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Documentation of the TMAP Parameter "Pollutants in seabird eggs" in The Netherlands in 2013

1. Egg sampling

Eggs were sampled according to OSPAR (1997), Becker et al. (2001) and VDI (2009). In general, per year, species and site, 10 eggs were sampled (cf. Fig. 1.1 and Table 1.1).



Fig. 1.1: Sampling sites of Oystercatcher and/or Common Tern eggs in the trilateral Wadden Sea. The Netherlands: 1 Balgzand, 2 Griend, 3 Julianapolder, 3-2 Schiermonnikoog, 4 Delfzijl; Germany, Lower Saxony: 5 Dollart, 5-2 Baltrum, 6 Minsener Oog, 7 Mellum (6 and 7 =Jade), 8 Hullen, 9 Neufelderkoog (8 and 9 = Elbe estuary); Germany, Schleswig Holstein: 10 Trischen, 11 Norderoog/Hallig Hooge, Denmark: 12 Langli. At sites 3, 5, 7, 8 and 12 only Oystercatcher eggs, at sites 3-2, 6 and 9 only Common Tern eggs were taken.

Table 1.1. Trumber of Oystercatenet and Common Tern eggs sampled per site in 201.

	Spe	cies
Site	Oystercatcher	Common Tern
Balgzand	10	10
Griend	10	10
Julianapolder	10	-
Schiermonnikoog	-	10
Delfzijl	10	10

2. Chemical analytics

The samples were analyzed as shortly described by OSPAR (1997), Sommer et al. (1997) and Becker et al. (2001). You can get a detailed description of methods on request.

3. Documentation of data, description of the EXCEL-file and of the variables

The data from The Netherlands in 2013 are documented in an EXCEL-file (already sent on disk). The variables in the EXCEL-file are explained in the file-head.

4. Assessment of the results

To present a short assessment of the data, we prepared and present statistics for the following most important chemicals or chemical groups:

ORGANOCHLORINES:

PCB sum	62 congeners
6PCBs	6 PCB-congeners (PCB28, PCB52, PCB101, PCB138, PCB153, PCB180)
НСВ	
HCH sum	α-HCH, β-HCH, γ-HCH
DDT sum	p,p'-DDE, p,p'-DDT, p,p'-DDD
Chlordane sum	Sum of Chlordan and Nonachlor-compounds: trans-Chlordan, cis-Chlordan, trans-Nonachlor, cis-Nonachlor

To make the following text more readable, for "PCB sum" the simplifying term "PCB", for "DDT sum" the term "DDT", for "HCH sum" the term "HCH" and for "Chlordane sum" the term "Chlordanes" is used.

HEAVY METALS:

Hg (mercury)

All concentrations are given in ng/g fresh weight of the eggs with an accuracy of one digit after the decimal point (which is shown as a comma).

Statistics:

For analyzing temporal trends for the years 2009 - 2013, Spearman rank correlations were calculated (two-tailed) for the years 2009 - 2013. To identify potential differences in pollutant concentrations between 2012 and 2013, Mann-Whitney-U-tests were conducted. In the figures, arithmetic means \pm 95% confidence intervals are presented. If the confidence intervals do not overlap, significance of at least p < 0.05 is indicated.

5 **Results**

5.1 Spatial patterns of selected contaminants in the Wadden Sea in 2013

For the Oystercatcher, we identified the following areas with relatively high contamination. They are listed in order of mean contamination level, beginning with the highest (see Abb. 5.1.1; Tab. 8.1.1):

Mercury:	Griend, Elbe, Trischen
PCB:	Dollart, Trischen, Julianapolder
HCB:	Delfzijl, Elbe, Trischen
DDT:	Dollart, Trischen, Elbe
HCH:	Halligen, Trischen, Balgzand
Chlordanes:	Balgzand, Griend, Dollart

In the Common Tern, concentrations of Hg, PCB, HCB and DDT tended to be higher at most sites than in the Ostercatcher, whereas HCH and Chlordane concentrations tended to be lower. In the following areas relatively high contaminations of Common Tern eggs were detected (given in the order of contamination level, beginning with the highest average level, see Abb. 5.1.1; Tab. 8.1.1):

Elbe, Trischen, Halligen
Elbe, Trischen, Delfzijl
Elbe, Halligen, Baltrum
Elbe, Trischen, Delfzijl
Elbe, Balgzand, Trischen
Balgzand, Griend, Delfzijl

In the **Oystercatcher**, the geographical pattern of **Hg** concentrations in 2013 was largely similar to that of 2012 (last report): Again **Hg** peaked at Trischen and more or less surrounding sites. A further peak was detected at Griend, which reached this year highest values. We see also the same trend as the year before: Because of the rising concentrations in formerly lower contaminated areas the differences between the areas seemed to be lower. This development may result in an established level of 200 ng/g **Hg** in Oystercatcher eggs in all areas of the Wadden Sea.

For **PCB** a prominent concentration peak was 2013 recognized at the Dollart. Lower peaks in comparable levels were found at Trischen, Julianapolder, Elbe, Jade and Delfzijl. But on the other hand we saw 2013 the same trend as 2012 that the PCB concentrations equalise at the different locations. This was mainly caused by increased concentrations in the western Wadden Sea.

As in the years before the clearly highest concentration of **HCB** was measured in 2013 at Delfzijl where it was a little lower than in 2012. A second, much smaller peak was again recorded at the river Elbe. For **DDT**, the highest concentrations in 2013 were clearly found at the Dollart but also concentration in Oystercatcher eggs from Trischen reached high levels.

For **HCH** we see the first time a strong concentration peak in eggs from the Halligen, which was mainly caused by high β -HCH levels in three eggs. Beside a second peak at Trischen, high HCH levels were found at Balgzand and Griend.

For **Chlordanes**, peak values have been recorded again at the western parts of the Wadden Sea, mainly in Oystercatcher eggs from Balgzand and Griend, as well as in eggs from Delfzijl and Dollart.

Except the strong peak of **HCB** at Delfzijl and of **HCH** at Halligen, we found for all pollutants only small differences between the different sites. The increased concentrations in both the western and the eastern Wadden Sea have resulted in a stronger approximation of the values at a generally increased contamination level.

Levels of environmental chemicals in Oystercatcher eggs from Langli in Denmark were low in 2013 in case of PCB, HCB, DDT and HCH compared to the other sites, whereas Hg and Chlordane reached median levels.



Fig. 5.1.1: Spatial patterns of pollutant concentrations in Oystercatcher and Common Tern eggs from the Wadden Sea in 2013 (means with standard deviations)

In the **Common Tern**, contamination patterns of **Hg**, **PCB**, **HCB**, **DDT** and **HCH** identified in 2013 were in general rather similar to those in 2012 or the years before, showing peak values at Elbe and at Trischen as well as at Delfzijl and in case of **Hg** and **HCB** at Halligen. In **HCH** a prominent concentration peak was again recorded at the river Elbe and a second lower one at Trischen and Balgzand. Concentrations of **Chlordanes** tended to be higher in the western parts of the Wadden Sea. However concentrations measured in 2013 were lower than in 2012 at many sites.

5.2 Annual variation in pollutant concentration in the period 2009-2013

Mean contamination values for the different substance groups in the period 2009-2013 are given in Figures 8.2.1-8.2.6 in the Appendix. A summarizing overview of short-term (2012-2013) and midterm (2009-2013) temporal changes in the Dutch and German Wadden Sea are given in Tables 5.2.1 and 5.2.2.

Balgzand

Temporal Trends 2009-2013:

Oystercatcher Significant **increases** were detected in HCB and Chlordanes, a significant **decrease** occurred in Hg.

Common Tern Significant increases were detected in PCB, HCB, DDT, HCH and Chlordanes.

In 2013 a significantly **higher** level of HCB and Chlordanes than in 2012 were found in Common Tern eggs. In Oystercatcher eggs concentration of PCB and DDT was **lower** in 2013 compared to 2012.

Griend

Temporal Trends 2009-2013:

Oystercatcher Significant **increases** were detected in Hg, HCB, and Chlordanes. Significant **increases** were detected in HCB and Chlordanes.

Between 2012 and 2013, PCB and Chlordane concentrations **decreased** in Oystercatcher eggs. In Common Tern eggs, the levels of HCB and Chlordanes **increased** in that period whereas levels of Hg and HCH **decreased**.

Julianapolder/Schiermonnikoog

Temporal Trends 2009-2013:

- Oystercatcher Significant **increases** in the concentrations of HCB and Chlordanes were identified at Julianapolder.
- Common Tern Significant **increases** in the concentrations of PCB, HCB and Chlordanes were identified at Schiermonnikoog.

In Oystercatcher eggs from 2013 sampled at Julianapolder, the level of HCH was significantly **higher** than in 2012. In Common Tern eggs from 2013 sampled at Schiermonnikoog, the concentrations of Hg and HCH were significantly **lower** than in 2012.

<u>Delfzijl</u>

Temporal Trends 2009-2013:

Oystercatcher	The concentrations of Hg decreased significantly. HCB concentrations did not
	change significantly.
Common Tern	Significant increases in the concentrations of Hg, HCB, DDT and Chlordanes.

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Between 2012 and 2013 PCB and DDT **decreased** in Oystercatcher eggs. Number of Common Tern eggs sampled in 2012 was below five, so no statistical comparison of the two years was possible.

Table 5.2.1: Overview over the development of selected pollutants in eggs of Oystercatcher and Common Tern in the Wadden Sea from 2009-2013, according to Spearman rank correlations. -: significant decline, +: significant increase.

	Oystercatcher							Common Tern										
	Balgzand	Griend	Julianapolder	Delfzijl	Dollart	Jade	Hullen	Trischen	Halligen	Balgzand	Griend	Schiermonnikoog	Delfzijl	Baltrum	Jade	Neufelderkoog	Trischen	Halligen
Hg	-	+		-	-				-				+					
PCB Summe					+			+	+	+		+				+	+	+
HCB	+	+	+			-	+	+	+	+	+	+	+	+	+		+	+
DDT Summe					+				+	+			+	+				+
HCH Summe						-			+	+					+			
Chlordan-Nonachlor-Summe	+	+	+		+	+	+	+	+	+	+	+	+	+	+		+	+

Table 5.2.2: Overview over the development of selected pollutants in eggs of Oystercatcher and Common Tern in the Wadden Sea from 2012-2013, according to Mann-Whitney-U-tests. -: significant decline, +: significant increase. Common Tern sample size from Delfzijl is too low for statistical comparison between 2012 and 2013.

		Oystercatcher							Common Tern									
	Balgzand	Griend	Julianapolder	Delfzijl	Dollart	Jade	Hullen	Trischen	Halligen	Balgzand	Griend	Schiermonnikoog	Delfzijl	Baltrum	Jade	Neufelderkoog	Trischen	Halligen
Hg								-			-	-	?	-	-	-		
PCB Summe	-	-		-			-						?		-		-	
HCB					-				+	+	+		?	+		-		+
DDT Summe	-			-									?		-	-	-	-
HCH Summe			-					+	+		-	-	?	-	-	-	-	
Chlordan-Nonachlor-Summe		-			-					+	+		?				-	

6. General Assessment

6.1 Spatial Trends

The concentrations of the mentioned substances in **Oystercatcher** eggs showed in 2013 only small geographical differences. Generally the measured concentrations were slightly lower compared to 2012, but still at a high level. HCB concentration at Delfzijl was five times higher compared to other sites in the Wadden Sea. In Oystercatcher eggs highest levels of PCB and DDT were detected at the Dollart, whereas HCH reached highest levels at Halligen.

In the **Common Tern**, clearly prominent concentration peaks of all contaminants except Chlordanes were found again at the Elbe estuary. Consequently, a continuous decrease of pollution was recorded with increasing distance from the Elbe estuary which is in accordance to the observations from previous years. So we found the second highest levels mostly at Trischen, as the area is influenced most by the waters of the river Elbe. But in case of HCB we saw a lesser influence of the river Elbe compared to the other contaminants: The concentration showed only small geographical differences and tended to align in Common tern eggs at a level of ca. 15 ng/g fresh weight.

In general, the species-specific spatial contamination patterns in 2013 remained similar to those recorded in 2012 and the years before: The contamination pattern of the Common Tern again showed clear concentration peaks at the river Elbe. And that of the Oystercatcher again indicated a clear HCB peak at Delfzijl and for the first time a strong concentration peak of HCH at Halligen. The species-specific patterns may have been influenced by differences in the feeding ecology, with the Oystercatcher possibly foraging to a higher degree in terrestrial habitats at some sites. This might explain why in particular at the river Elbe, no maximum concentrations have been found in the Oystercatcher but in the Common Tern.

This may also explain the high HCH levels in eggs from the Halligen, which was mainly caused by high β -HCH levels in three eggs. β -HCH implies a contamination with technical HCH which was used as an insecticide before the implementation of Lindan (concentrated γ -HCH). Apparently there are still remainders of these old insecticides.

Furthermore, for the benthic feeding Oystercatcher, contaminants bound to sediment particles which might have been transported previously, are probably more important than for the pelagic feeding Common Tern. In addition, the partly whole-year presence of the Oystercatcher in the Wadden Sea might have affected the contamination patterns recorded. In general, the data underline the importance of the large rivers Elbe and Ems for the influx of several contaminants into the Wadden Sea. For Hg, PCB, HCB and DDT, contamination levels tended to be higher in the Common Tern than those in the Oystercatcher (except for HCB at Delfzijl), reflecting the higher trophic level of the piscivorous Common Tern with an even more effective bioaccumulation of pollutants. However, for Chlordanes contamination was clearly higher in Oystercatcher than in Common Tern at most study sites.

The mentioned concentration peak of HCB in Oystercatcher eggs from Delfzijl could be seen since many years and may be caused by contaminated water and sediment in the Sea Harbour Channel in Delfzijl (Eggens & Bakker 2001). Therefore the lower concentrations in 2013 compared to 2012 were pleasant, but in the period 2008-2012 (see TMAP reports 2011 and 2012) increasing concentrations were evident after the HCB concentration had decreased until 2000 (Eggens & Bakker 2001).

6.2 Temporal trends

During the **five-year-period 2009-2013** contamination level of many measured substances increased mostly at all study sites. A decrease was only detected in case of Hg concentrations in **Oystercatcher** eggs form four sites (Balgzand, Delfzijl, Dollart, Halligen) and in case of HCB or HCH from the Jade. In the Oystercatcher eggs Chlordane increased during the five-year-period at eight of nine sampled sites. HCB increased at six as well as PCB at three and DDT at two sites. Hg and HCH increased at

one site respectively. Remarkable was the situation on the Halligen where all measured substances except Hg increased. In total we found 21 cases of significantly increased substance levels (factor 1,5 up to 6): The PCB concentration at Dollart increased by a factor of three as well as DDT by factor of 1,5 (see figures at 8.2). The levels of Chlordane increased up to a factor of 6 in the western parts of the Wadden Sea (Balgzand, Julianapolder, Griend).

The mentioned decreases of Hg and HCB levels in Oystercatcher eggs were at all sites beneath a factor of two but continuously and with a very low variance in all years.

In the **Common Tern** eggs no significant decrease in the period during 2009-2013 could be detected.

We see an increase of Chlordanes and HCB at eight of nine study sites. PCB increased at five, DDT at four and HCH at two of the nine study sites. Hg increased at one site respectively. In total we found 28 significant increased substance levels in Common tern eggs and to a greater extent as in Oystercatcher eggs. The HCB concentrations were remarkable, increased by a factor of five at Halligen as well as by a factor of two or three at Balgzand, Schiermonnikoog, Delfzijl, Baltrum, Jade and Trischen, whereas the HCB concentration at Griend rose linearly by a factor of four. The concentrations of Chlordanes increased up to a factor of five in the Dutch parts of the study area (e.g. Griend) whereas the PCB concentrations in Common Tern eggs doubled at Elbe and Trischen or the PCB and DDT concentrations at Balgzand rose up to the threefold.

When **comparing the year 2013 with 2012** clearly more contamination decreases (11) than increases (3) were recorded in **Oystercatcher** eggs (Tab. 5.2.2.). But there was no visible pattern in the data. De- and increases occurred at different places of the Wadden Sea and in all chemical groups. Two points were remarkable: First at Halligen, normally a less affected area, only significant increases were detectable; and second in the Oystercatcher eggs from the river Elbe we see a decrease of the PCB-total but an increase of the most toxic, dioxin-like ones.

In contrast to 2012 in the **Common Tern** eggs more contamination decreases (19) than increases (6) were recorded in 2013 similar to the analysed Oystercatcher eggs (Tab. 5.2.2.): HCH decreased at six, DDT at five and Hg at four of the eight study sites with a sufficient sample size for a statistical comparison. Most decreases were recorded at the Elbe estuary and at Trischen (each four substance groups) as well as at the Jade (three substance groups). Increases occurred mainly in the HCB concentrations in Common Tern eggs and in the Chlordane concentrations in eggs from the western parts of the Wadden Sea.

The results indicate that both short-term and longer-term analyses are necessary to adequately describe changes in chemical pollution of the Wadden Sea.

A comparison of 2013 with 2012 showed a decrease in the contamination levels in the bird eggs between these two years. These findings were clearer in Common Tern eggs as in Oystercatcher ones. But focussing on the five-year-period between 2009 and 2013 the data showed a different picture: In total we found 49 significant increases in the contamination levels and only six decreases. So, chemical contamination increased on mid-term level at many sites of the Wadden Sea and in most analysed chemical groups. This fact was much clearer in areas which were formerly less affected like Balgzand or at the Halligen or by classes of chemical substances which were formerly lower concentrated such as HCB or Chlordanes.

Even with the data from 2013 the question if the contamination level of the environmental chemicals rose again or showed fluctuations around established levels in birds (cf. Becker & Dittmann 2009) could not finally be attempted. On the long term scale there were conspicuous decreases in the concentrations of all environmental chemicals mainly in the middle of the 1990 (Becker et al. 2001). On the other hand on the mid-term scale there were again increasing concentrations but on a lower level than before the 1990. Here we have to ask for the human activities or natural processes that are responsible for the increasing levels of environmental chemicals which are forbidden by law since many years. The data of this report indicate that increases occur mainly at the previously lower contaminated sites. The influx of the great rivers seems not to be responsible for these higher levels, as the data show generally increasing levels throughout the Wadden Sea e.g. in case of HCB.

6.3 Summarized Assessment

Summarizing, the results from 2013 indicate similar species-specific spatial contamination patterns as in the previous years. Whereas the Elbe is again the most prominent site of contamination in the Common Tern, in the Oystercatcher particularly high concentrations of HCB have been found at Delfzijl. In the Oystercatcher, the spatial contamination peaks were lesser than in the previous years. This was mainly caused by contamination increases in formerly comparatively low polluted areas. The mid-term comparison show contamination increases in both species and at nearly all sites.

Hg in Oystercatcher eggs seems to have an established level of 200 ng/g in all areas of the Wadden Sea.

When comparing the year 2013 with the previous one a clear decrease in the contamination levels could be detected, except in the HCB levels. In Common Tern eggs there was a greater decrease than in Oystercatcher ones. Remarkable were the common increase of the HCB levels in the eggs of both species. Highest HCB levels were found, like many years before, at Delfzijl, but fortunately at a little lower level as in 2012.

The recorded increases of many contaminants were clearly higher than annual fluctuations around established levels. Furthermore we see an undesired reversal of the former decreasing trends and might expect higher levels in the future. This is also of interest on the background of the Ecological Quality Objectives (EcoQOs) defined by OSPAR in recent years for coastal bird eggs from the North Sea area (Dittmann et al. 2012).

7. Literature

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This report was compiled by Frank R. Mattig, Ursula Pijanowska and Peter H. Becker.

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(Prof. Dr. Peter H. Becker)

8. Appendix

8.1 Concentrations of selected contaminants in eggs of Oystercatcher and Common Tern at different sites of the Dutch Wadden Sea 2013

Table 8.1.1: Concentrations of chemicals in Oystercatcher and Common Tern eggs sampled in The Netherlands in 2013. Mean concentrations in ng/g (fresh mass of egg content), standard deviations and number of samples are presented.

	Balgzand	Griend	Julianapolder	Delfzijl
Oystercatcher	(N=10)	(N=10)	(N=10)	(N=10)
Hg	192,4 ± 71,9	228,2 ± 67,0	154,7 ± 44,0	93,1 ± 19,2
6 PCB (law)	204,0 ± 66,6	140,6 ± 52,7	327,8 ± 108,3	258,1 ± 63,6
PCB sum	430,3 ± 132,3	295,8 ± 105,1	681,4 ± 210,6	517,4 ± 123,0
НСВ	$3,3 \pm 1,5$	$3,3 \pm 1,1$	$2,8 \pm 1,1$	$28,3 \pm 26,7$
ppDDE	16,5 ± 4,2	12,1 ± 4,0	$30,5 \pm 7,2$	35,1 ± 11,1
ppDDT	$2,3 \pm 1,4$	$2,3 \pm 1,6$	3,0 ± 1,7	0,9 ± 1,0
ppDDD	2,4 ± 0,3	$2,3 \pm 0,2$	2,2 ± 0,1	$2,5 \pm 0,3$
DDT sum	21,3 ± 4,3	16,7 ± 3,8	35,7 ± 8,2	38,5 ± 12,1
alpha-HCH	$0,6 \pm 0,8$	0,3 <u>±</u> 0,4	$0,0$ \pm $0,0$	$0,0$ \pm $0,0$
beta-HCH	1,9 ± 1,1	2,1 ± 0,9	$1,3 \pm 0,4$	0,4 ± 0,3
gamma-HCH	1,0 ± 1,0	$0,6\pm0,6$	$0,1 \pm 0,3$	$0,0$ \pm $0,0$
HCH sum	3,4 ± 2,8	$3,1 \pm 1,3$	1,4 ± 0,6	0,4 ± 0,3
Chlordane sum	$2,7 \pm 1,7$	$2,5 \pm 1,0$	$1,8 \pm 0,4$	$1,9 \pm 0,7$

	Balgzand	Griend	Schiermonnikoog	Delfzijl				
Common Tern	(N=10)	(N=10)	(N=10)	(N=10)				
Hg	$267,1 \pm 88,3$	244,0 ± 101,7	229,2 ± 37,2	369,4 ± 120,8				
6 PCB (law)	371,9 ± 160,7	292,7 ± 110,5	356,4 ± 182,6	425,8 ± 193,6				
PCB sum	791,0 ± 331,5	593,3 ± 215,6	747,5 ± 359,4	905,1 ± 397,0				
HCB	$16,1 \pm 7,0$	12,1 ± 3,2	9,1 ± 3,8	13,3 ± 3,4				
ppDDE	51,1 ± 17,6	38,0 ± 17,5	56,3 ± 29,7	75,6 ± 22,9				
ppDDT	$0,0$ \pm $0,0$	1,5 ± 1,5	$0,0$ \pm $0,0$	$0,0$ \pm $0,0$				
ppDDD	$2,4\pm0,4$	2,7 ± 1,5	$2,3 \pm 0,3$	$2,9 \pm 0,8$				
DDT sum	53,5 ± 17,7	42,3 ± 17,2	58,6 ± 29,8	78,5 ± 22,8				
alpha-HCH	$0,0$ \pm $0,0$	$0,0$ \pm $0,0$	$0,0$ \pm $0,0$	$0,0$ \pm $0,0$				
beta-HCH	$2,4 \pm 2,1$	0,6 ± 0,4	$0,8\pm0,5$	$1,0\pm0,6$				
gamma-HCH	$0,4\pm0,4$	$0,2 \pm 0,2$	$0,1 \pm 0,3$	$0,0$ \pm $0,0$				
HCH sum	$2,7$ \pm $2,3$	$0,8\pm0,5$	$0,9\pm0,6$	$1,0\pm0,6$				
Chlordane sum	$1,8 \pm 0,5$	1,3 ± 0,7	0,4 ± 0,5	$1,2 \pm 1,1$				

8.2 Temporal trends of pollutant concentrations at different sites during 2009-2013



Fig. 8.2.1: Temporal development of pollutant concentrations in Oystercatcher and Common Tern eggs from Balgzand, NL, in the period 2009-2013. Arithmetic means are given with the 95% confidence interval.



Griend

year

Fig. 8.2.2: Temporal development of pollutant concentrations in Oystercatcher and Common Tern eggs from Griend, NL, in the period 2009-2013. Arithmetic means are given with the 95% confidence interval.





Fig. 8.2.3: Temporal development of pollutant concentrations in Oystercatcher eggs from Julianapolder and in Common Tern eggs from Schiermonnikoog, NL, in the period 2009-2013. Arithmetic means are given with the 95% confidence interval.

Delfzijl



Common Tern



year

Fig. 8.2.4: Temporal development of pollutant concentrations in Oystercatcher and Common Tern eggs from Delfzijl, NL, in the period 2009-2013. Arithmetic means are given with the 95% confidence interval.