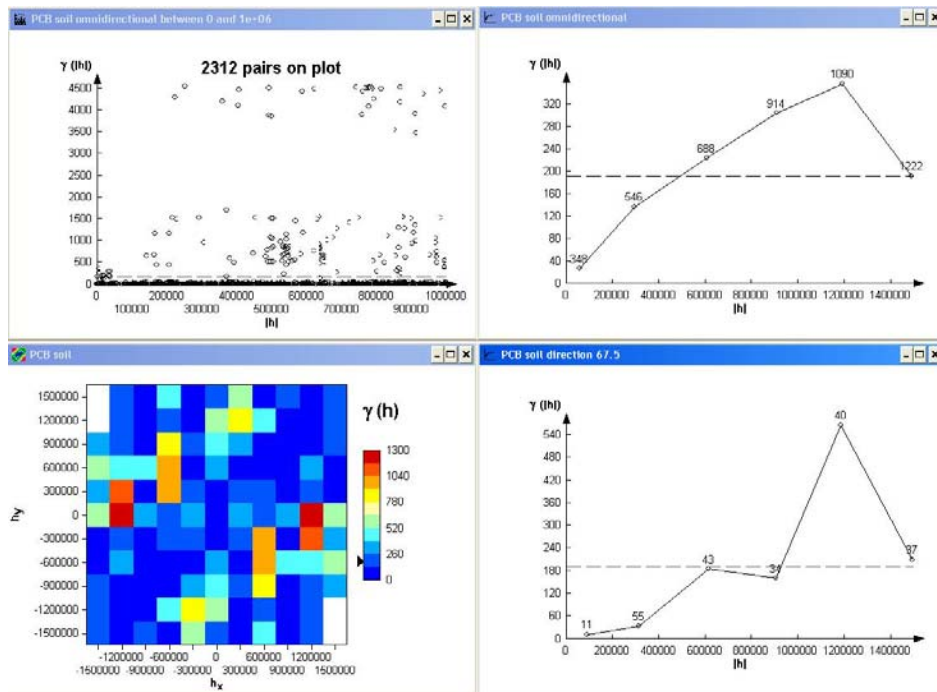


Figure 27: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 1997-1999

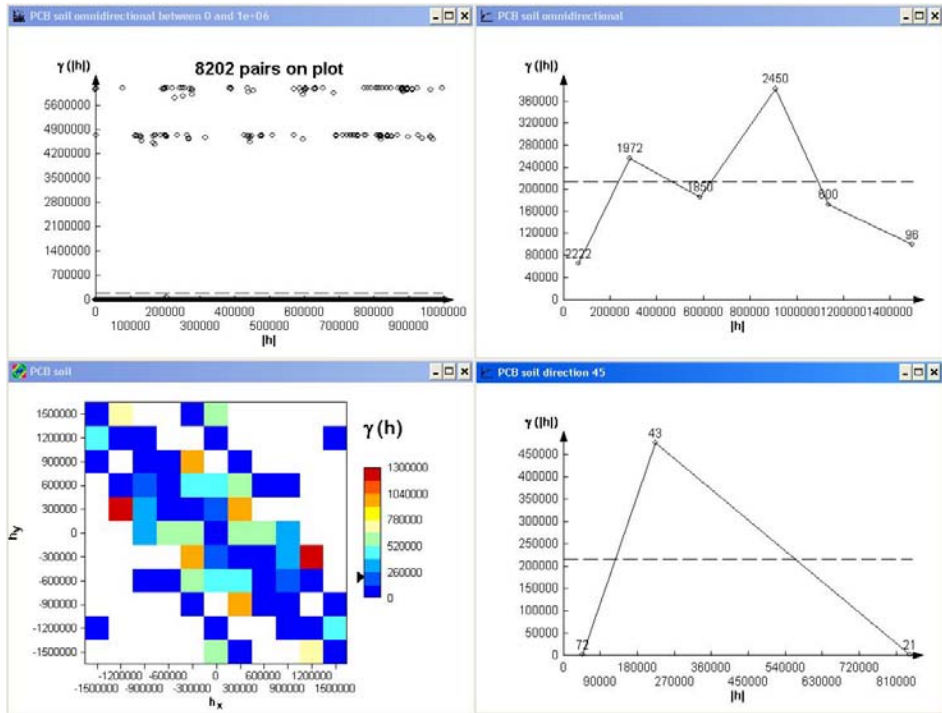


Figure 28: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2003-2005

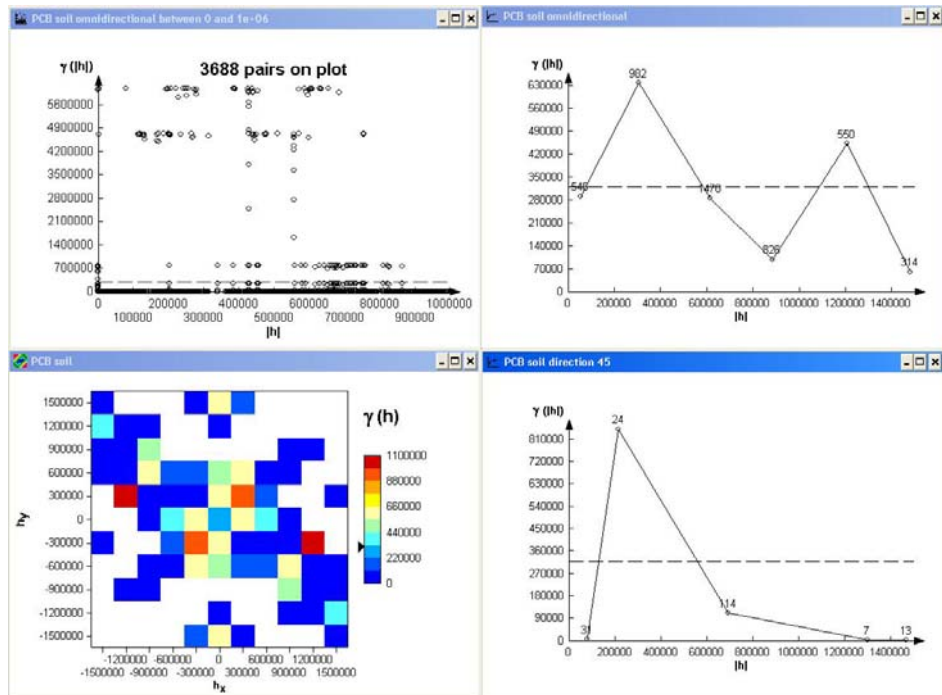


Figure 29: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2004-2006

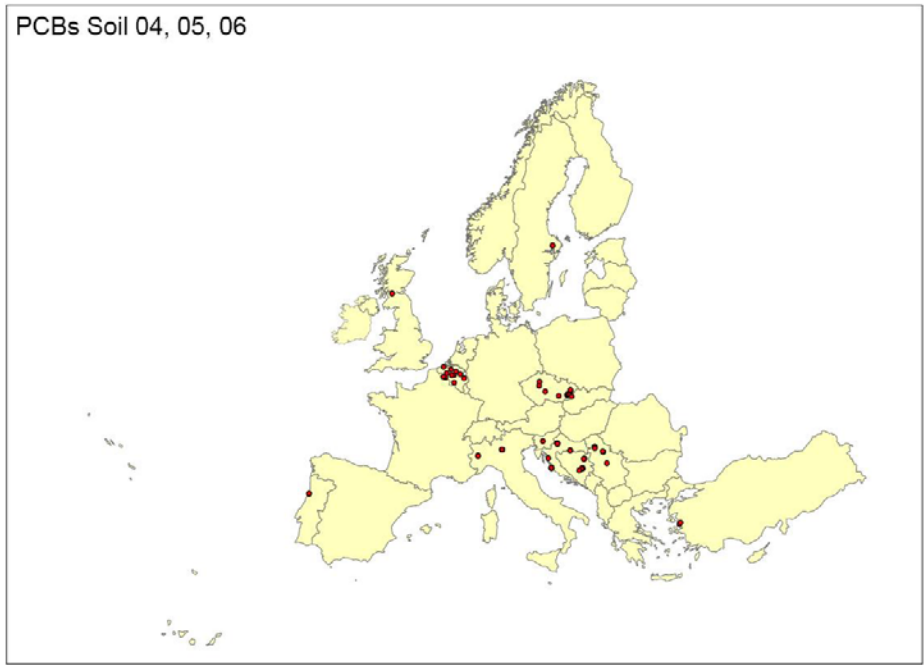


Figure 30: map showing how PCBs samples on soil in the period 2004-2006 are spatially clustered

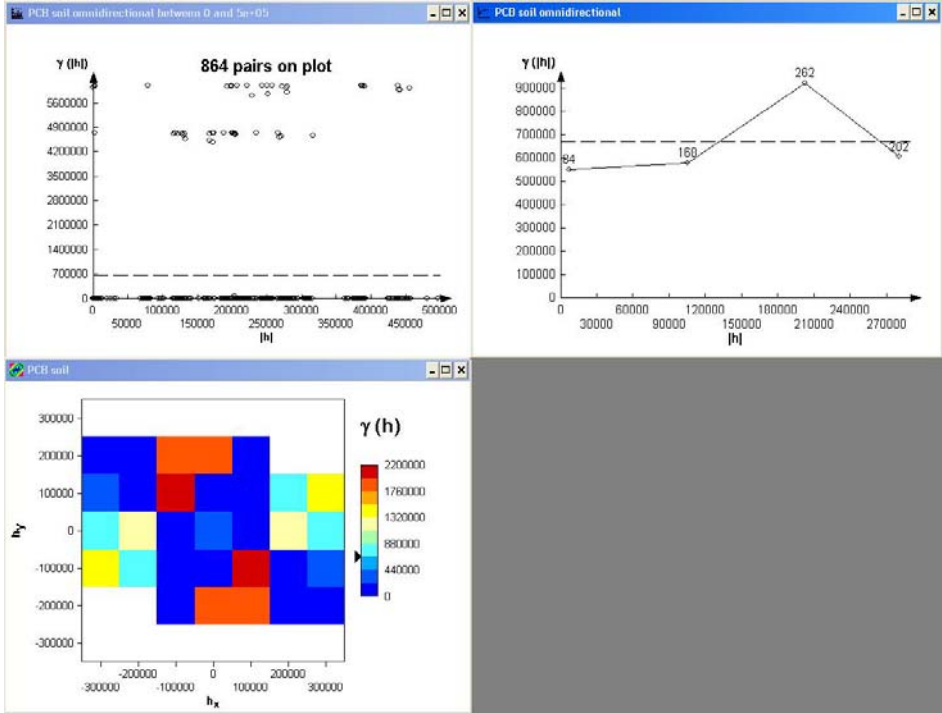


Figure 31: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2004-2006 in the Balkan Peninsula

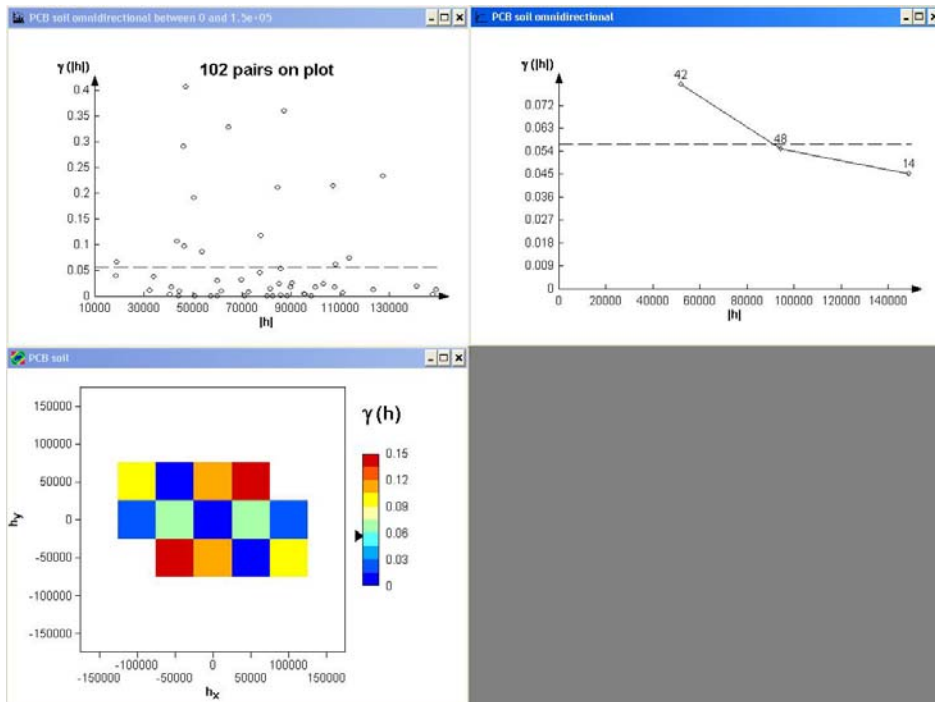


Figure 32: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2004-2006 in Belgium

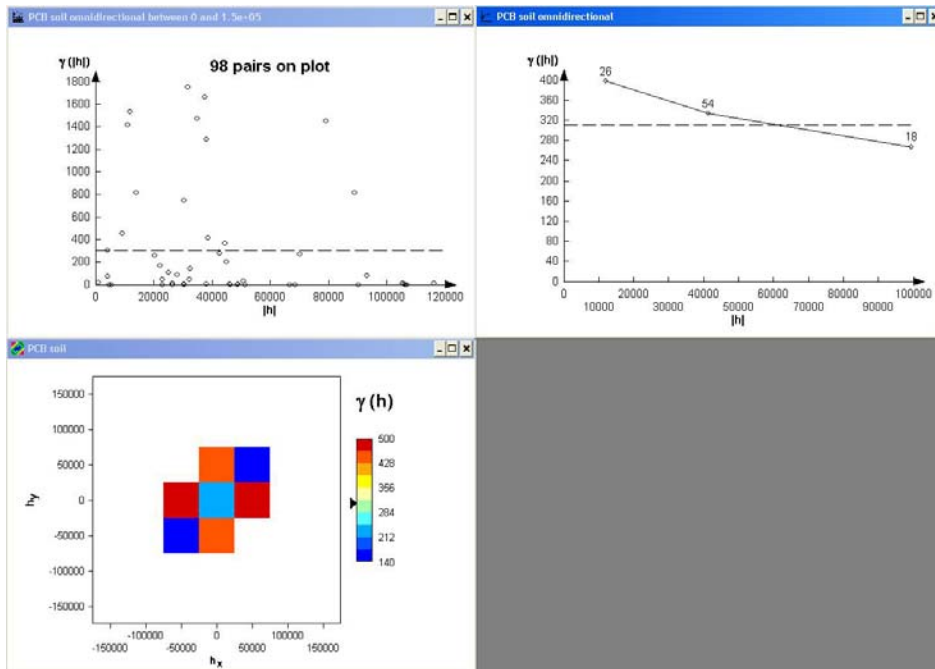


Figure 33: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2004-2006 in the Czech Republic

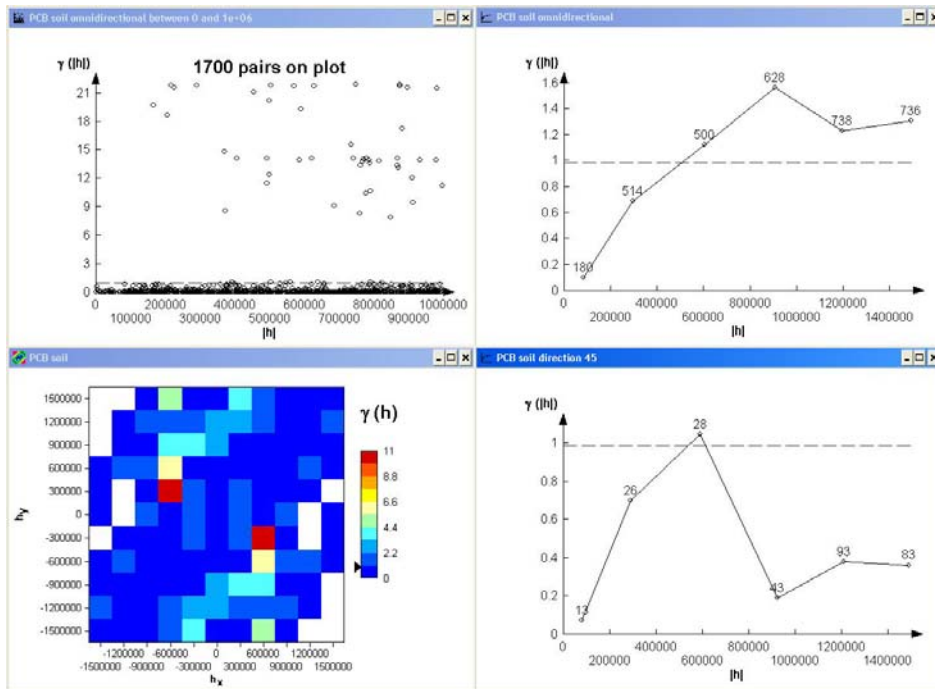


Figure 34: variogram cloud, variogram surface, and directional variograms for the PCB 118 soil concentrations, period 1998-2000

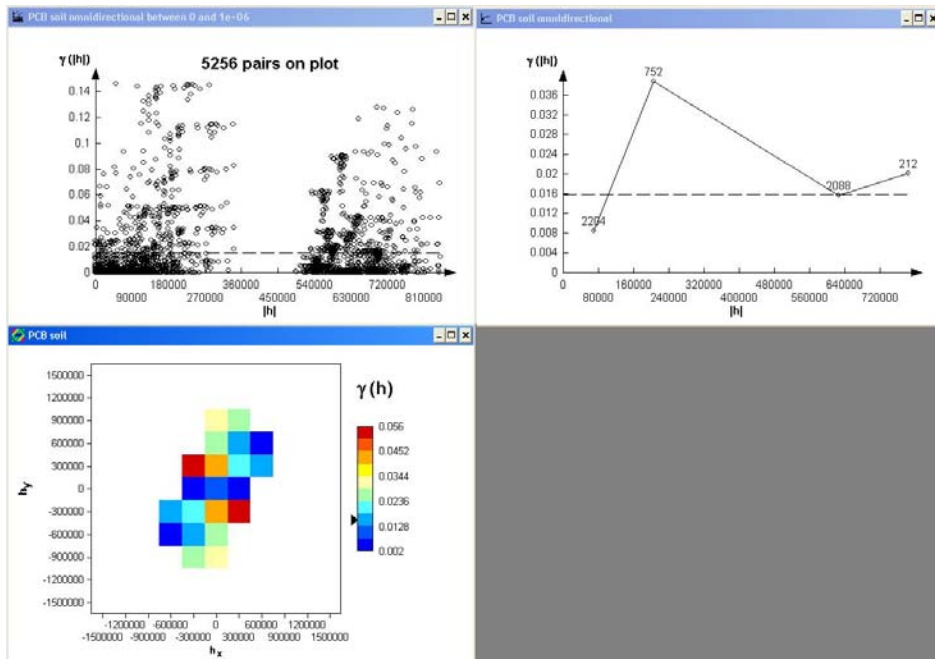


Figure 35: variogram cloud, variogram surface, and directional variograms for the PCB 118 soil concentrations, period 2001-2003

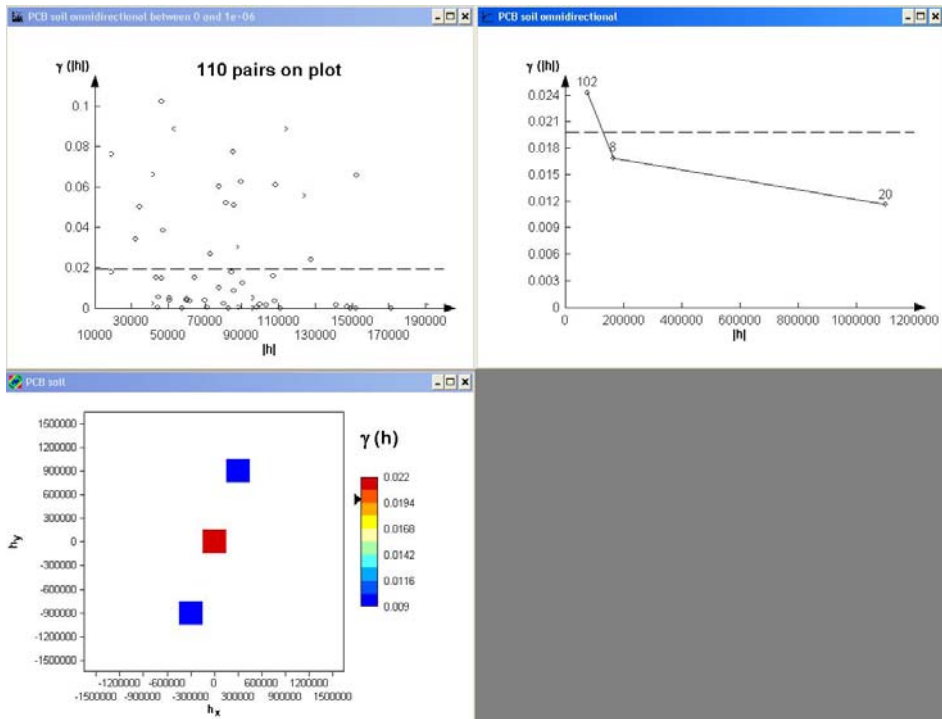


Figure 36: variogram cloud, variogram surface, and directional variograms for the PCB 118 soil concentrations, period 2005-2007

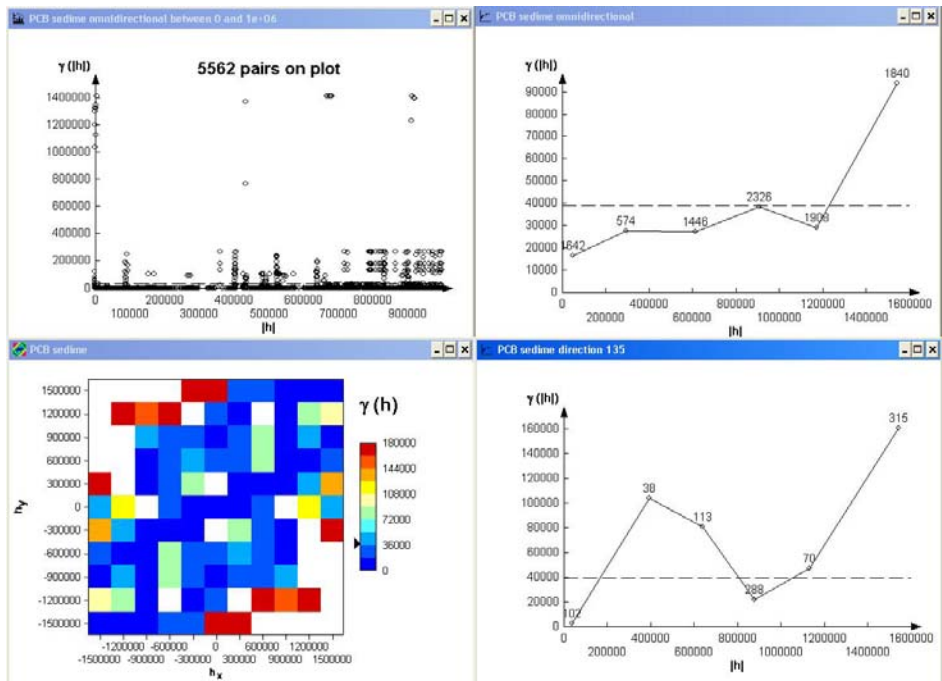


Figure 37: variogram cloud, variogram surface, and directional variograms for the PCBs sediment concentrations, period 2001-2003

		Tri-annual period	Near trend	Distant trend	No trend
PCBs	Soil	1996-1998	x		
		1997-1999	x		
		1998-2000	x		
		2000-2002	x	x	
		2001-2003		x	
		2002-2004			x
		2003-2005	x		
	Sediment	2004-2006			x
		1998-2000			x
		1999-2001	x	x	
		2001-2003		x	
	Water	2005-2007	x		
	Air	2005-2007	x	x	

Table 7: Summary table showing the geostatistical trends analyzed for PCBs

		Tri-annual period	Near trend	Distant trend	No trend
PCDD/Fs	Soil	1998-2000	x	x	
		2002-2004		x	
		2004-2006			x
	Sediment	1994-1996			x
		1996-1998			x
		1998-2000			x
		2001-2003			x
		2005-2007			x
	Water	2005-2007		x	
	Air	1994-1996			x
		1996-1998			x
		1997-1999			x
		2002-2004			x
		2004-2006			x

Table 8: Summary table showing the geostatistical trends analyzed for PCBs

5. Conclusions

Building a database of monitored concentrations of PCBs and PCDD/Fs entails considerable efforts. For the database presented here, an equivalent 2 man-months of bibliographic research by non-specialist scientific support personnel was invested, which gives an idea of the cost of compilations of data. The data collected show a relatively high heterogeneity, as environmental data from various local, small scale projects may have an enormous spatial and seasonal difference. The latter, for air and vegetation data, when sampled in different seasons may bring in a factor of 10 variations for dioxins, and little less for PCBs. In water the variability may be even higher. The variation in time shown by the data seems to be affected by the combination of different spatial clusters of data; also the evolution in time of the different experimental setups may have an influence. A time series of measurements at a single given location is the one way to assess the temporal trends of chemical concentrations. An option is to refer to existing (and increasingly available) monitoring stations such as the ones described in the German UBA report “Dioxine, Daten aus Deutschland - 5. Bericht” (<http://www.umweltdaten.de/publikationen/fpdf-l/3328.pdf>) or the EMEP network (although not all

[Europe is so far covered; Castro-Jiménez et al. 2009](#)). Different data series should be compared using normalized data or the time derivatives of concentrations.

Time series of data in Soils often highlight weak time trends because of the relatively long memory of this medium which reflects a more integral response to chemicals, while spatially distributed monitoring may yield interesting data sets (e.g. Meijer et al., 2003). As capturing spatial trends requires comparable, simultaneous measurements at different locations in space, which is presently far from being routinely done in the European Union or elsewhere for most chemicals, the compilation of published data in soils seems the most promising exercise. Despite the limitations highlighted above, the data collected from the literature reveal some (although weak) spatial trends that suggest the possibility of using data compilations for the evaluation of models of chemical fate at continental scale. Compilations of measured concentrations of PCDD/Fs were prepared for the European Commission in the past years (D.Buckley-Golder, Compilation of EU Dioxin exposure and health data, 1999: <http://ec.europa.eu/environment/dioxin/download.htm#CompilationofEUDioxinexposureandhealthdata>) in order to estimate the population exposure to these chemicals; however, it does not seem that more recent compilation exercises have been undertaken in Europe.

In order to obtain similar performances in other media, longer-memory, integral observations should be collected: increasingly, indirect measurement methods are being proposed such as passive samplers for atmospheric gas-phase concentration and the monitoring of lipids e.g. from animal tissues or dairy produce, which deliver a sort of “time-integral” of exposure as they reflect exposure over rather long periods.

In addition, having a database of measured environmental concentrations (MEC) may be very useful in order to complement and corroborate predicted environmental concentrations (PEC) that are employed in the exposure and risk assessment under the REACH regulation, particularly for persistent, bioaccumulative and toxic (PBT) chemicals. Such a dataset may help building confidence, or highlight limitations since predictive models seem to be able to reproduce observations only to a limited extent. However, the data included in the present work should be further validated for this purpose. It is therefore recommended that specialised staff to be dedicated to the data suitability analysis for this purpose or that available data validation procedures under Chemical safety assessment (CSA) are implemented. Klimisch *et al* (1997) developed a scoring system to assess the reliability of data, particularly from toxicological and ecotoxicological studies, that may be extended to physico-chemical and environmental fate and behaviour studies. This is the method currently employed under REACH for data validation. The present data compilations it is in any case a first step of this direction.

ANNEX 1. Mobile sum of samples of each year \pm 1 year maps

PCBs

5.1.1 Air

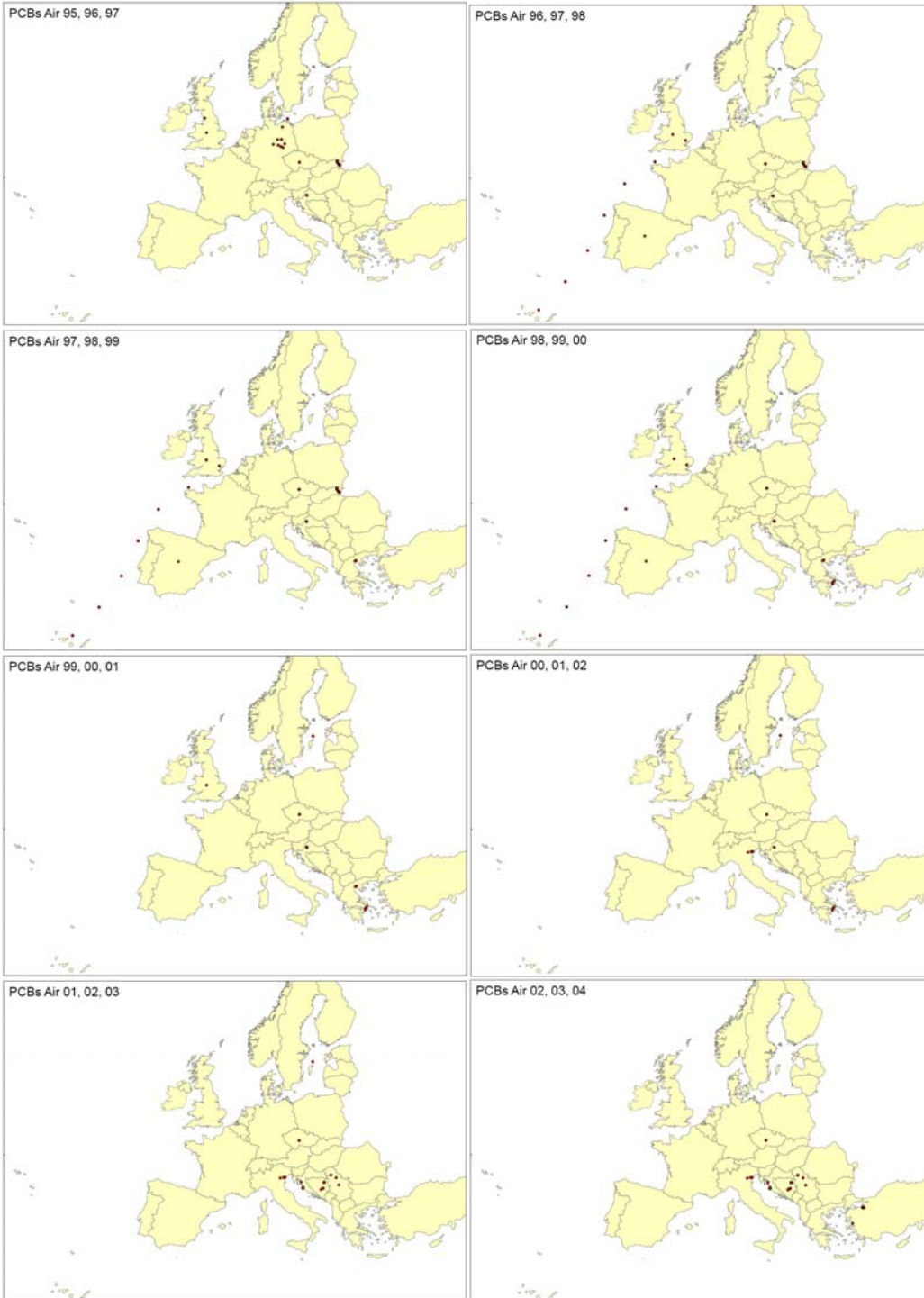
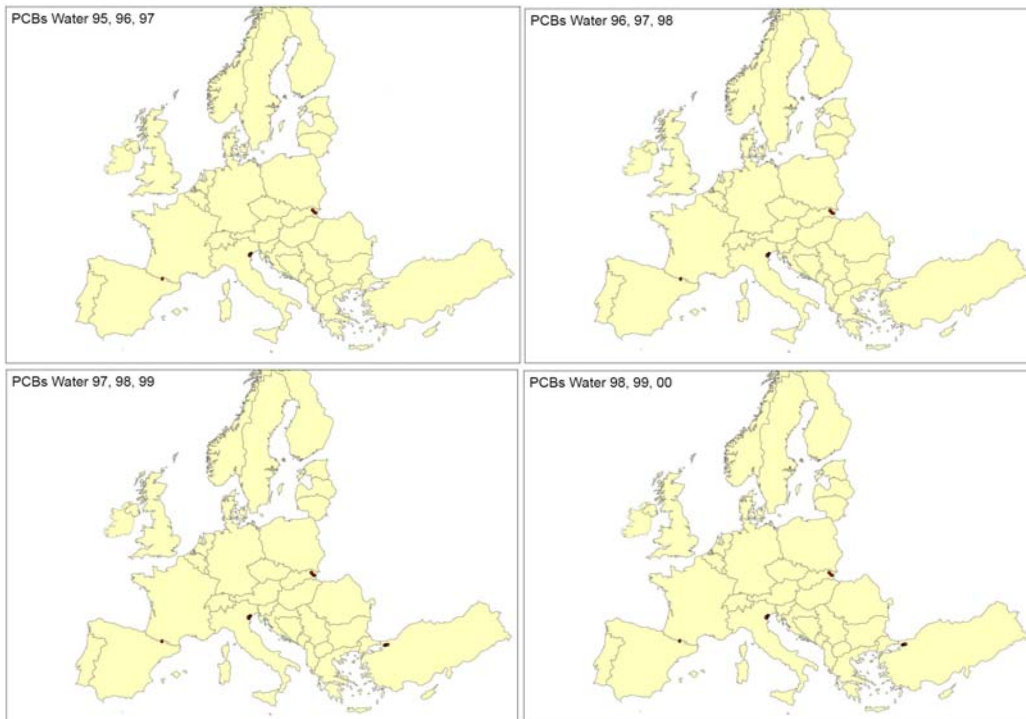




Figure 38: Mobile sum of samples of each year \pm 1 year maps, PCBs in air

5.1.2 Water



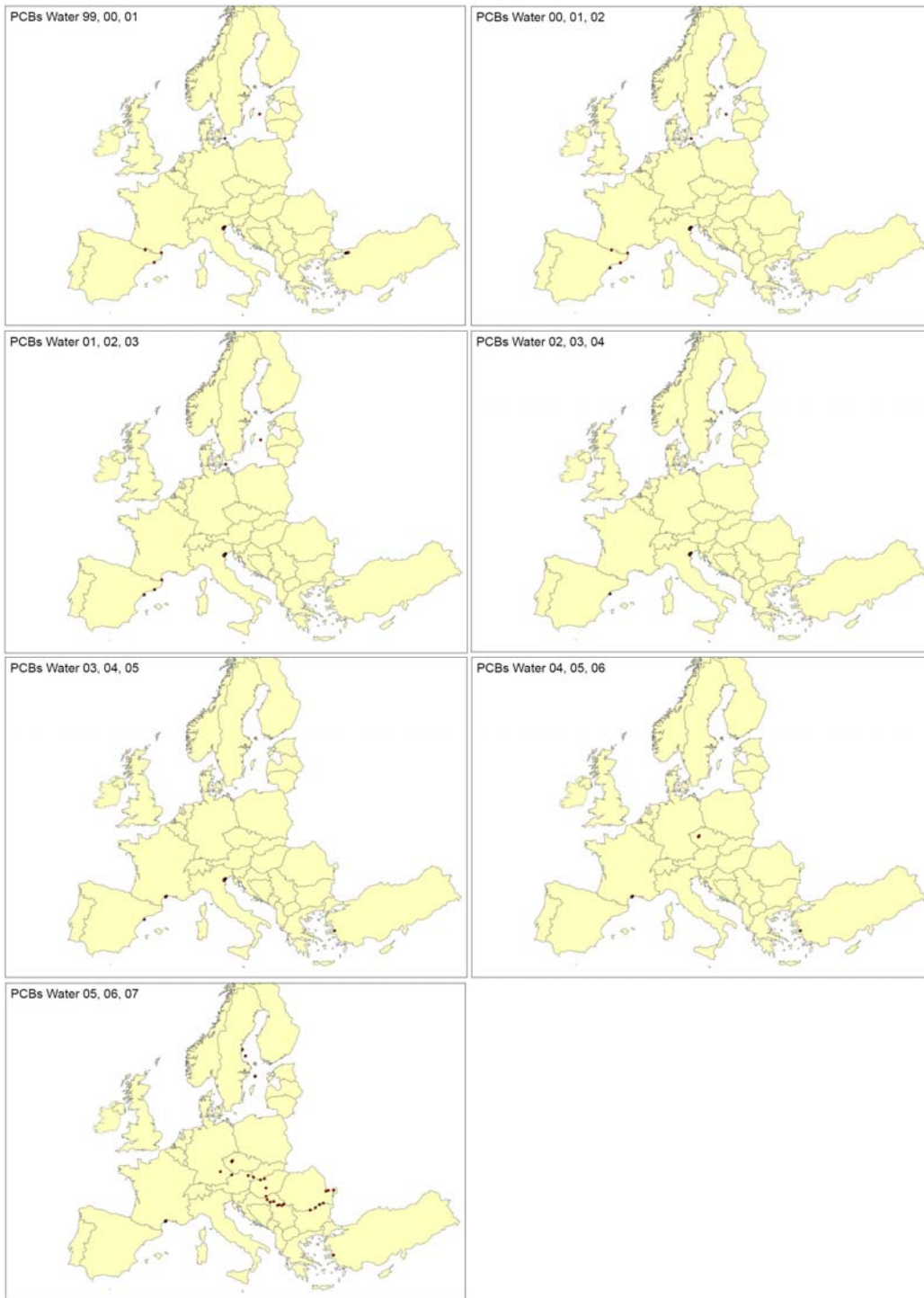
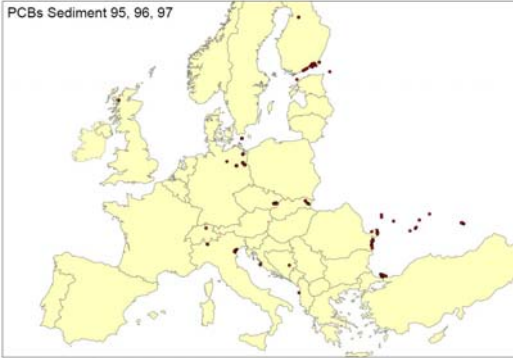


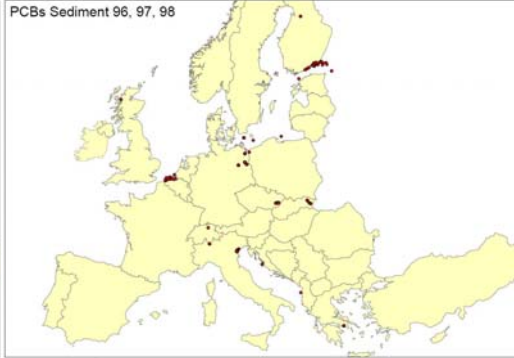
Figure 39: Mobile sum of samples of each year ± 1 year maps, PCBs in water

5.1.3 Sediment

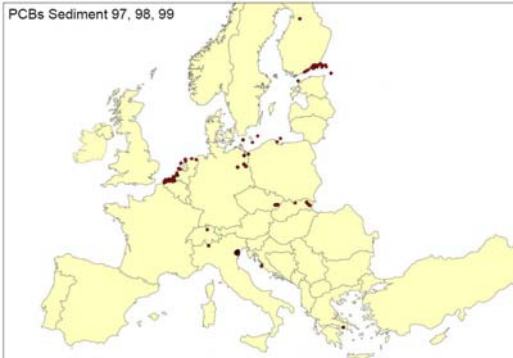
PCBs Sediment 95, 96, 97



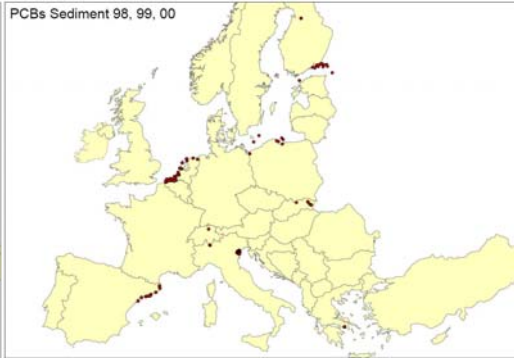
PCBs Sediment 96, 97, 98



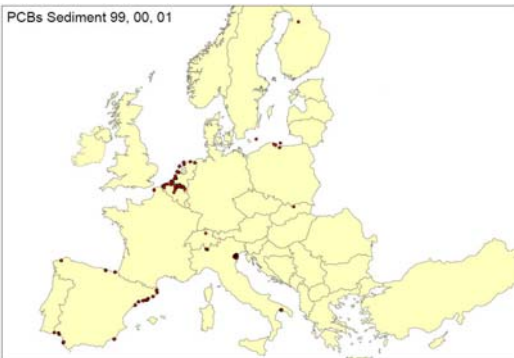
PCBs Sediment 97, 98, 99



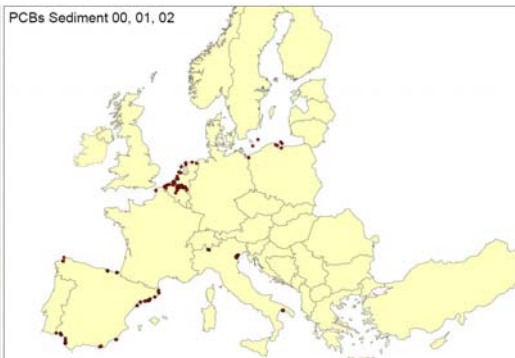
PCBs Sediment 98, 99, 00



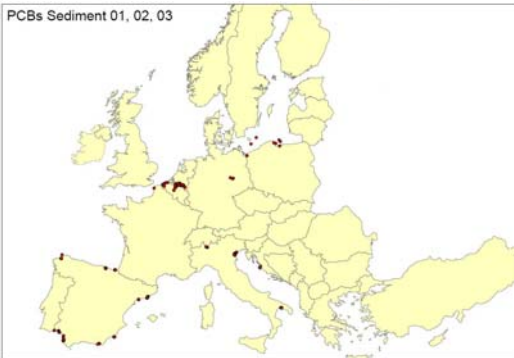
PCBs Sediment 99, 00, 01



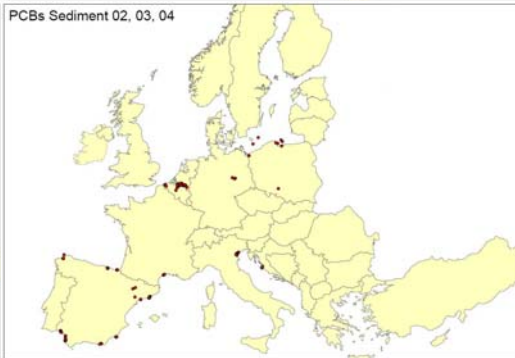
PCBs Sediment 00, 01, 02



PCBs Sediment 01, 02, 03



PCBs Sediment 02, 03, 04



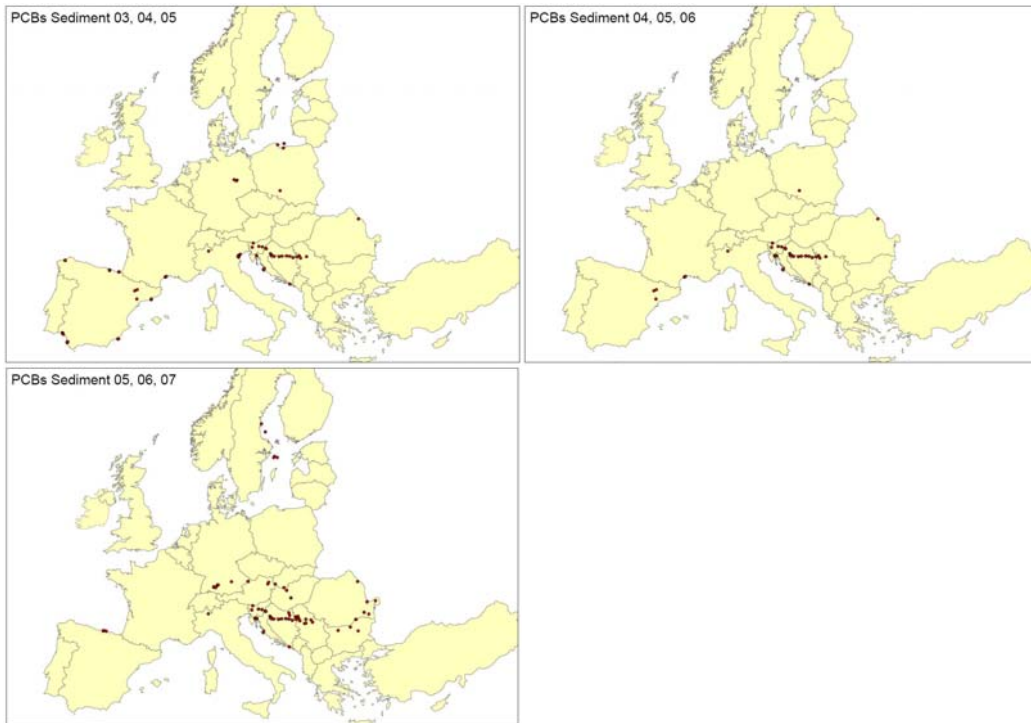


Figure 40: Mobile sum of samples of each year ± 1 year maps, PCBs in sediment

5.1.4 Soil

PCBs Soil 95, 96, 97



PCBs Soil 96, 97, 98



PCBs Soil 97, 98, 99



PCBs Soil 98, 99, 00



PCBs Soil 99, 00, 01



PCBs Soil 00, 01, 02



PCBs Soil 01, 02, 03



PCBs Soil 02, 03, 04

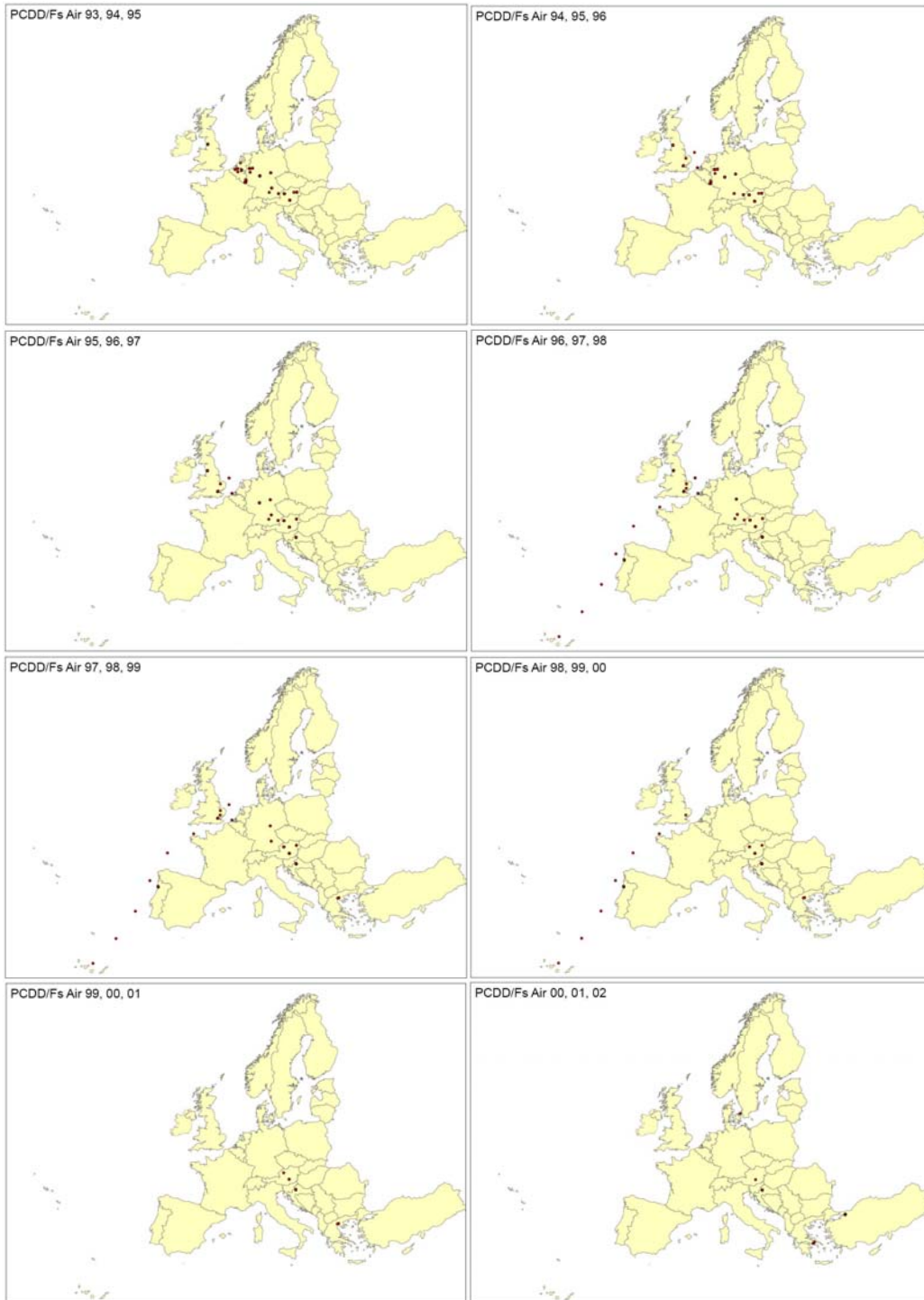




Figure 41: Mobile sum of samples of each year \pm 1 year maps, PCBs in soil

5.2 PCDD/Fs

5.2.1 Air



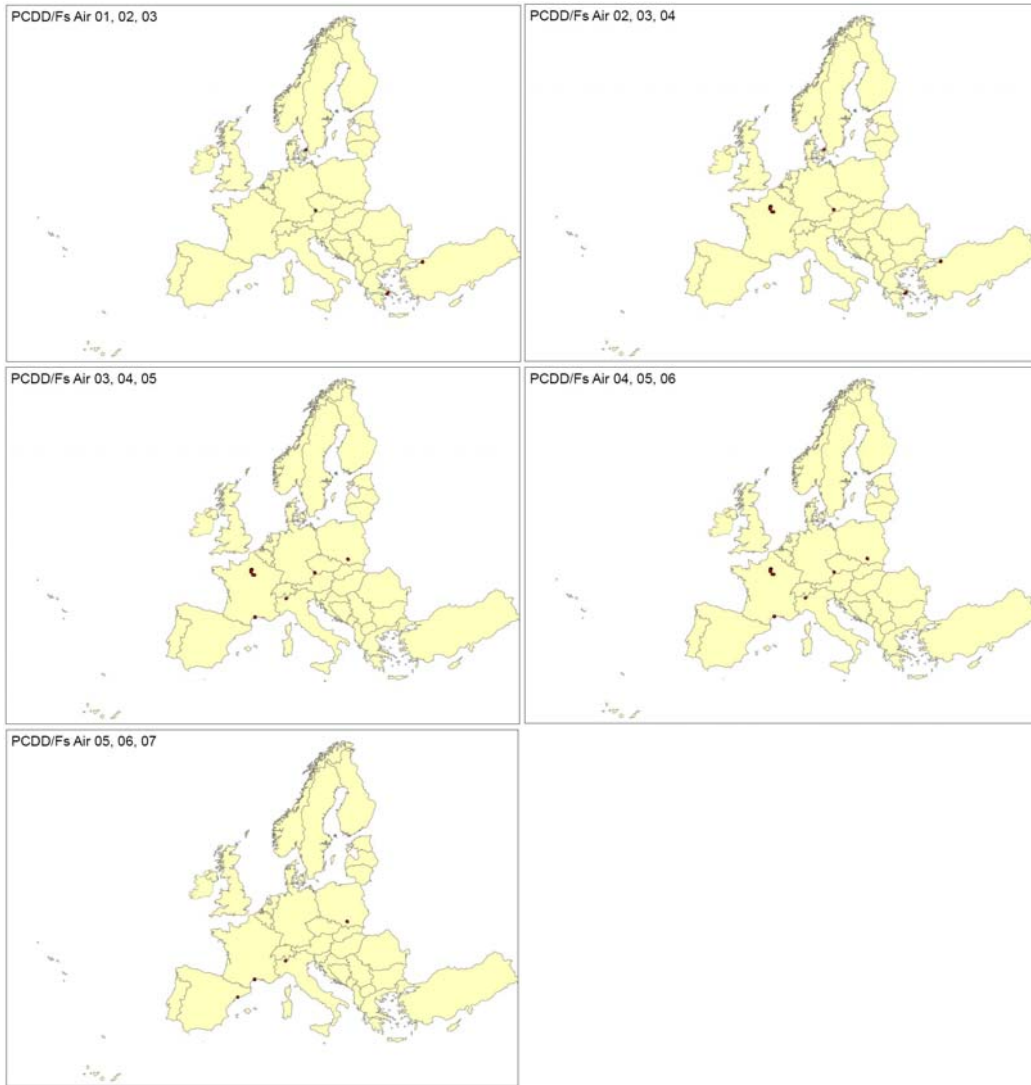


Figure 42: Mobile sum of samples of each year \pm 1 year maps, PCDD/Fs in air

5.2.2 Water

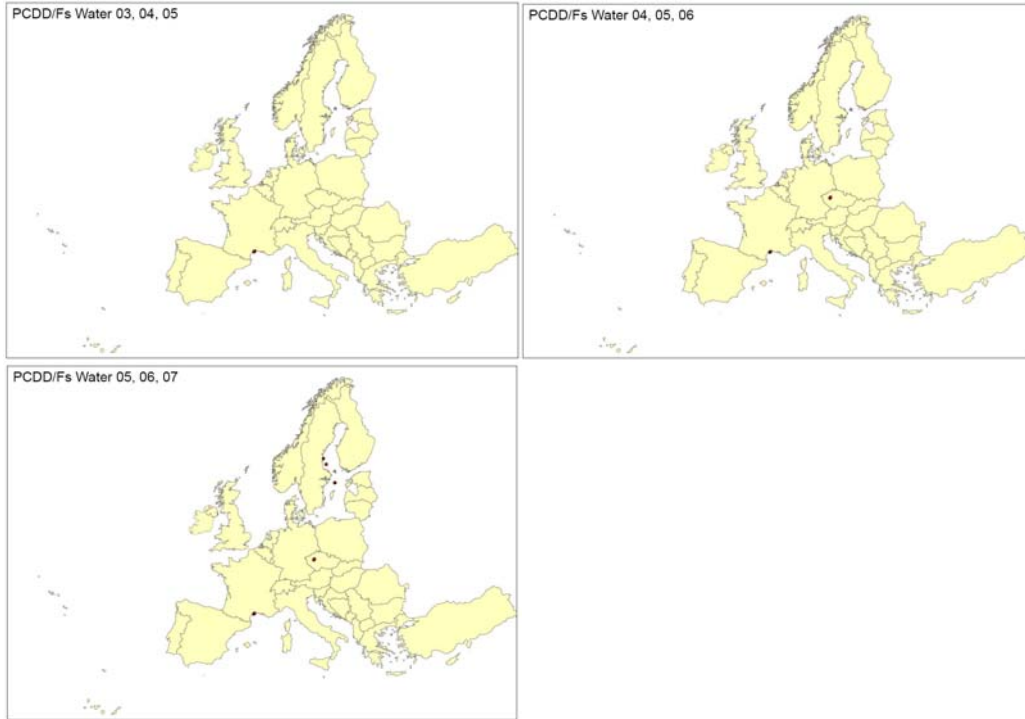
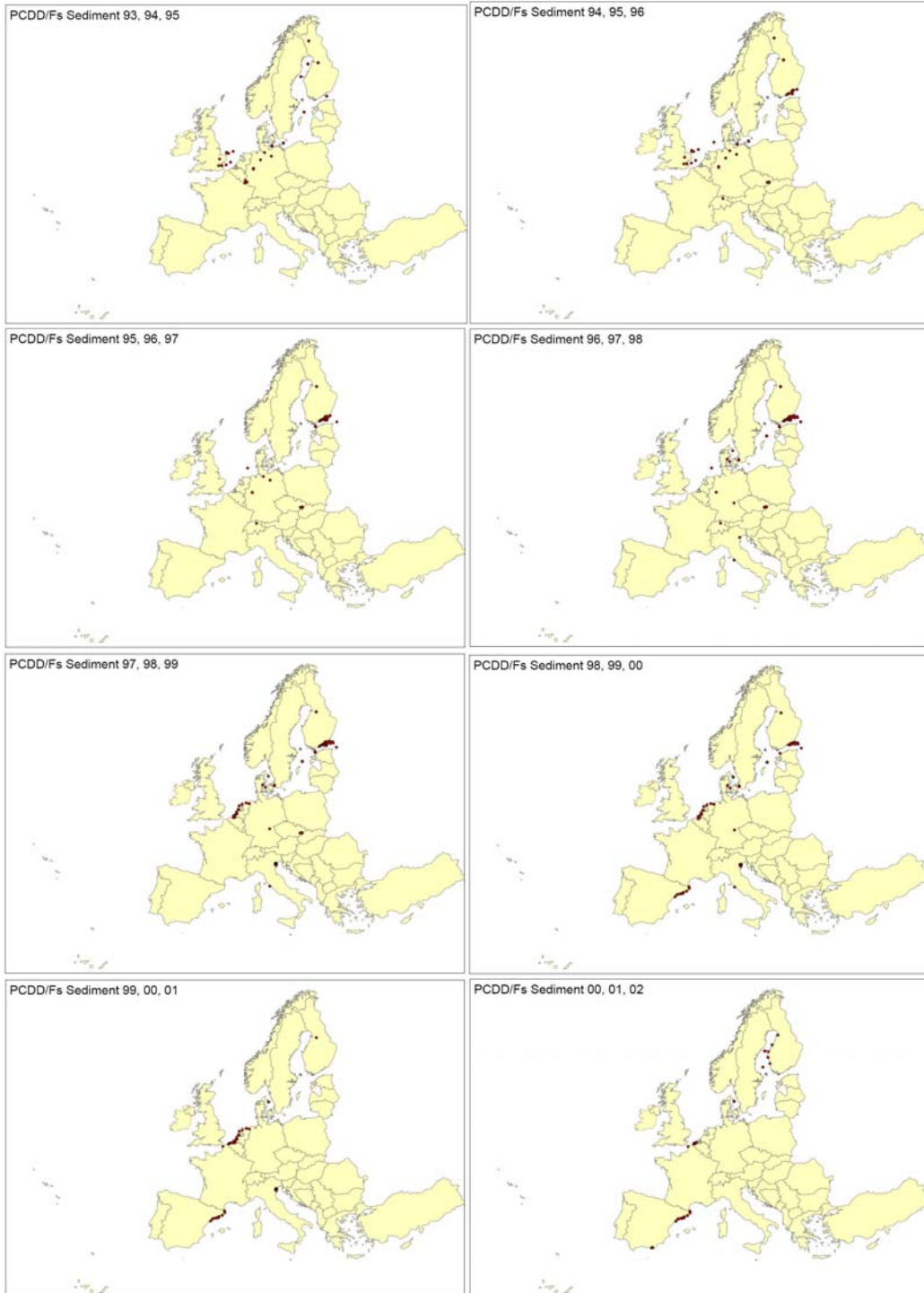


Figure 43: Mobile sum of samples of each year ± 1 year maps, PCDD/Fs in water

5.2.3 Sediment



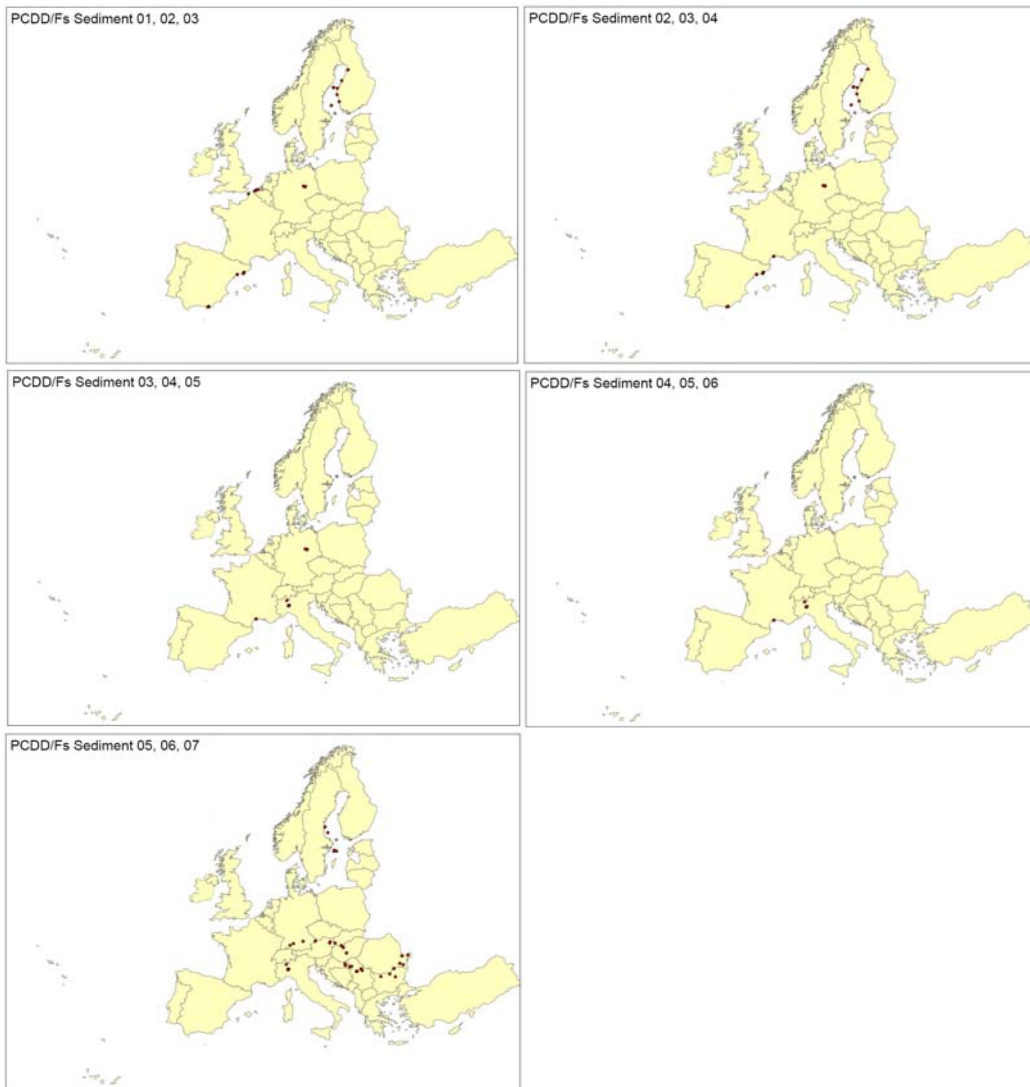


Figure 44: Mobile sum of samples of each year \pm 1 year maps, PCDD/Fs in sediment

5.2.4 Soil

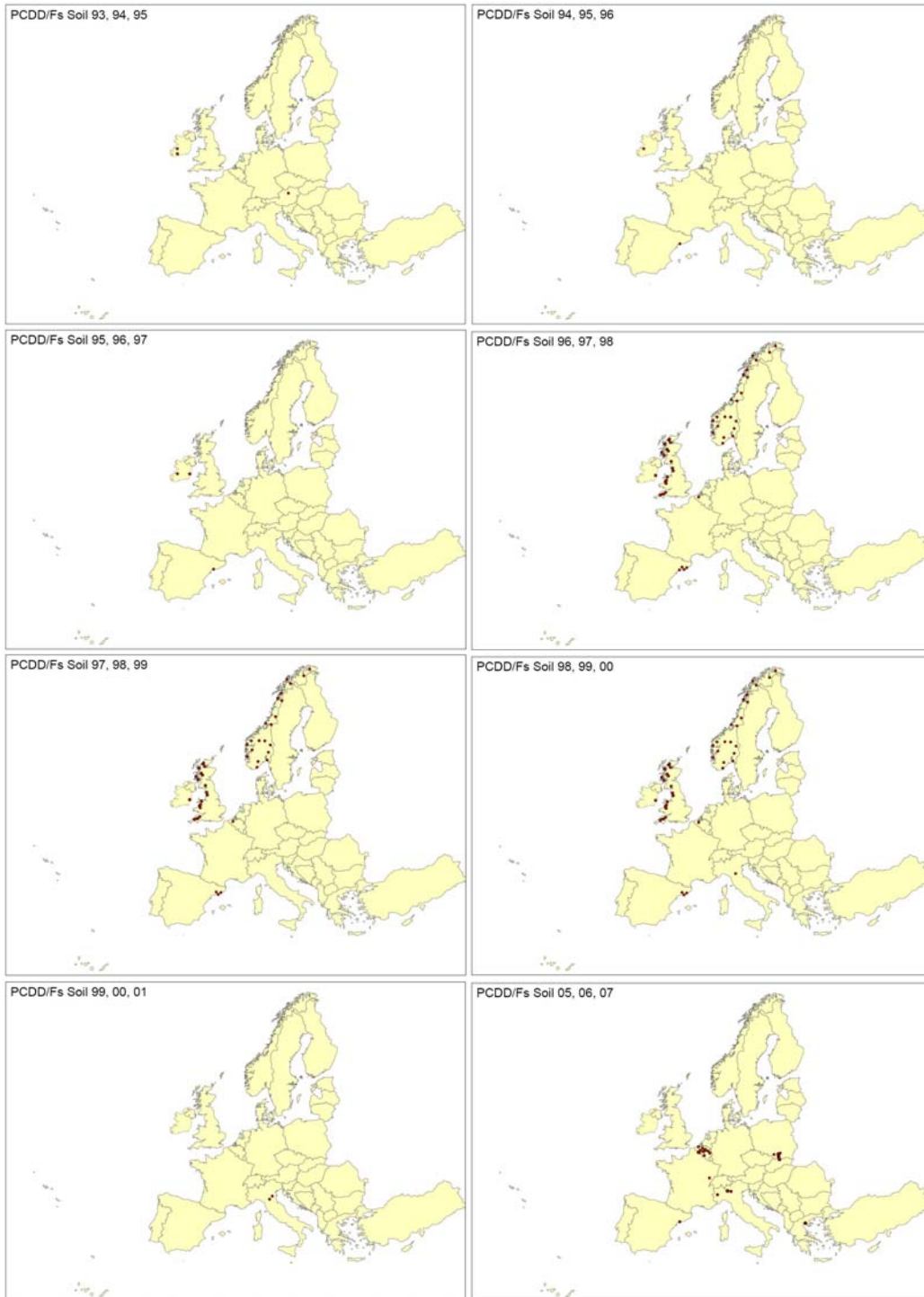




Figure 45: Mobile sum of samples of each year \pm 1 year maps, PCDD/Fs in soil

ANNEX 2. Results of the geostatistical analysis

5.3 PCBs

The geostatistical studies were done for soil in the following years:

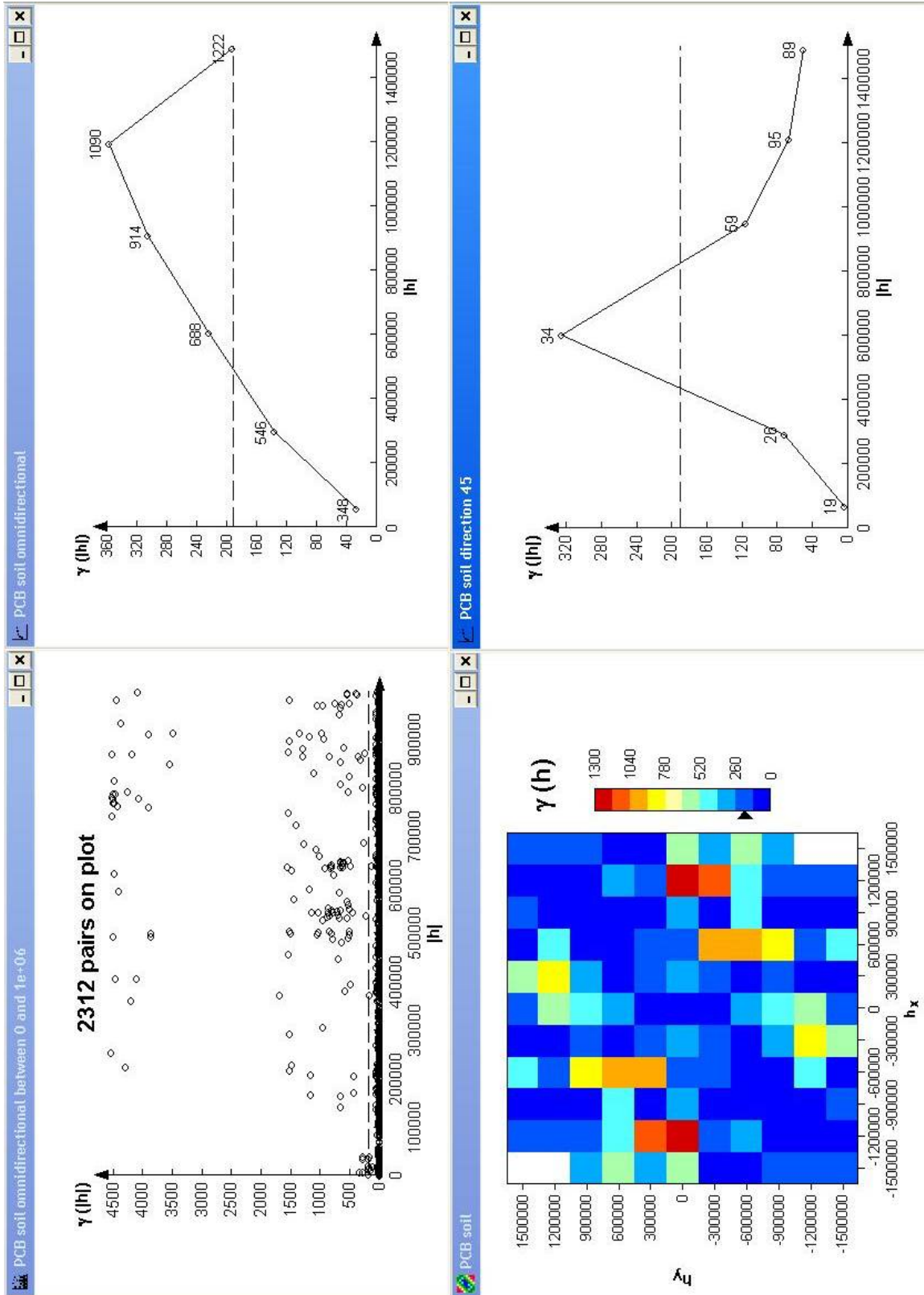


Figure 46: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 1996-1998

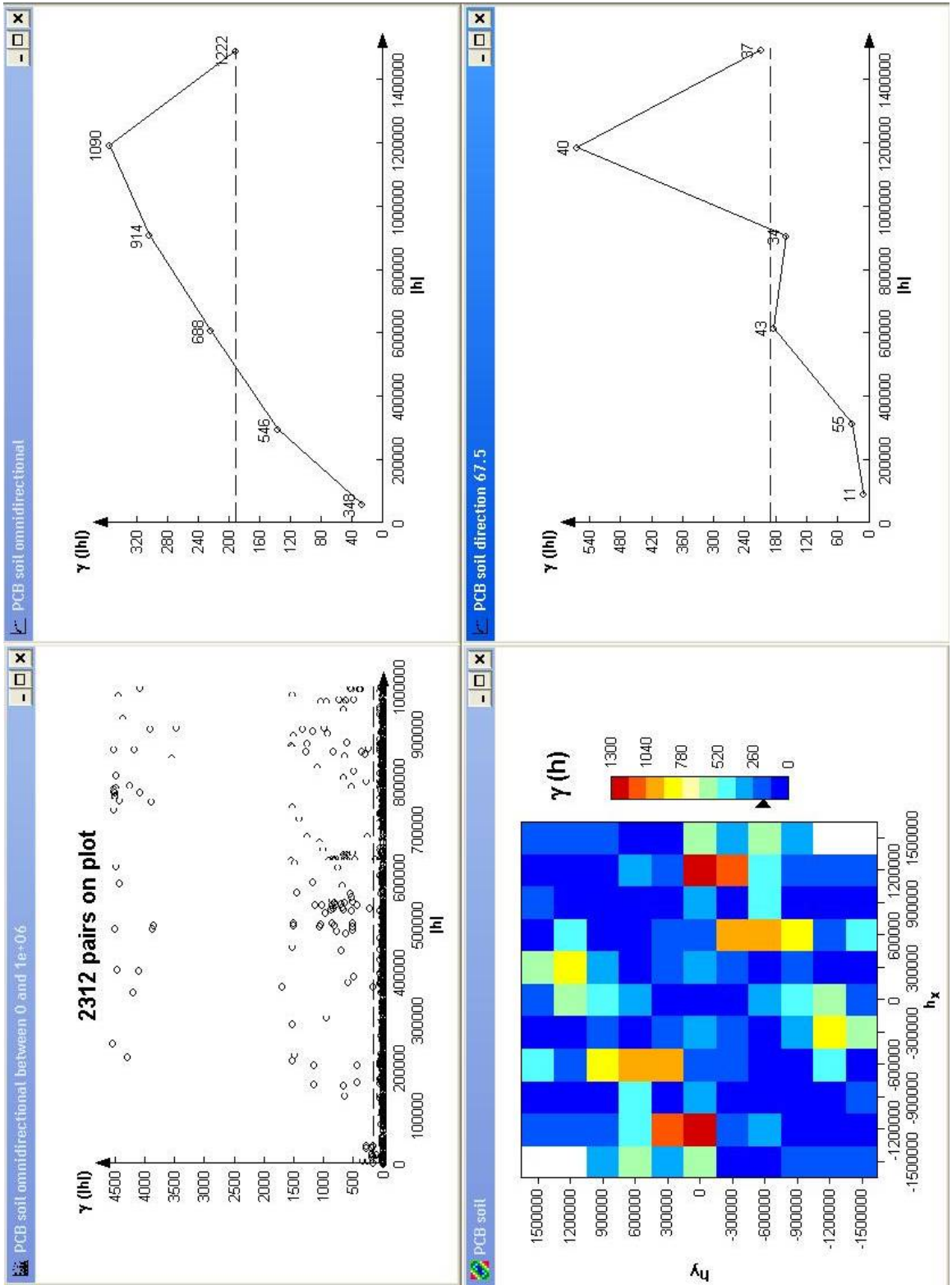


Figure 47: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 1997-1999

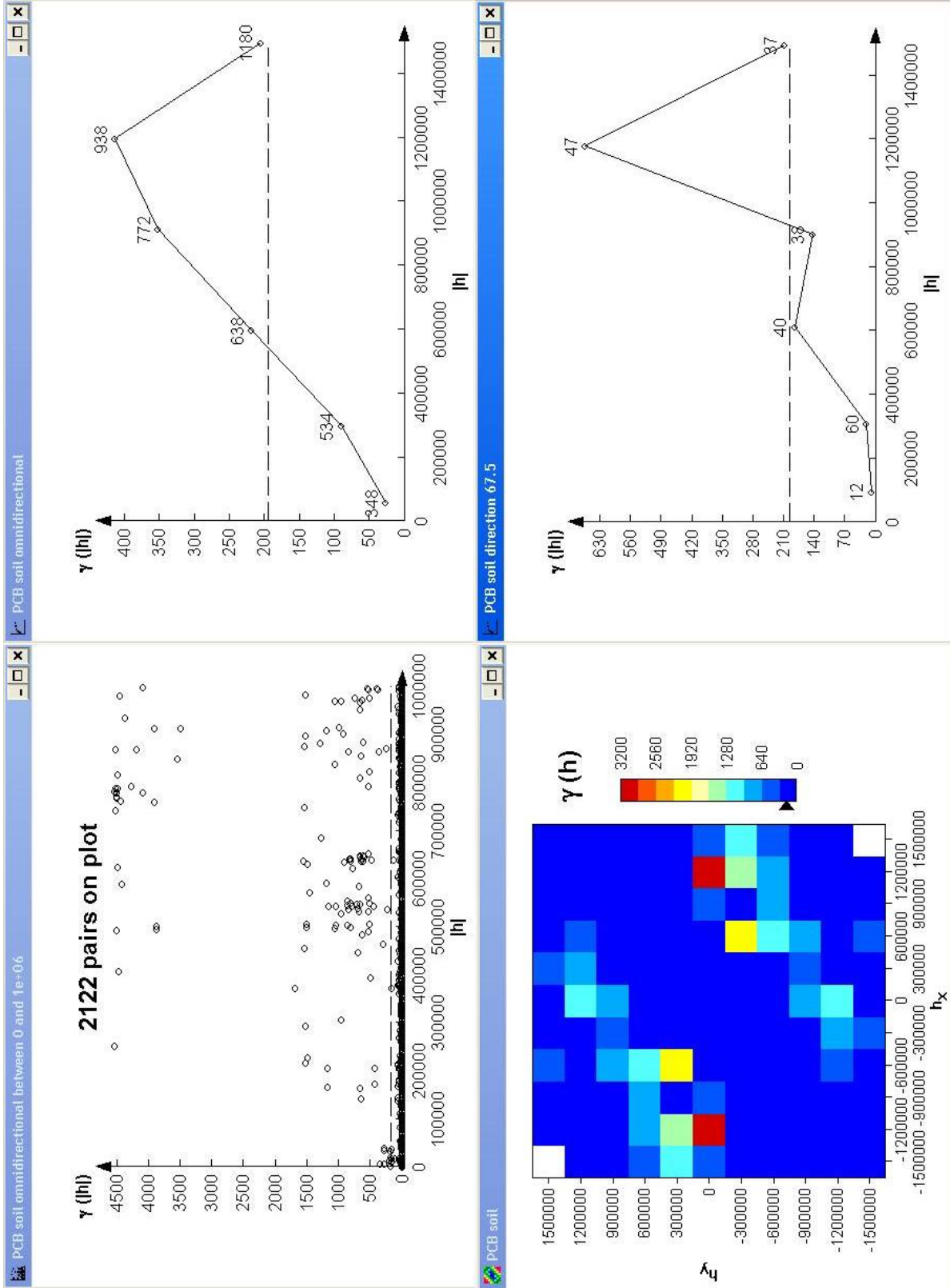


Figure 48: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 1998-2000

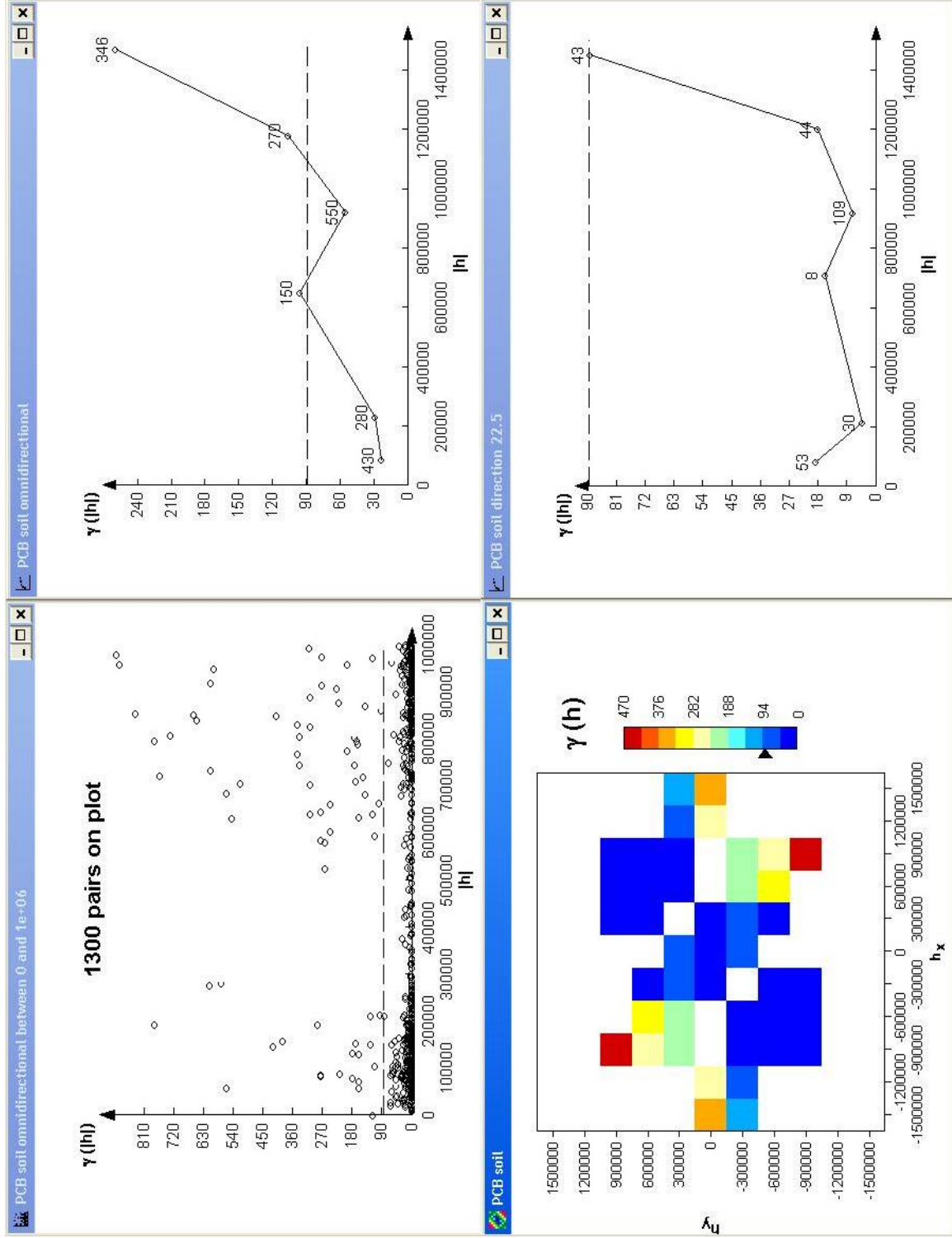


Figure 49: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2000-2002

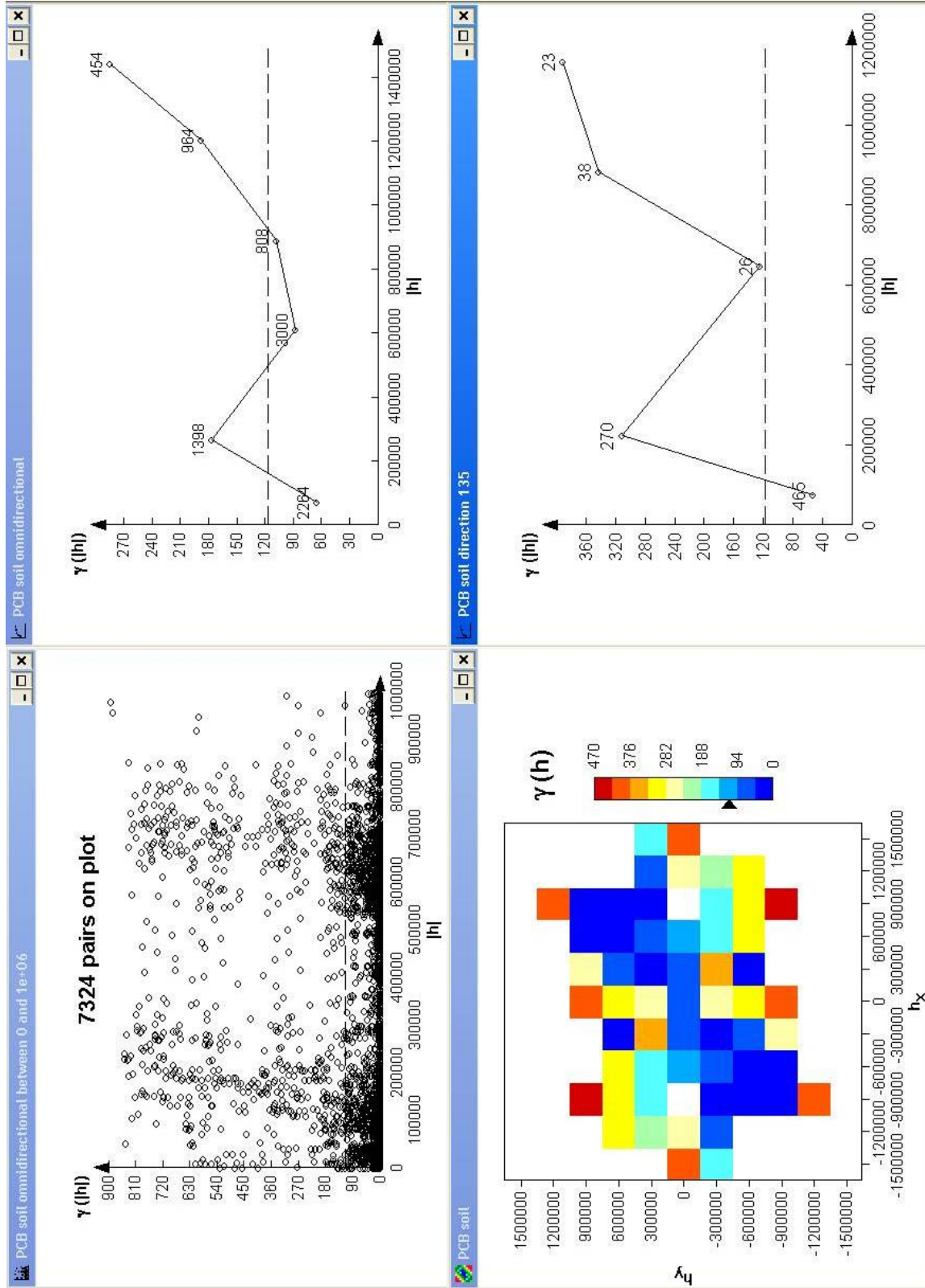


Figure 50: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2001-2003

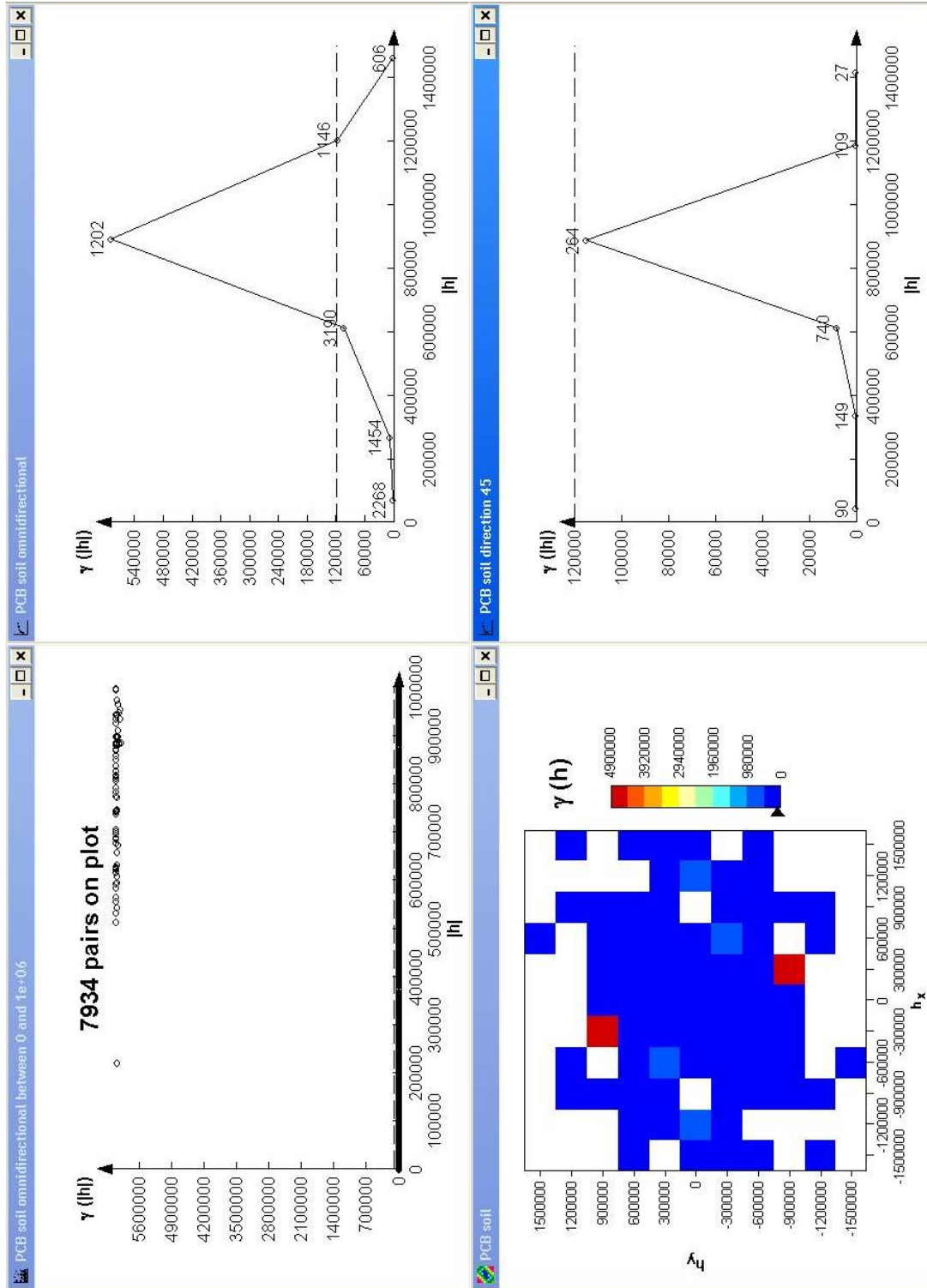


Figure 51: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2002-2004

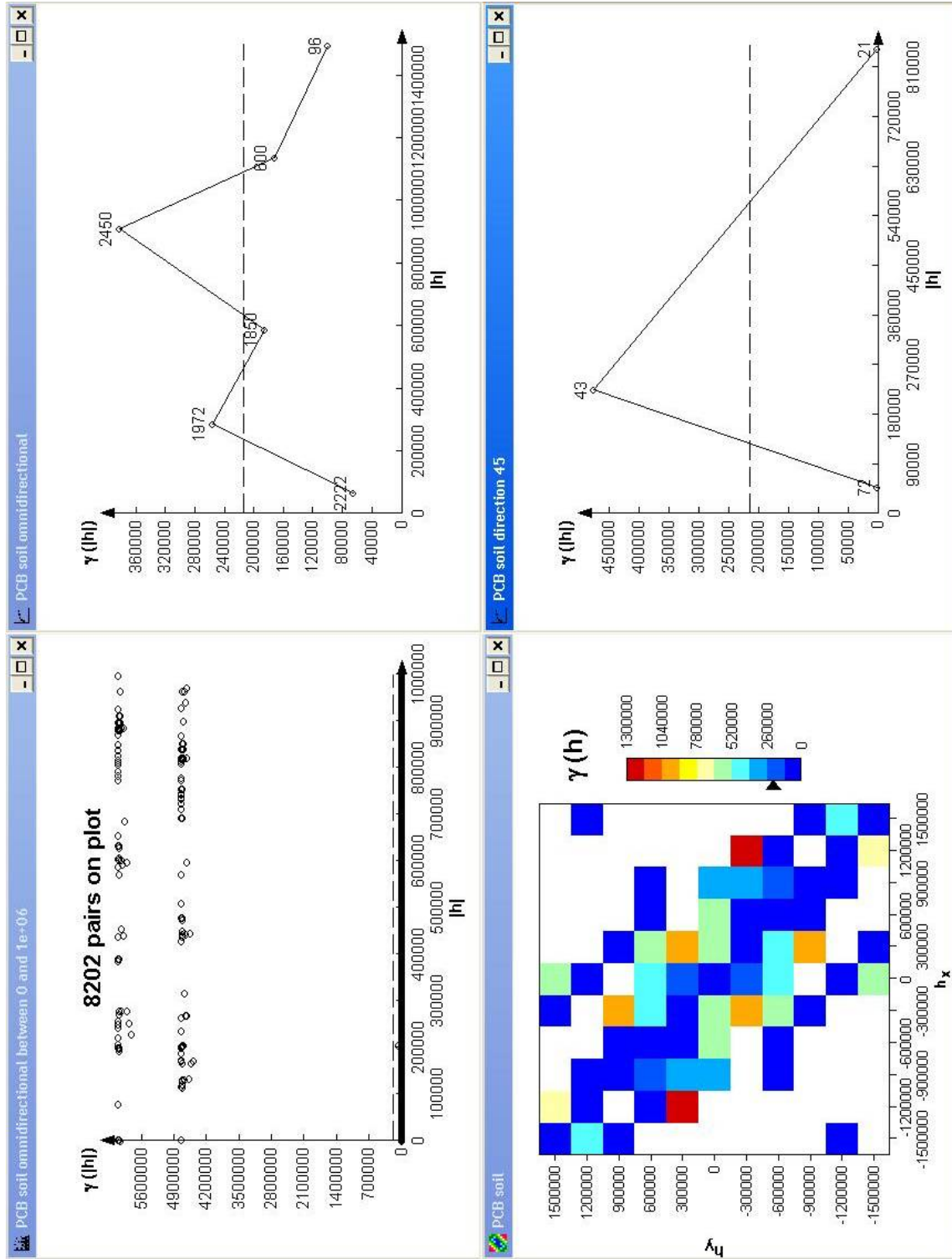


Figure 52: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2003-2005

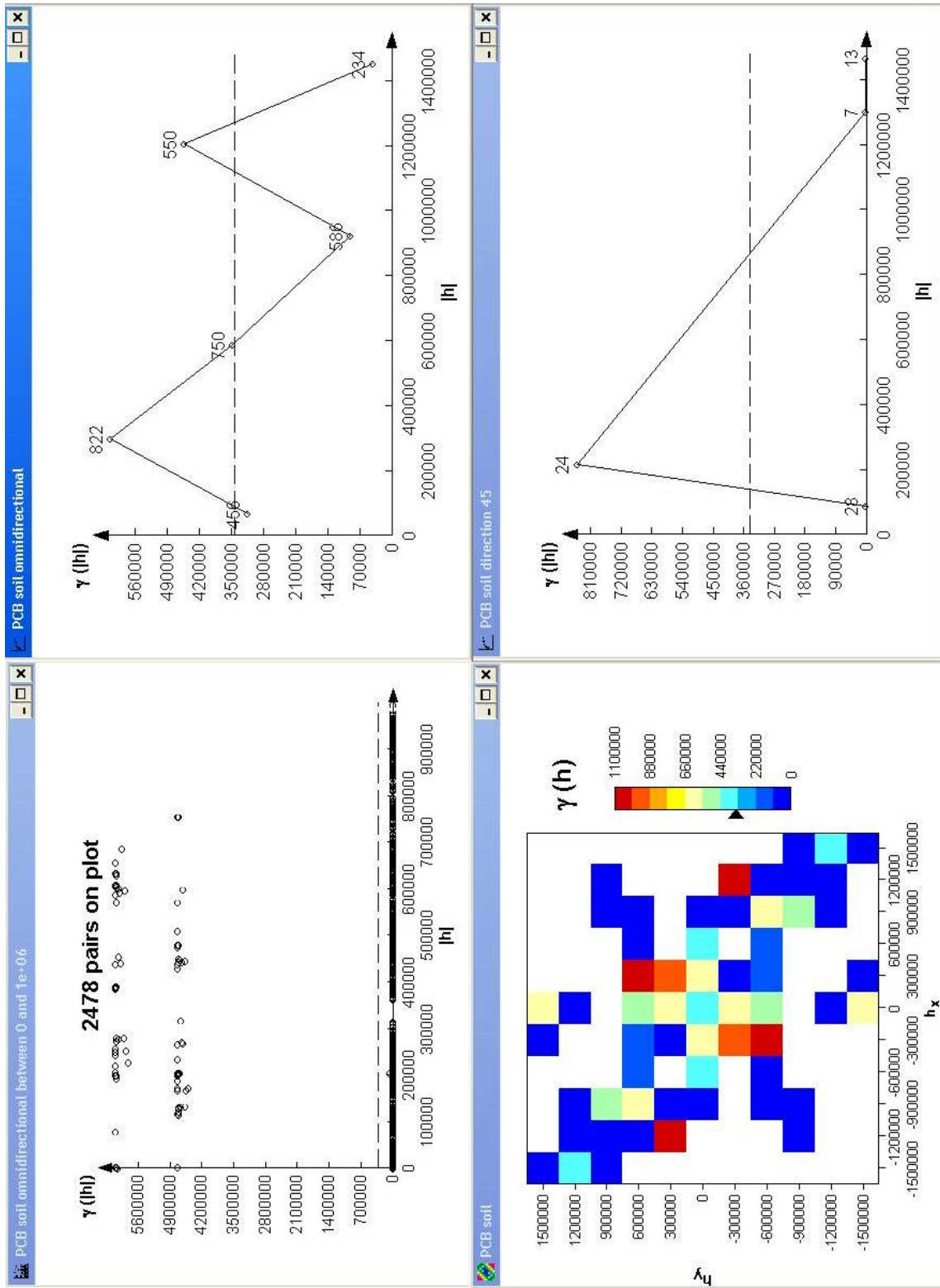


Figure 53: variogram cloud, variogram surface, and directional variograms for PCBs soil concentrations, period 2004-2006

The geostatistical studies done for sediments have been selected for the three years periods where the data has a better geographic coverage:

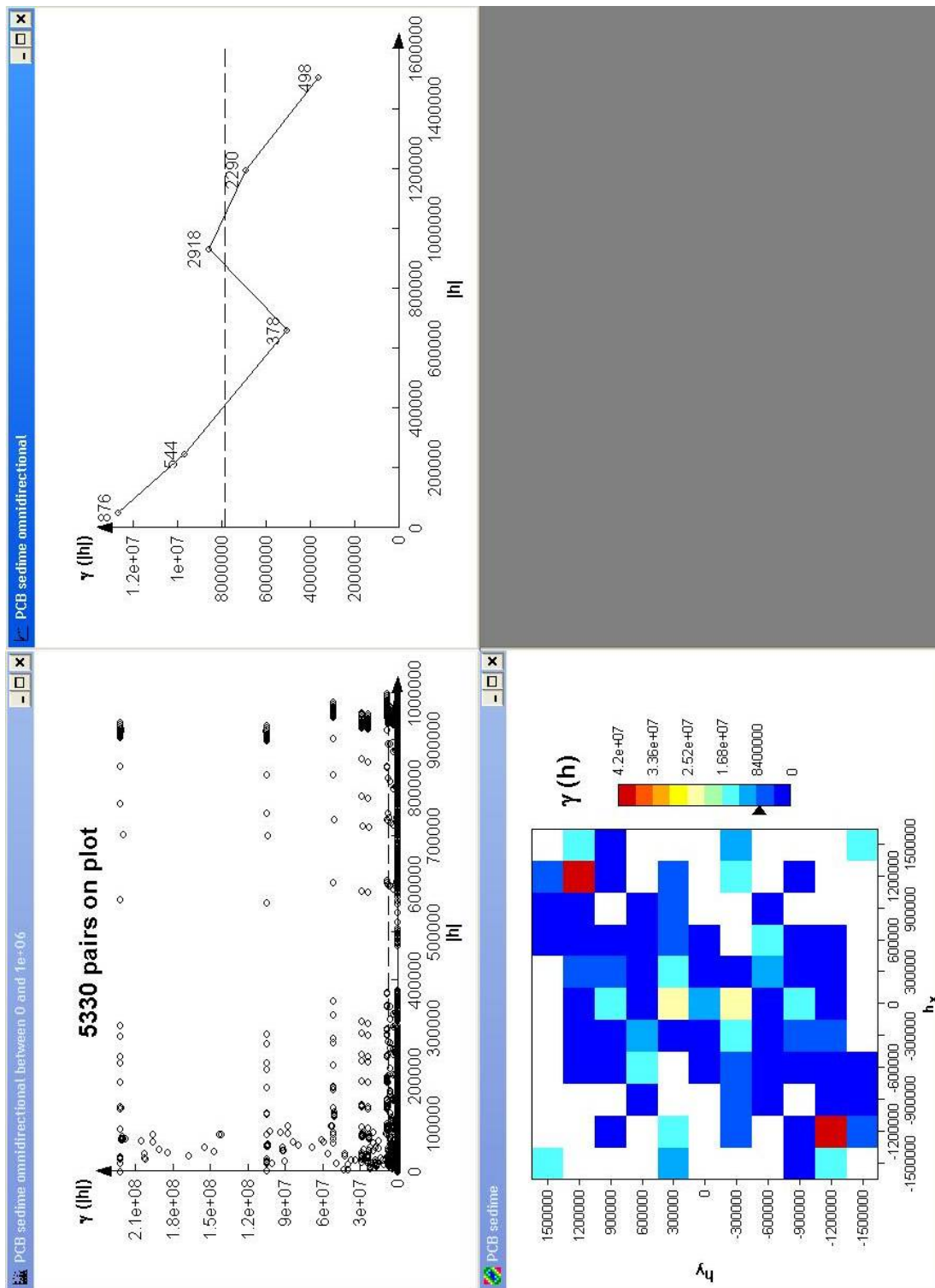


Figure 54: variogram cloud, variogram surface, and directional variograms for PCBs sediment concentrations, period 1998-2000

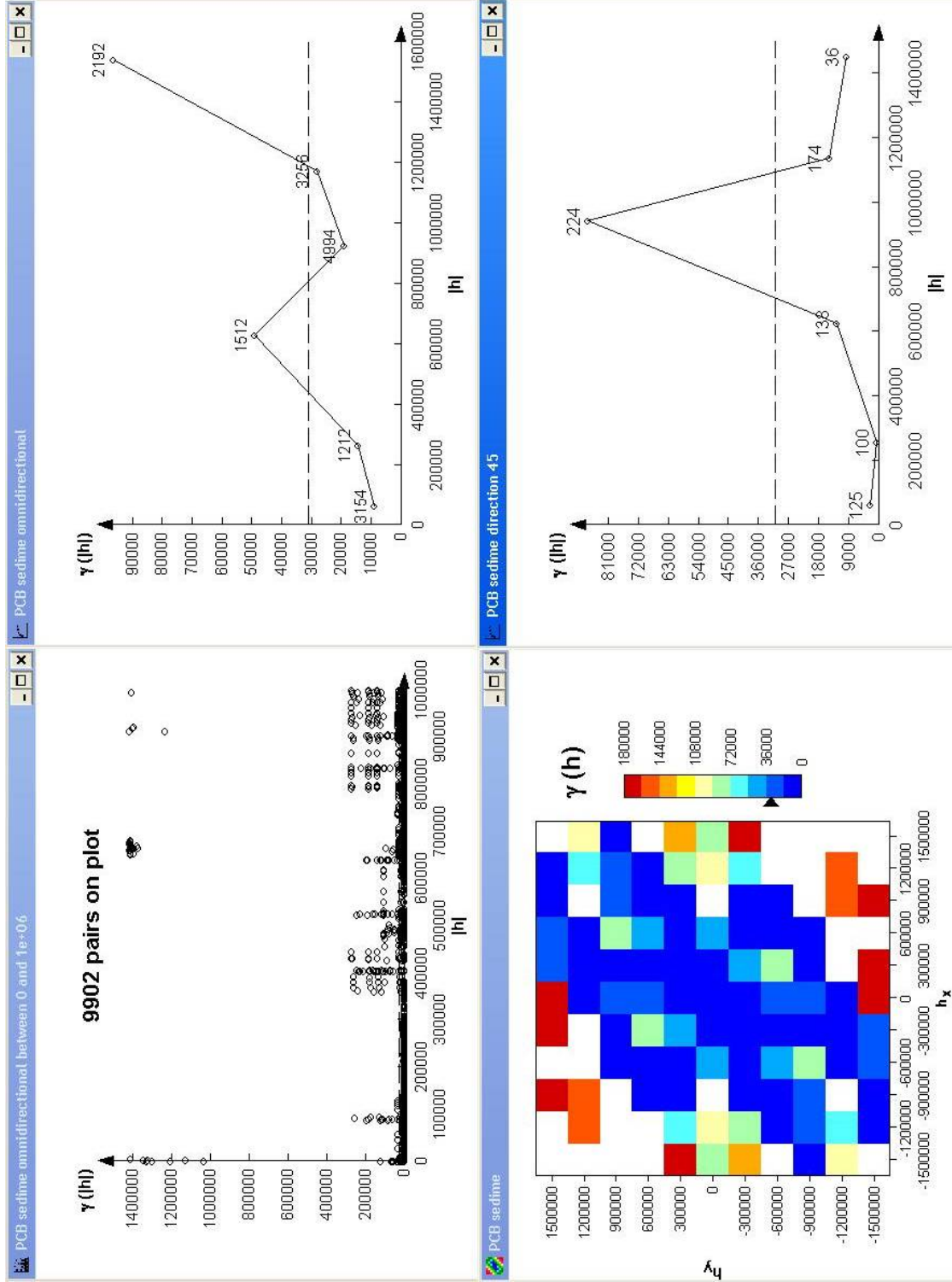


Figure 55: variogram cloud, variogram surface, and directional variograms for PCBs sediment concentrations, period 1999-2001

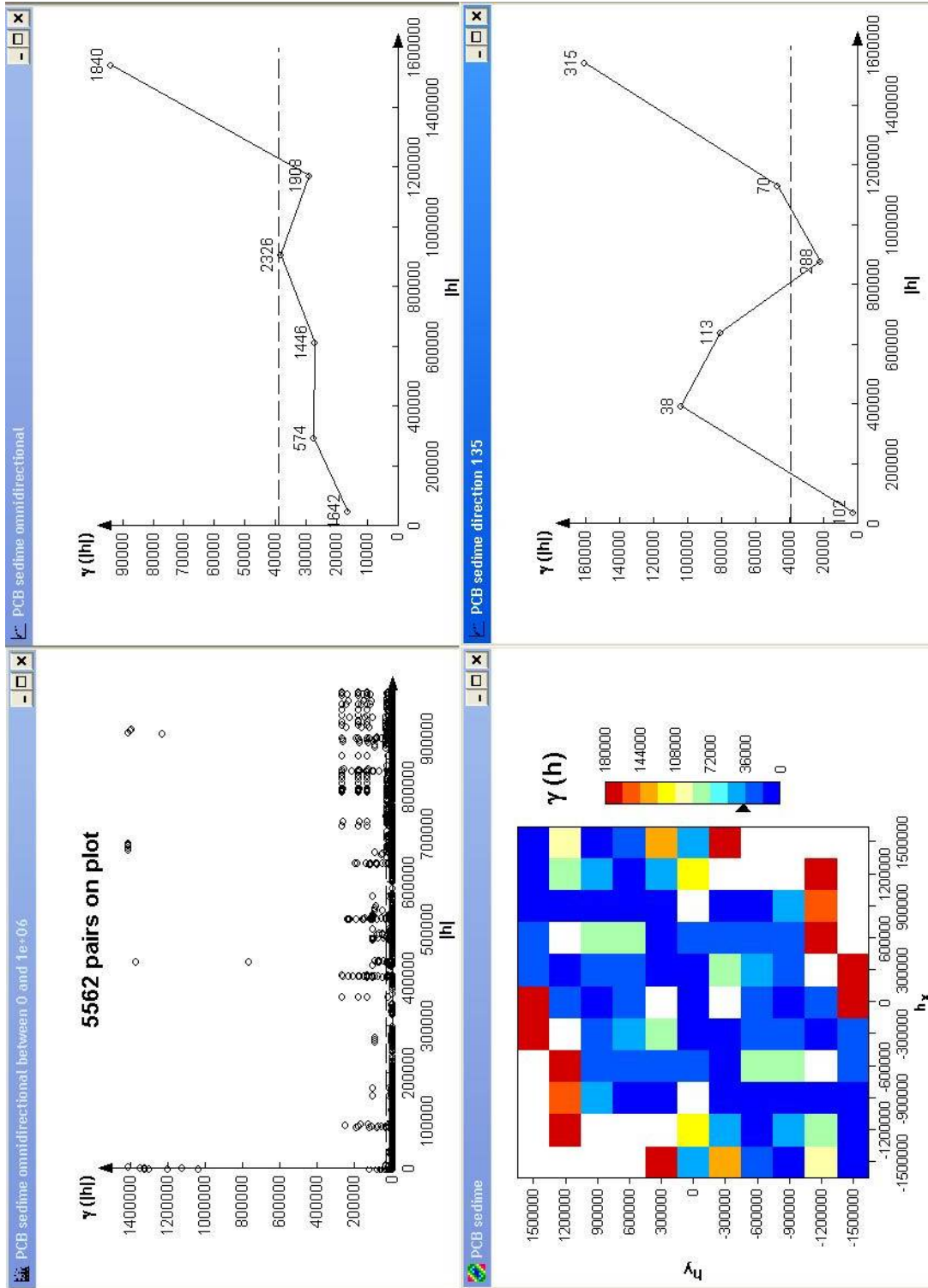


Figure 56: variogram cloud, variogram surface, and directional variograms for PCBs sediment concentrations, period 2001-2003

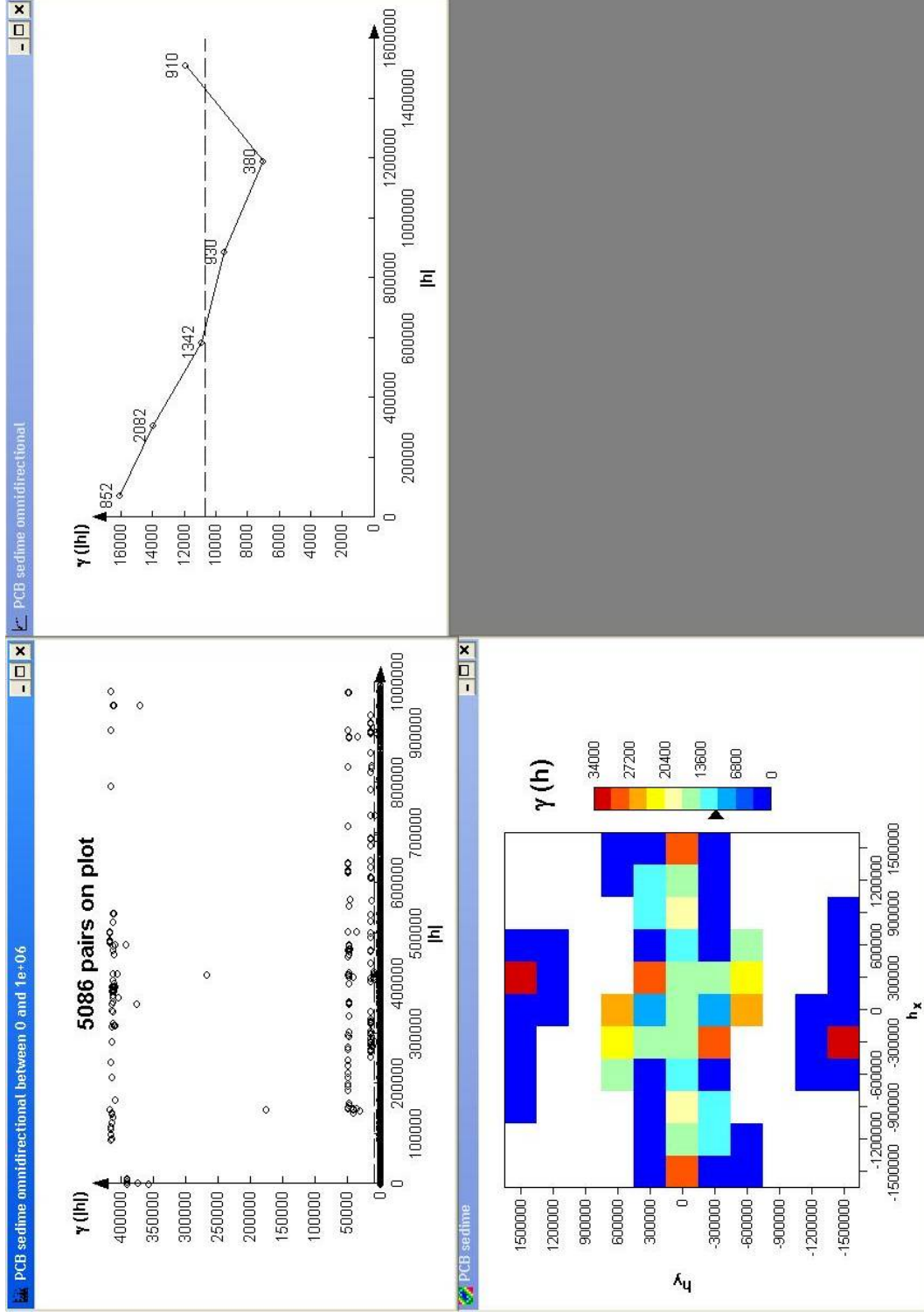
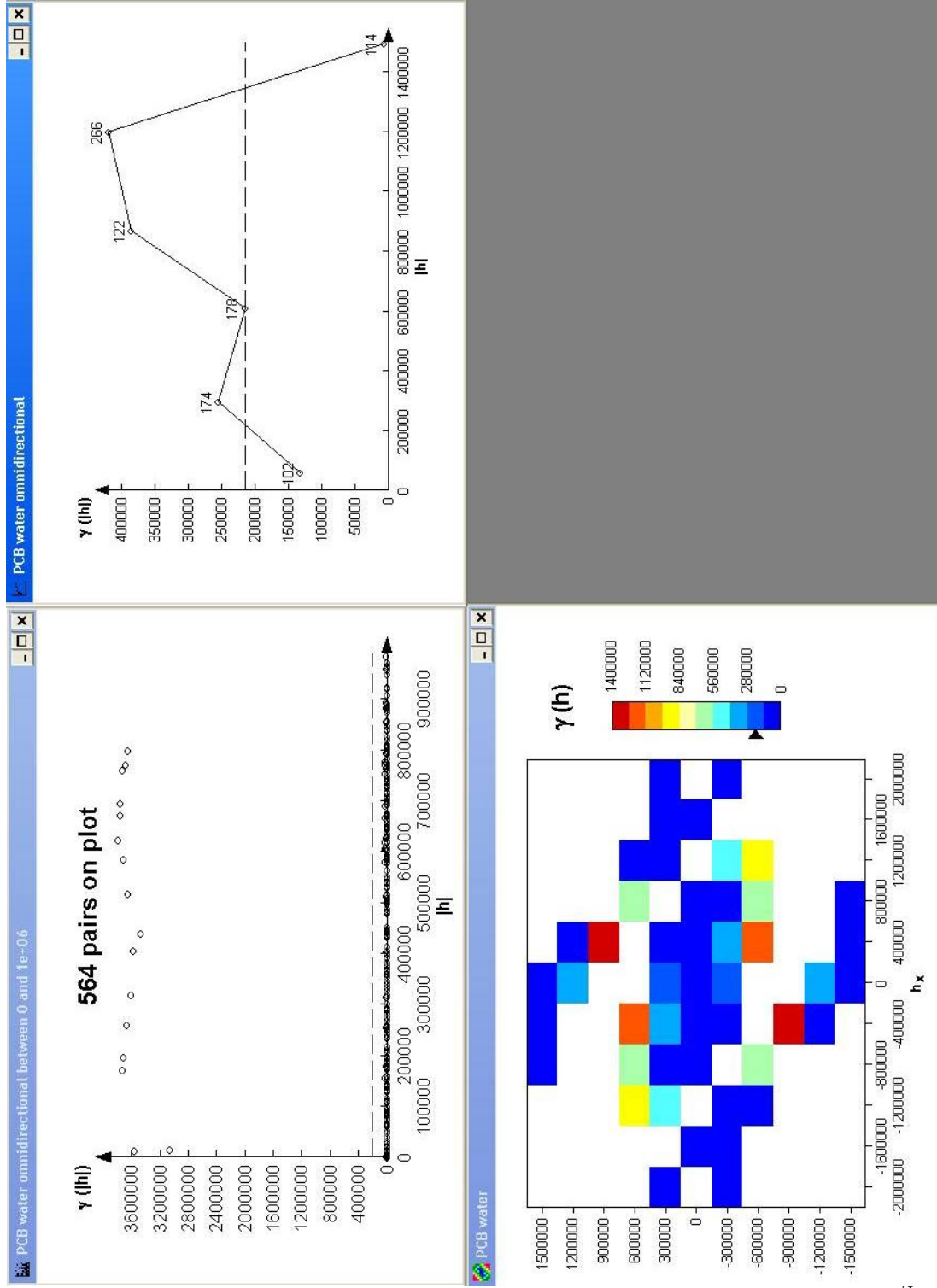


Figure 57: variogram cloud, variogram surface, and directional variograms for PCBs sediment concentrations, period 2005-2007

For water the period studied was 2005-2007:



F

For air the period studied was 2005-2007:

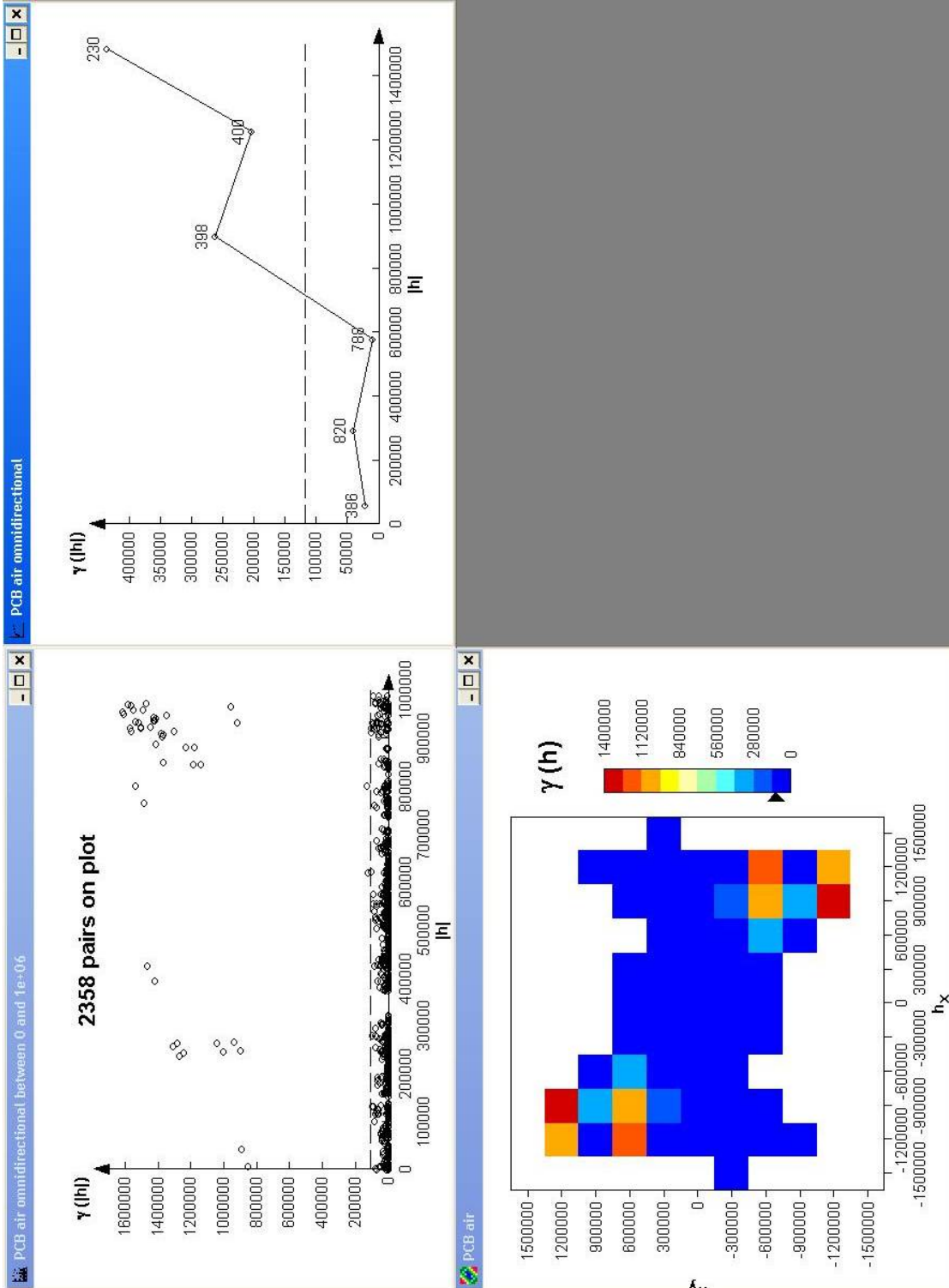


Figure 59: variogram cloud, variogram surface, and directional variograms for PCBs air concentrations, period 2005-2007

5.4 PCDD/Fs

These are the results found for air:

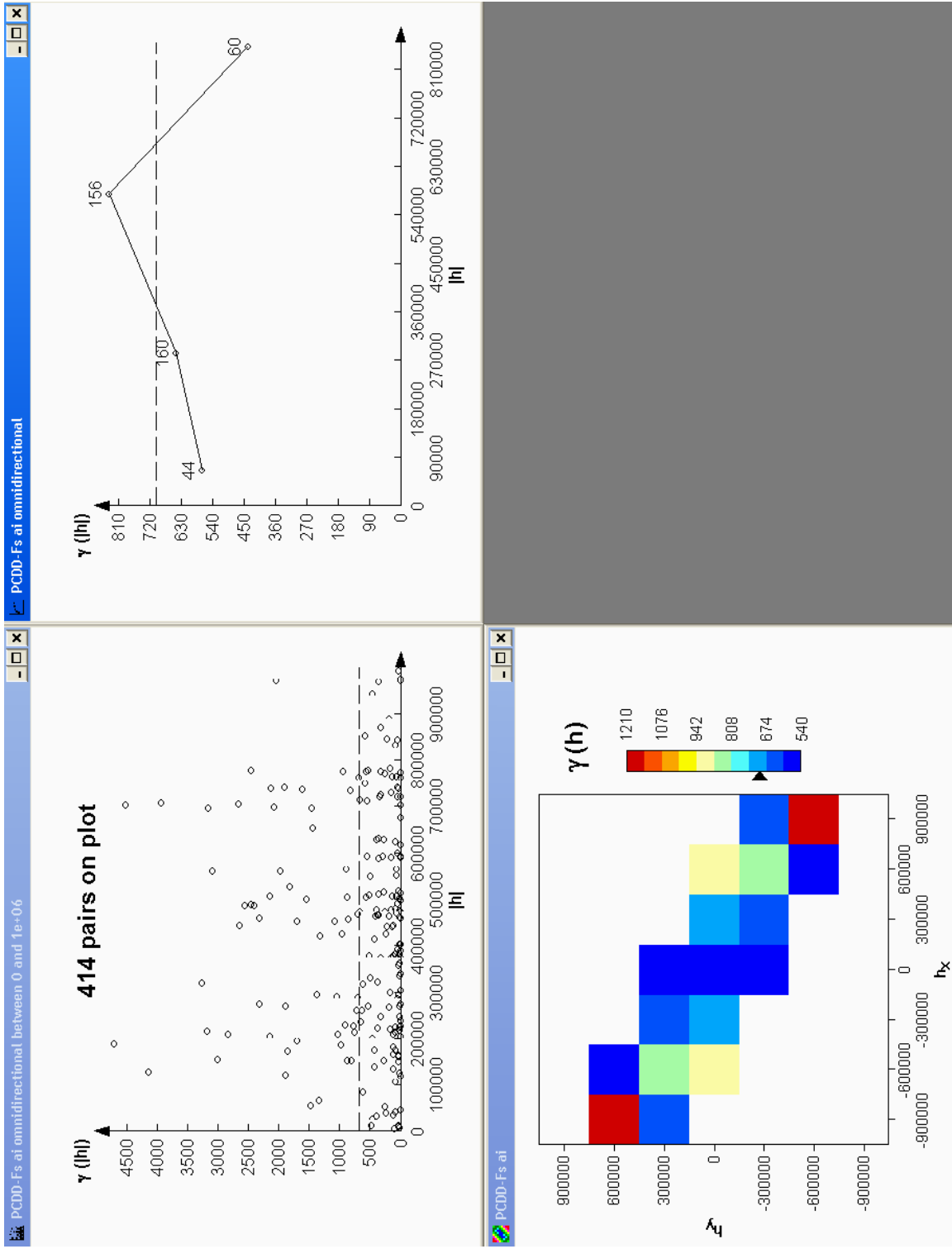


Figure 60: variogram cloud, variogram surface, and directional variograms for PCDD/Fs air concentrations, period 1994-1996

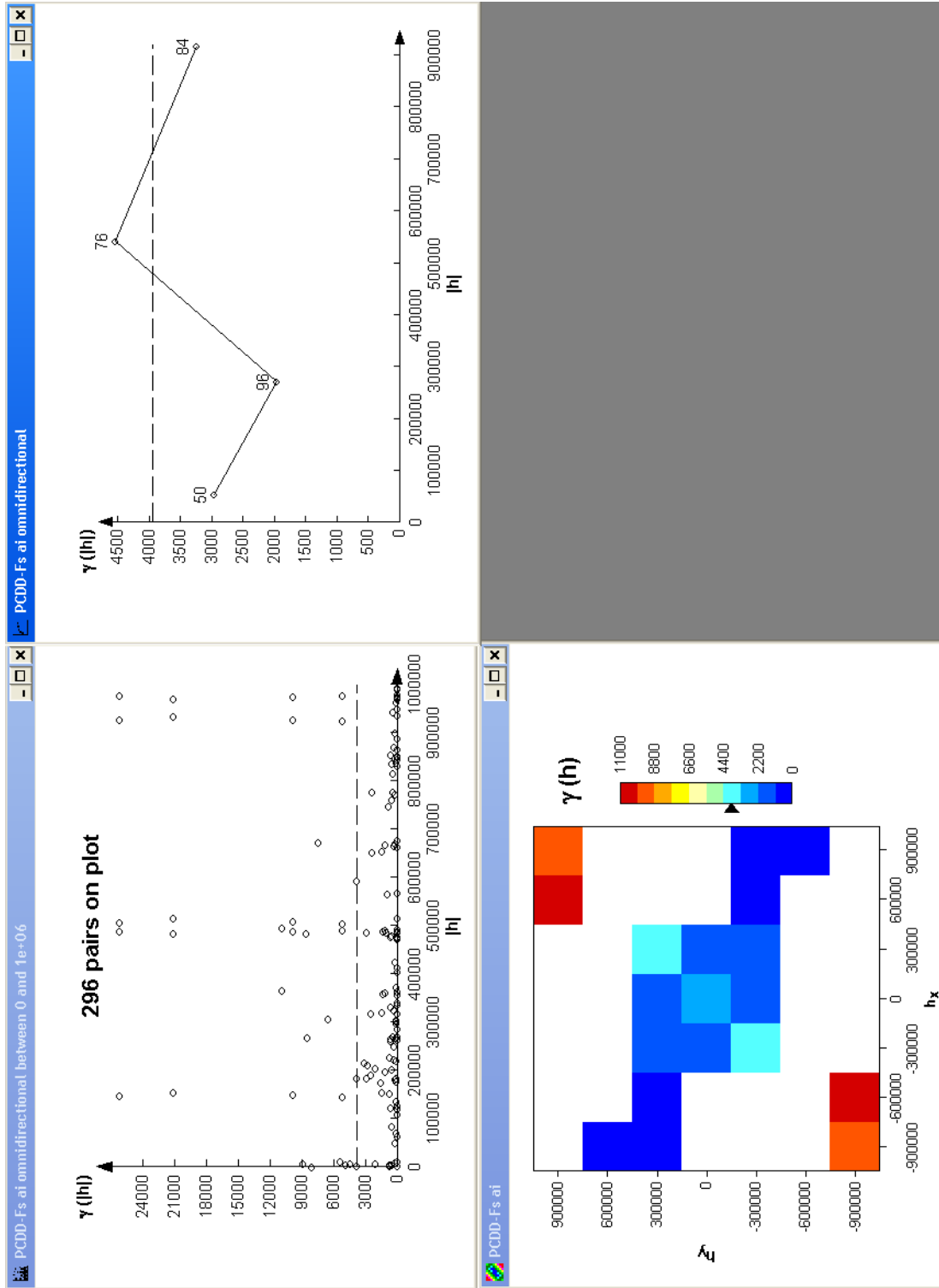


Figure 61: variogram cloud, variogram surface, and directional variograms for PCDD/Fs air concentrations, period 1996-1998

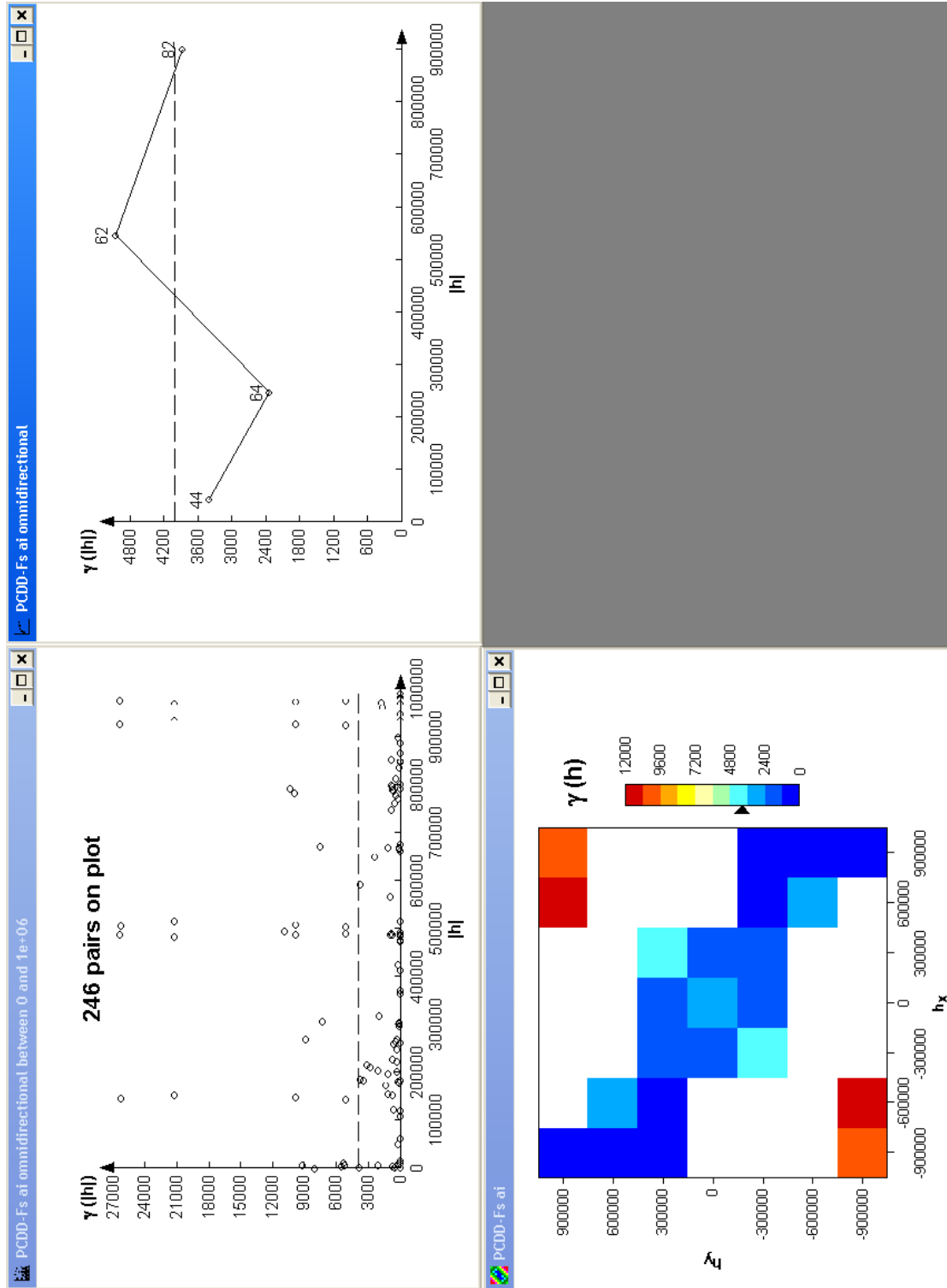


Figure 62: variogram cloud, variogram surface, and directional variograms for PCDD/Fs air concentrations, period 1997-1999

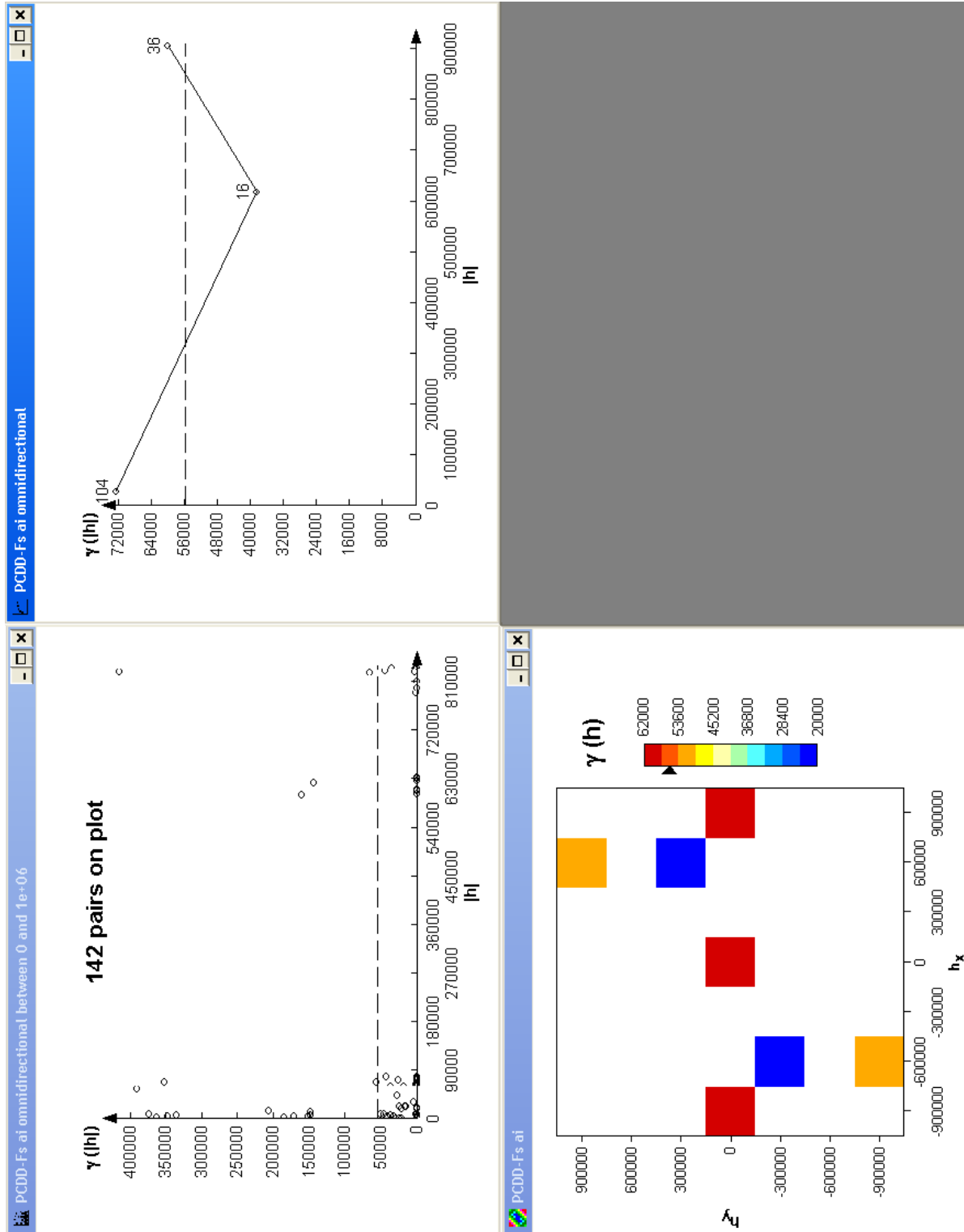


Figure 63: variogram cloud, variogram surface, and directional variograms for PCDD/Fs air concentrations, period 2002-2004

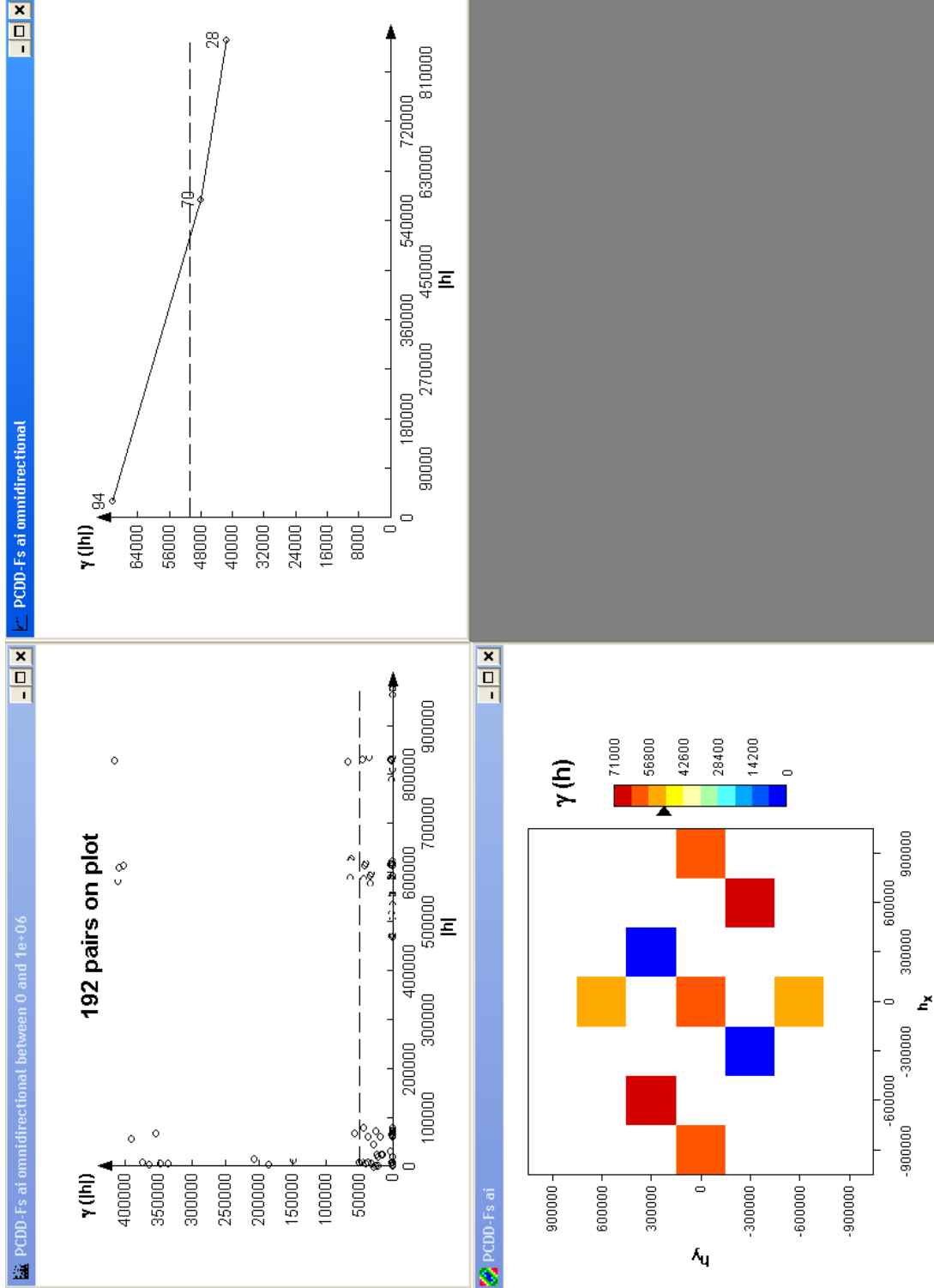


Figure 64: variogram cloud, variogram surface, and directional variograms for PCDD/Fs air concentrations, period 2004-2006

These are the results found for water:

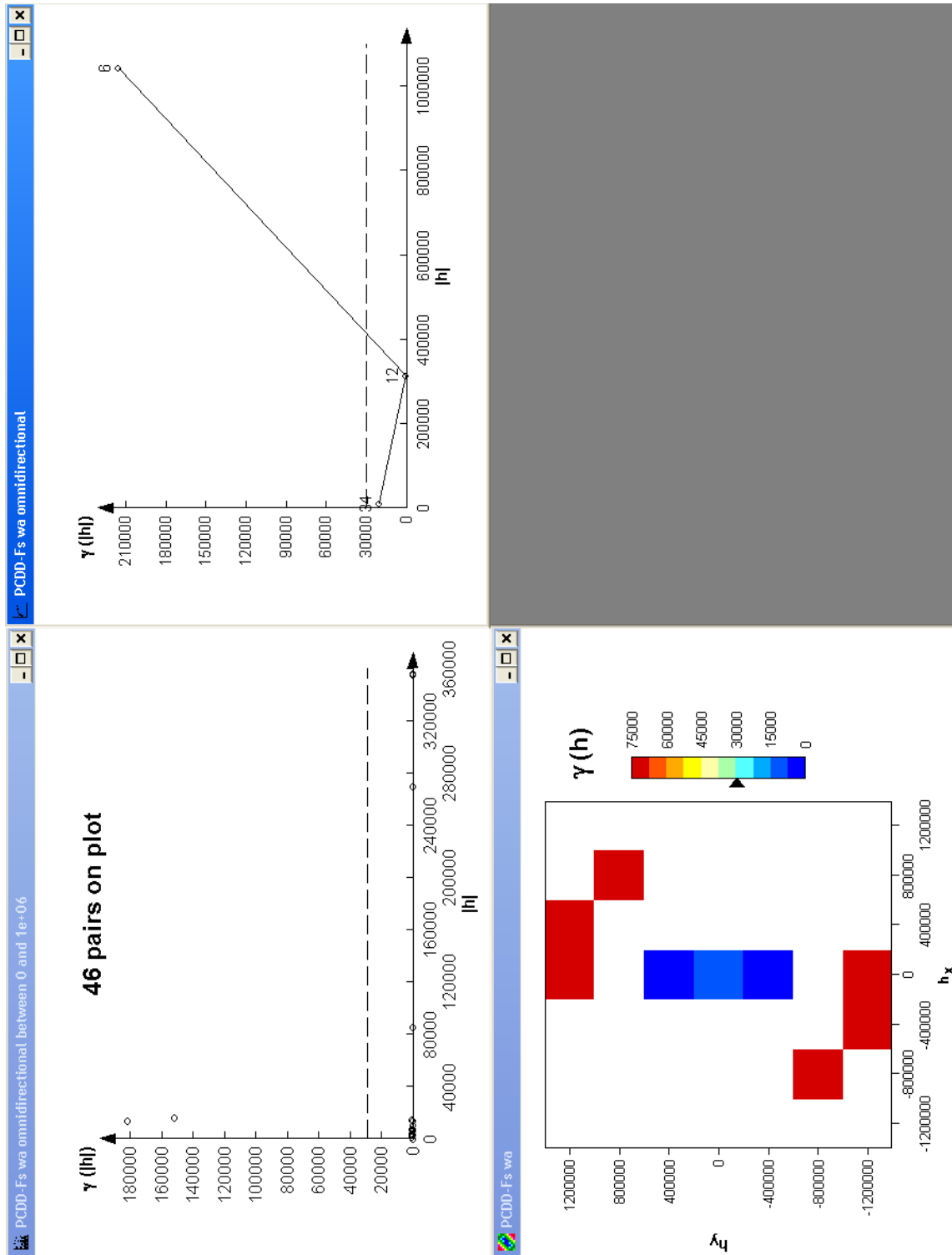


Figure 65: variogram cloud, variogram surface, and directional variograms for PCDD/Fs water concentrations, period 2005-2007

These are the results found for sediment:

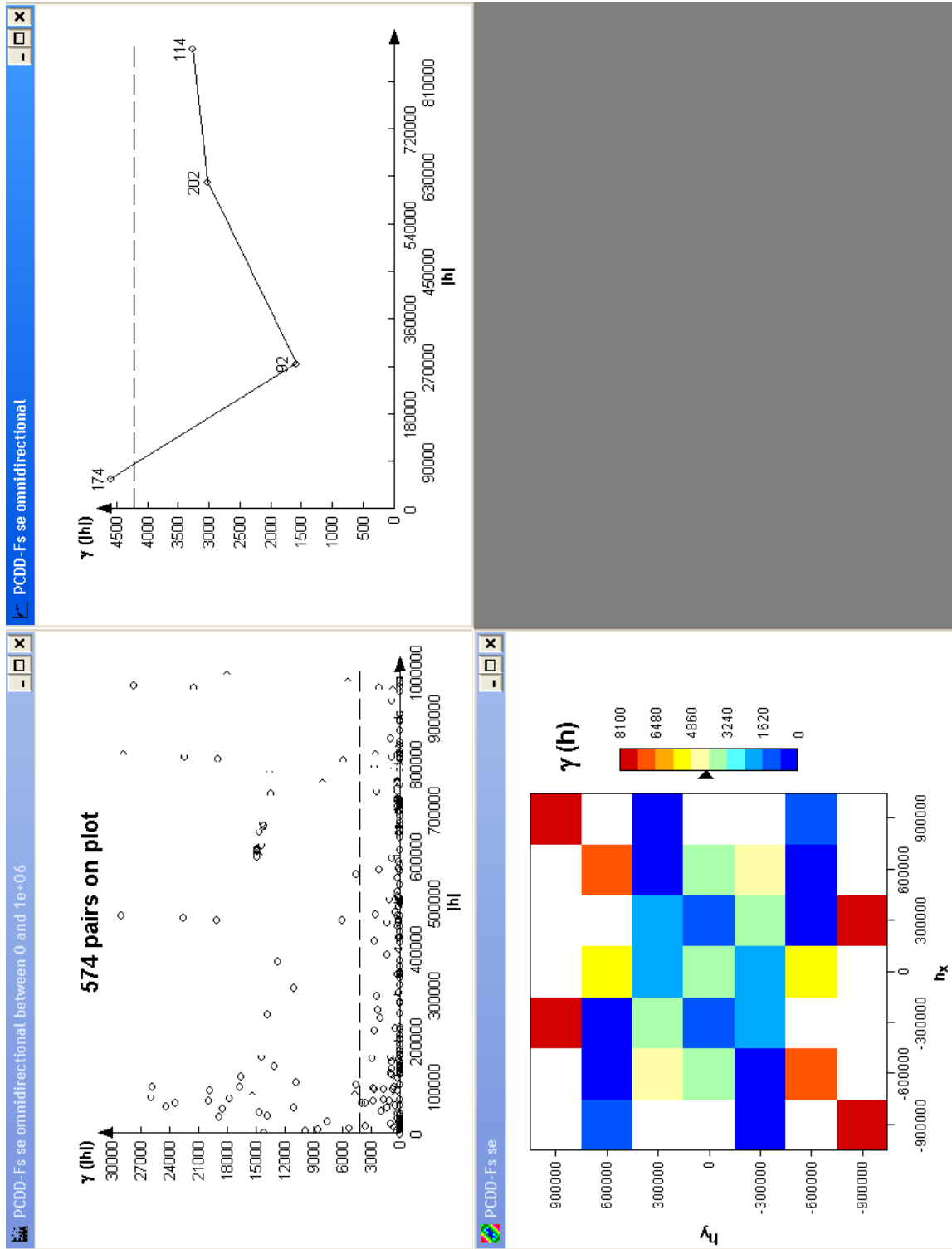


Figure 66: variogram cloud, variogram surface, and directional variograms for PCDD/Fs sediment concentrations, period 1994-1996

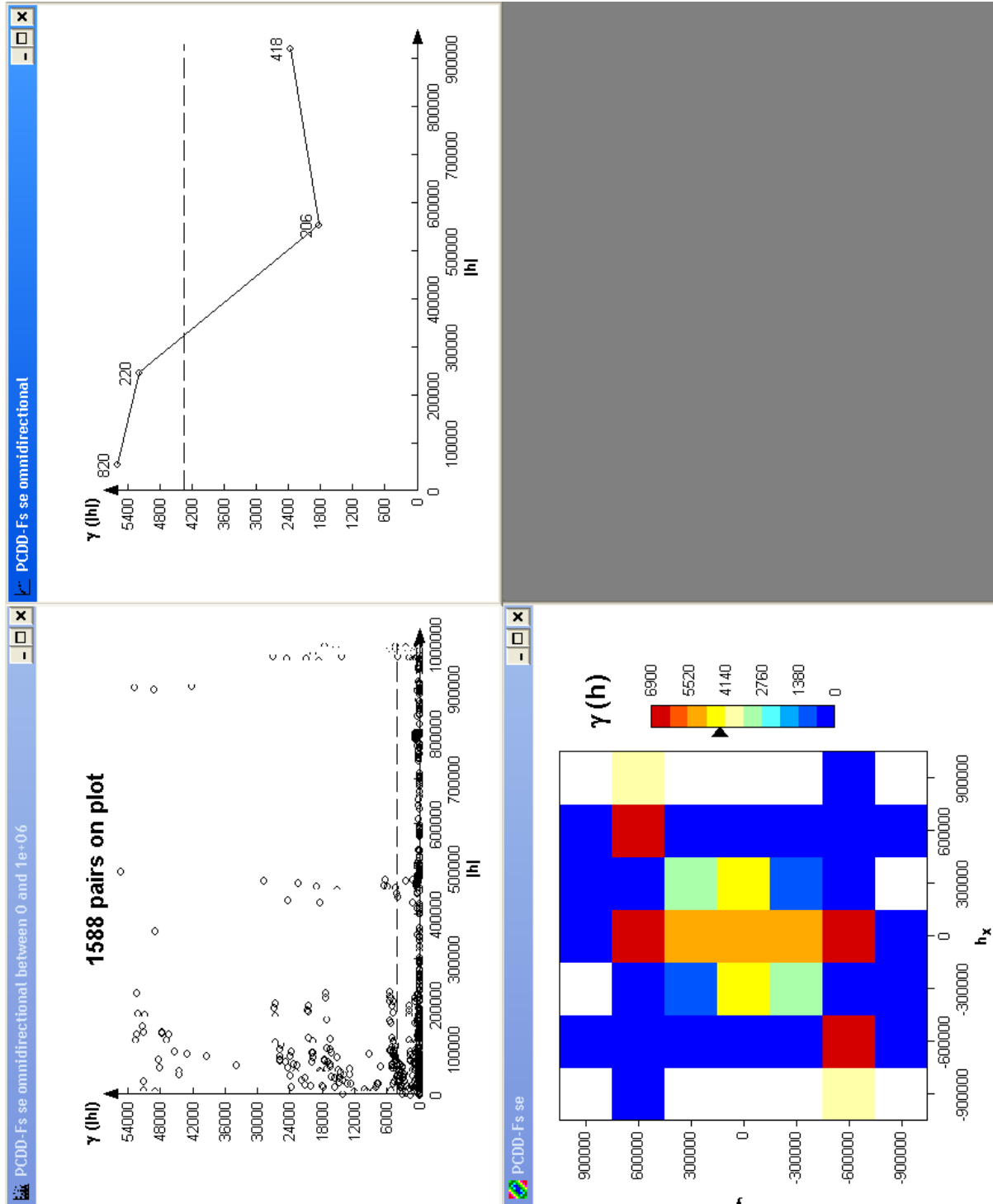


Figure 67: variogram cloud, variogram surface, and directional variograms for PCDD/Fs sediment concentrations, period 1996-1998

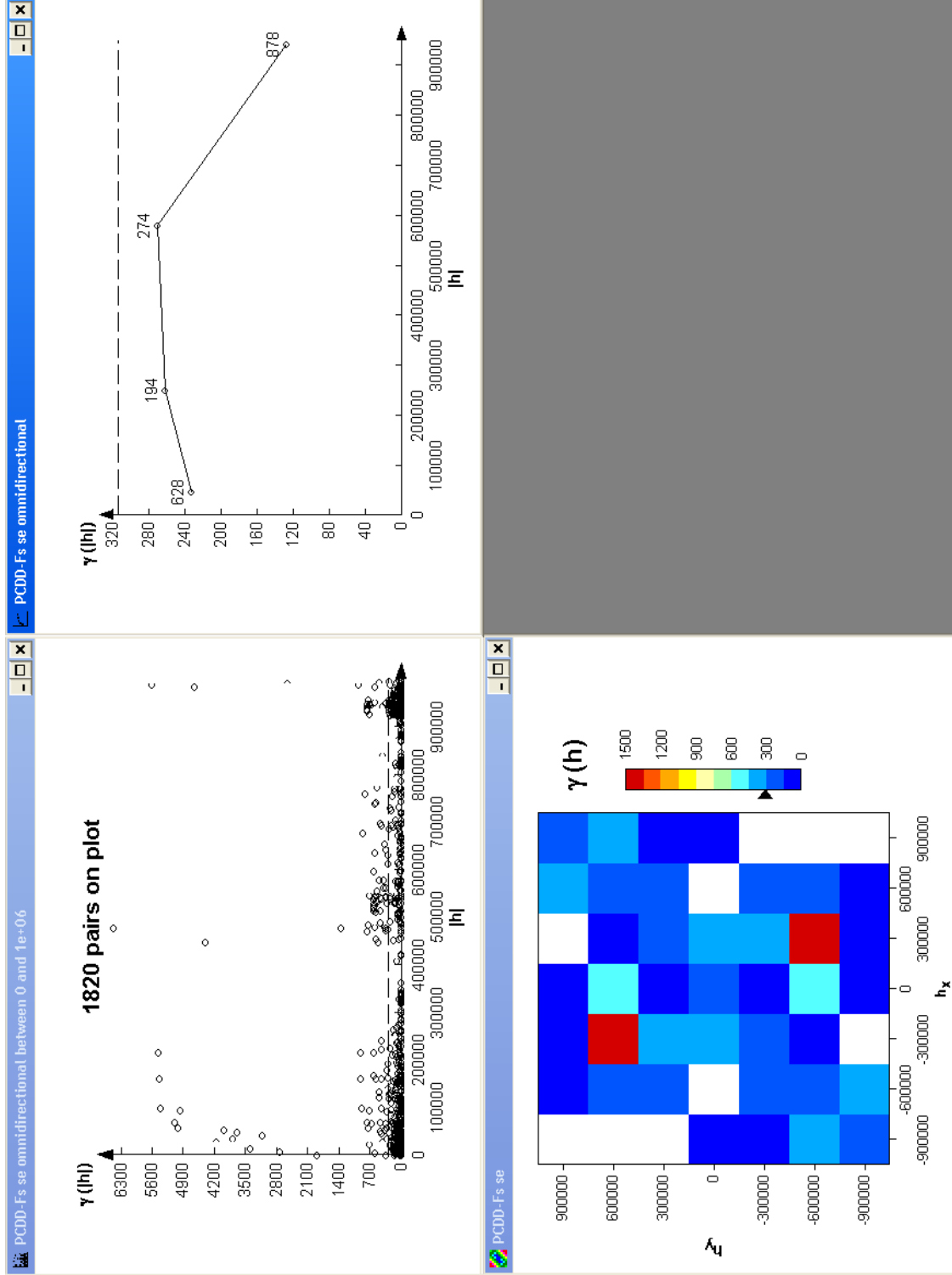


Figure 68: variogram cloud, variogram surface, and directional variograms for PCDD/Fs sediment concentrations, period 1998-2000

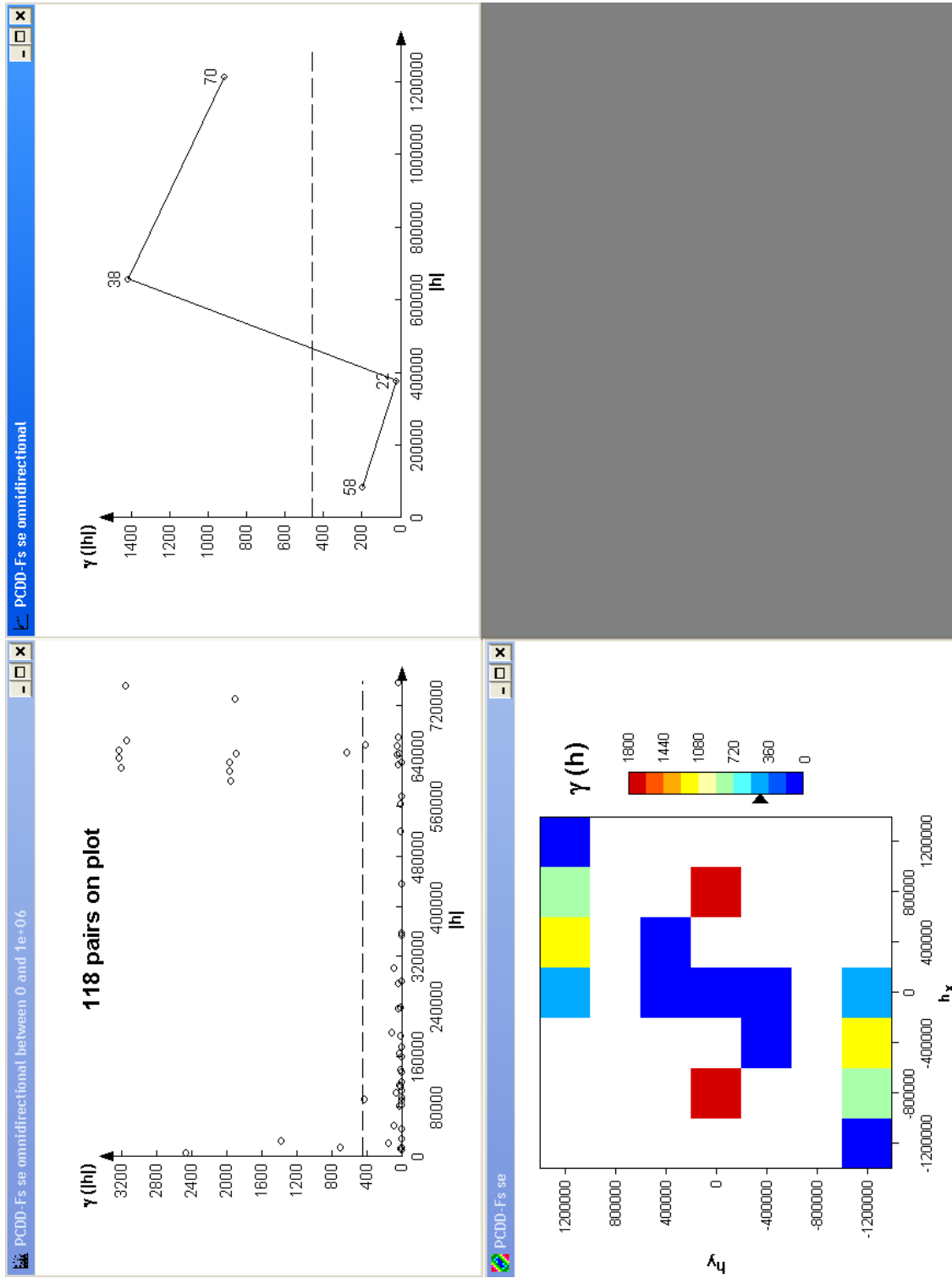


Figure 69: variogram cloud, variogram surface, and directional variograms for PCDD/Fs sediment concentrations, period 2001-2003

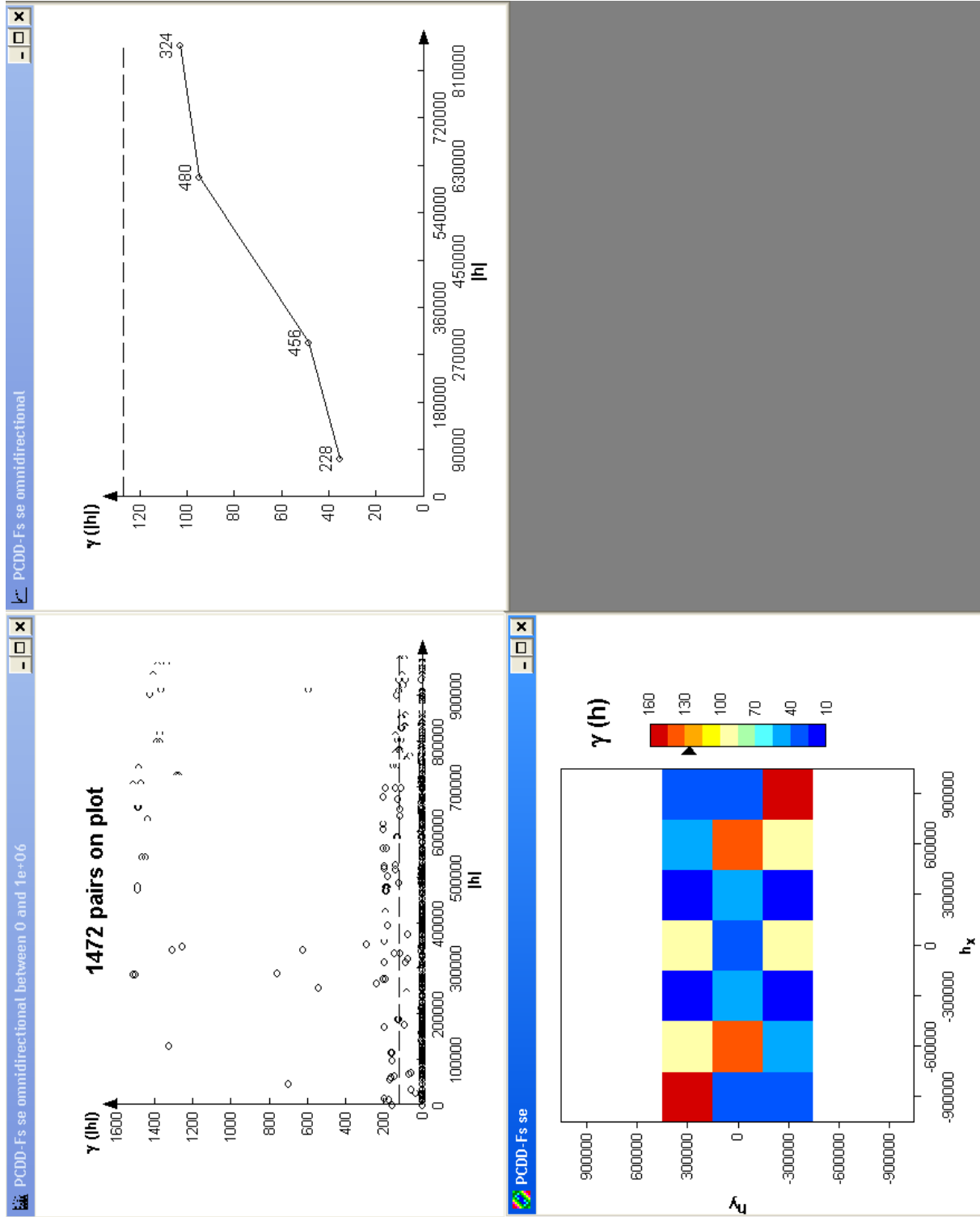


Figure 70: variogram cloud, variogram surface, and directional variograms for PCDD/Fs sediment concentrations, period 2005-2007

These are the results found for soil:

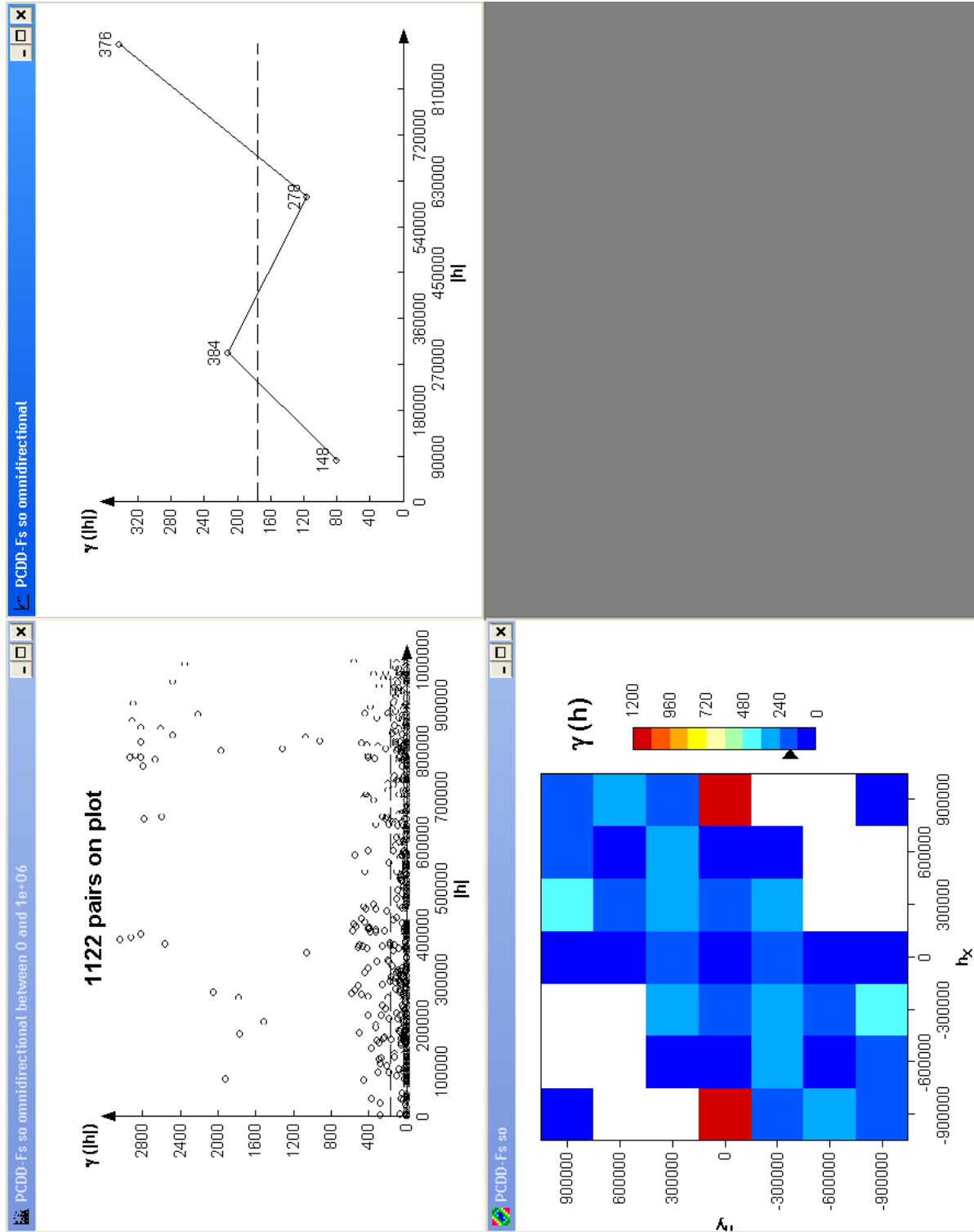


Figure 71: variogram cloud, variogram surface, and directional variograms for PCDD/Fs soil concentrations, period 1998-2000

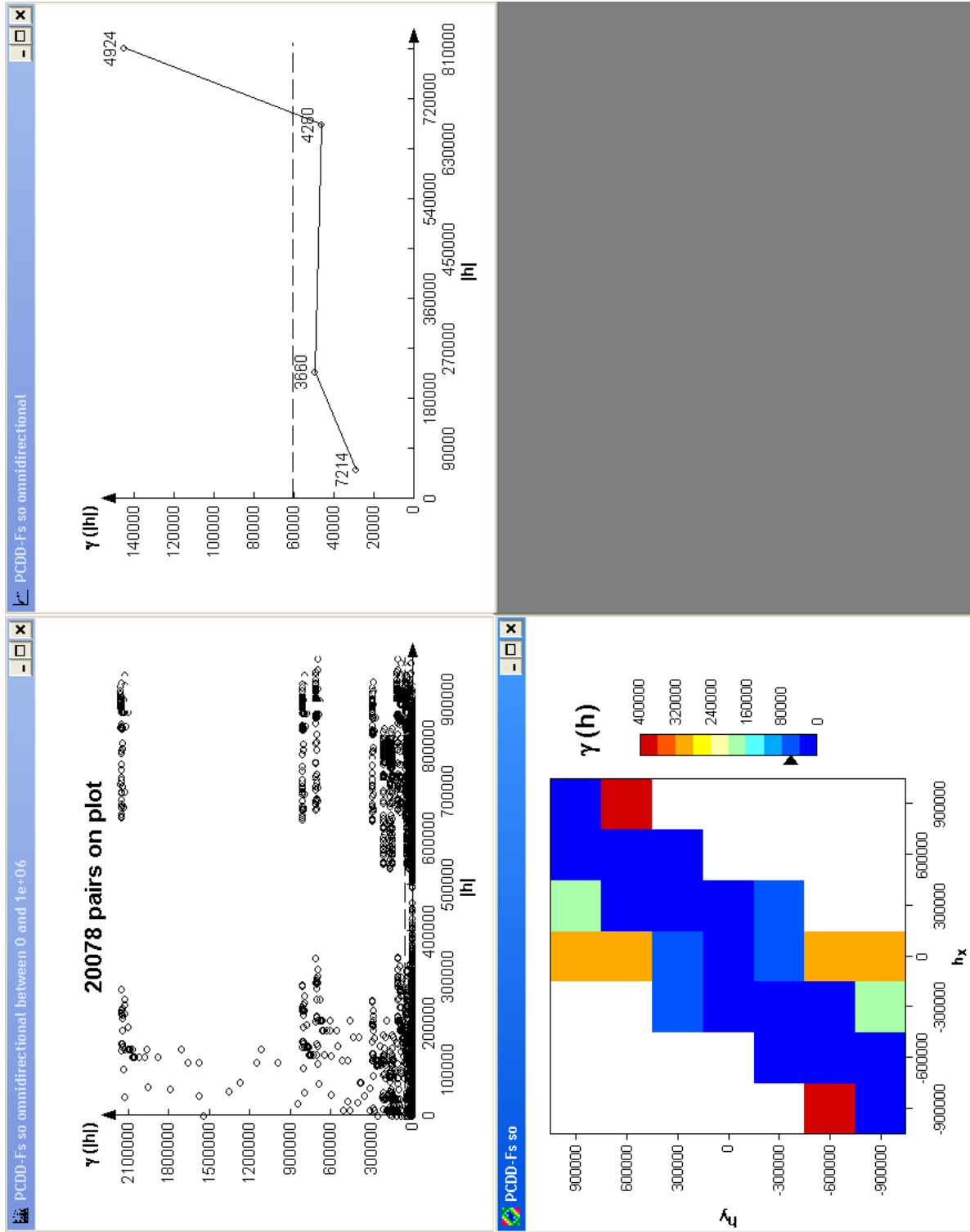


Figure 72: variogram cloud, variogram surface, and directional variograms for PCDD/Fs soil concentrations, period 2002-2004

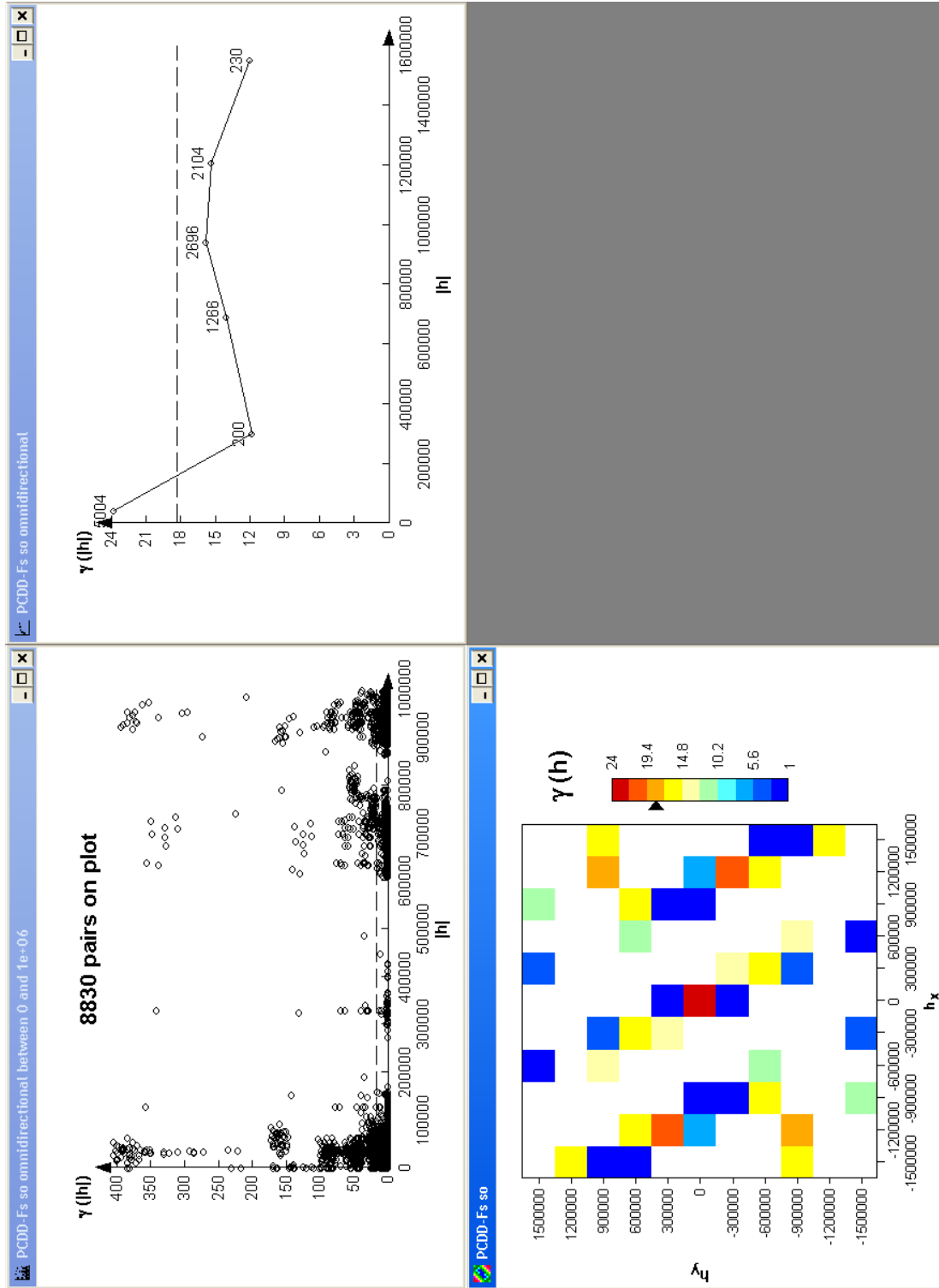


Figure 73: variogram cloud, variogram surface, and directional variograms for PCDD/Fs soil concentrations, period 2004-2006

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Title: A Compilation of Europe-Wide Databases from Published Measurements of PCBs, Dioxins and Furans

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Abstract

Chemical risk assessment always entails an evaluation of expected environmental concentrations of substances; these concentrations may be predicted using mathematical models or may be chosen on the basis of experimental observations and monitoring activities.

The work discusses the usefulness and limitations in building compilations of published monitoring data by describing a feasibility study on polychlorinated biphenyls (PCBs) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), for which observed concentration data were retrieved from scientific journal articles published between 2000 and 2009.

These chemicals are typical POPs and are of constantly high concern for their potential adverse effects on human health and ecosystems. PCBs were mainly used by the power industry in electrical transformers, capacitors, hydraulic equipment, and as lubricants. PCDD/Fs are formed as unintentional by-products of chemical manufacturing and incineration processes, as well as natural processes such as volcanic eruptions, and biomass burning. Emissions from incineration of industrial wastes such as metal reclamation and domestic heating (especially in central Europe) are considered as current sources of PCDD/Fs to the environment.

A database of published observed concentrations of PCBs and PCDD/Fs in air, soil sediments and water was built as described in details in the report. As continental scale assessment is aimed more at the evaluation of the regional distribution of contamination, we excluded from the database those measurements taken close to known PCB or PCDD/F pollution sources of exceptional entity such as waste incinerators or contaminated sites.

After presenting and interpreting the results of the literature search, we critically examine the completeness and usability of this information, and the usefulness of data compilations in the framework of chemical risk assessment.

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