

Supplementary information

Table S1: Calculated Σ TU EC₁₀ in 2012 at Dutch monitoring locations. The area and location, the coordinates of the location, type of water body, the average, minimum and maximum Σ TU EC₁₀, the number of samples taken in 2012 and the contaminants which were monitored at the specific location in 2012.

Area-location	latitude, longitude DD°MM.mmm"	water body	average Σ TU EC ₁₀	minimum Σ TU EC ₁₀	maximum Σ TU EC ₁₀	number of samples	contaminants included
Eems-Dollard, Bocht van Wattum	N 53 20.096, E 6 56.637	Estuarine	5.2E-02	1.3E-02	1.0E-01	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Wadden Sea, Boomkensdiep	N 53 21.537, E 3 13.007	Estuarine	6.2E-03	2.0E-04	1.2E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Wadden Sea, Dantziggat	N 53 24.068, E 5 43.619	Estuarine	1.1E-02	3.5E-03	2.3E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Wadden Sea, Doove Balg West	N 53 5.063, E 5 17.233	Estuarine	2.5E-02	7.6E-03	6.8E-02	11	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Grevelingen Lake, Dreischor	N 51 42.877, E 3 59.960	Estuarine	2.4E-02	1.2E-02	4.3E-02	13	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Meuse, Eijsden	N 50 46.770, E 5 41.947	River	1.0E-03	< LOD	2.1E-01	336	atrazine, diuron, isoproturon, terbutryn, terbutylazine
North Sea, Goeree 2 km off shore	N 51 50.817, E 3 50.083	Coastal	1.2E-02	6.4E-03	2.5E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Goeree 6 km off shore	N 51 52.183, E 3 52.416	Coastal	1.1E-02	6.9E-03	2.1E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Ems-Dollard, Groote Gat Noord	N 53 18.258, E 7 9.400	Estuarine	7.6E-02	3.1E-02	1.4E-01	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Western Scheldt, Hansweert	N 51 26.167, E 4 0.849	Estuarine	1.9E-01	6.6E-02	4.3E-01	13	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Ems-Dollard, Huibergat Oost	N 53 33.548, E 6 39.672	Estuarine	1.1E-02	5.0E-03	2.1E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Rhine, Lobith	N 51 51.252, E 6 5.471	River	1.9E-02	< LOD	6.5E-01	297	diuron, isoproturon, terbutylazine
Eastern Scheldt, Lodijkse Gat	N 51 30.917, E 4 7.623	Estuarine	2.8E-02	1.5E-02	3.4E-02	4	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Wadden Sea, Marsdiep	N 52 58.952, E 4 44.996	Estuarine	1.2E-02	8.0E-03	1.7E-02	4	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Noordwijk 10 km off shore	N 52 18.133, E 4 18.149	Coastal	1.1E-02	5.7E-03	1.9E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Noordwijk 2 km off shore	N 52 15.683, E 4 24.367	Coastal	1.2E-02	7.6E-03	2.1E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Noordwijk 20 km off shore	N 52 20.500, E 4 10.500	Coastal	5.0E-03	< LOD	1.1E-02	6	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Noordwijk 70 km off shore	N 52 34.167, E 3 31.883	Coastal	4.1E-04	< LOD	4.9E-03	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Rottumerplaat 3 km off shore	N 53 33.966, E 6 33.850	Coastal	8.5E-03	4.6E-03	1.5E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Schouwen 10 km off shore	N 51 43.200, E 3 29.717	Coastal	9.9E-03	5.4E-03	1.6E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Lake Veere, Soelekerke-polder Oost	N 51 32.532, E 3 43.849	Estuarine	8.9E-02	5.1E-02	1.5E-01	13	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Western Scheldt, Temeuzen boei 20	N 51 20.792, E 3 49.529	Estuarine	1.4E-01	8.1E-02	3.2E-01	13	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Terschelling 10 km off shore	N 53 27.667, E 5 6.050	Coastal	3.8E-03	< LOD	1.0E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Terschelling 135 km uit de kust	N 54 24.933, E 4 2.467	Coastal	< LOD	< LOD	< LOD	6	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine

North Sea, Terschelling 235 km uit de kust	N 53 10.333, E 3 9.450	Coastal	< LOD	< LOD	< LOD	4	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Terschelling 50 km uit de kust	N 53 46.050, E 4 46.017	Coastal	< LOD	< LOD	< LOD	6	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Western Scheldt, Vlissingen boei SSVH	N 51 24.720, E 3 33.937	Estuarine	5.1E-02	1.9E-02	1.1E-01	13	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
North Sea, Walcheren 2 km	N 51 32.933, E 3 24.649	Coastal	1.3E-02	5.4E-03	3.1E-02	12	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Eastern Scheldt, Wissenkerke	N 51 36.095, E 3 43.234	Estuarine	1.6E-02	6.3E-03	2.3E-02	13	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine
Eastern Scheldt, Zijpe	N 51 38.683, E 4 5.819	Estuarine	2.7E-02	9.0E-03	4.1E-02	4	atrazine, diuron, isoproturon, irgarol, terbutryn, terbutylazine

< LOD: Chemical concentrations provided by the Dutch ministry of infrastructure were below the limit of detection (LOD). LODs were not provided.

Table S2: LC-MS optimized settings for multiple reaction monitoring used for the analysis of the contaminants.

Contaminant	MW	MRM transitions (m/z)	Collision energy (eV)	Fragmentor voltage
Atrazine	215	216.1 → 174.0	16	136
Diuron	232	233.0 → 72.1	20	112
Irgarol	253	254.1 → 198.1	16	112
Isoproturon	206	207.1 → 72.1	20	104
Terbutylazine	229	230.1 → 174.0	16	116
Terbutryn	241	242.1 → 186.1	16	70

Table S3: Annual average concentrations of the key contaminants from 2003 to 2012 at Ems-Dollard, Western Scheldt, Wadden Sea and North Sea. MWTL monitoring data provided by the Dutch ministry of infrastructure and the environment.

location	year	annual average concentration (µg/L)					
		atrazine (0.6) ¹	diuron (0.2) ¹	isoproturon (0.3) ¹	irgarol (0.0025) ¹	terbutryn (0.0065) ¹	Terbutylazine (0.19) ²
Ems-Dollard <i>Groote Gat Noord</i>	2003	0.004	0.035	0.039	0.001	0.002	0.009
	2004	0.003	0.043	0.036	0.002	0.002	0.008
	2005	0.003	0.040	0.025	0.002	0.002	0.008
	2006	0.002	0.037	0.017	0.002	0.002	0.007
	2007	n.d.	0.027	0.031	0.002	0.001	n.d.
	2008	n.d.	0.019	0.016	0.002	0.001	n.d.
	2009	0.001	0.014	0.010	0.003	0.001	0.011
	2010	0.001	0.008	0.006	0.002	0.001	0.014
	2011	0.001	0.008	0.006	0.002	0.001	0.014
	2012	0.002	0.005	0.007	0.002	0.002	0.016
	Western Scheldt <i>Hansweert</i>	2003	0.061	0.222	0.053	0.003	0.003
2004		0.040	0.199	0.048	0.003	0.003	0.006
2005		0.035	0.145	0.048	0.007	0.003	0.009
2006		0.021	0.113	0.042	0.008	0.003	0.011
2007		n.d.	0.082	0.029	0.008	0.003	n.d.
2008		n.d.	0.084	0.042	0.010	0.003	n.d.
2009		0.007	0.041	0.029	0.008	0.002	0.025
2010		0.006	0.029	0.022	0.005	0.002	0.017
2011		0.005	0.023	0.015	0.003	0.002	0.018
2012		0.008	0.022	0.029	0.003	0.002	0.038
Wadden Sea <i>Marsdiep</i>		2003	0.005	0.015	0.005	0.001	0.001
	2004	0.004	0.014	0.005	0.001	0.001	0.001
	2005	0.004	0.011	0.004	0.001	0.001	0.001
	2006	0.003	0.009	0.004	0.001	0.001	0.001
	2007	n.d.	0.007	0.003	0.001	0.001	n.d.
	2008	n.d.	0.006	0.006	0.002	0.001	n.d.
	2009	0.002	0.005	0.004	0.002	0.001	0.001
	2010	0.002	0.003	0.002	0.001	0.001	0.002
	2011	0.001	0.002	0.001	<LOD	<LOD	0.002
	2012	0.001	0.002	0.002	<LOD	<LOD	0.002
	North Sea <i>Noordwijk</i>	2003	0.006	0.015	0.005	0.001	0.001
2004		0.004	0.017	0.005	0.001	0.001	0.001
2005		0.004	0.016	0.006	0.001	0.001	0.001
2006		0.003	0.010	0.006	0.001	0.001	0.001
2007		n.d.	0.008	0.004	0.001	0.001	n.d.
2008		n.d.	0.007	0.005	0.002	0.001	n.d.
2009		0.002	0.005	0.004	0.002	0.001	0.002
2010		0.001	0.003	0.003	0.001	<LOD	0.001
2011		0.001	0.003	0.002	<LOD	<LOD	0.001
2012		0.001	0.003	0.003	<LOD	<LOD	0.001

¹ Annual average (AA-EQS) concentration European Commission. Directive 2013/39/EU

² Maximum allowable risk level (RIVM, 2014)

n.d. no data available

< LOD: Chemical concentrations provided by the Dutch ministry of infrastructure were below the limit of detection (LOD). LODs were not provided.

Table S4: Maximum concentrations of the key contaminants from 2003 to 2012 at Ems-Dollard, Western Scheldt, Wadden Sea and North Sea. MWTL monitoring data provided by the Dutch ministry of infrastructure and the environment.

location	year	maximum concentration (µg/L)					
		atrazine (2) ¹	diuron (1.8) ¹	isoproturon (1) ¹	irgarol (0.016) ¹	terbutryn (0.034) ¹	Terbutylazine (0.19) ²
Ems-Dollard <i>Groote Gat Noord</i>	2003	0.005	0.056	0.130	0.002	0.002	0.026
	2004	0.005	0.066	0.180	0.003	0.002	0.020
	2005	0.005	0.067	0.055	0.003	0.003	0.022
	2006	0.004	0.051	0.049	0.003	0.002	0.014
	2007	n.d.	0.049	0.170	0.003	0.003	n.d.
	2008	n.d.	0.029	0.033	0.004	0.002	n.d.
	2009	0.002	0.020	0.025	0.004	0.002	0.026
	2010	0.003	0.012	0.019	0.003	0.002	0.030
	2011	0.002	0.012	0.026	0.003	0.002	0.042
	2012	0.005	0.009	0.021	0.002	0.002	0.033
	Western Scheldt <i>Hansweert</i>	2003	0.130	0.370	0.120	0.004	0.005
2004		0.065	0.290	0.081	0.005	0.004	0.010
2005		0.088	0.200	0.088	0.011	0.005	0.023
2006		0.030	0.160	0.093	0.012	0.004	0.023
2007		n.d.	0.120	0.066	0.011	0.003	n.d.
2008		n.d.	0.130	0.088	0.016	0.004	n.d.
2009		0.009	0.055	0.069	0.011	0.003	0.042
2010		0.008	0.037	0.042	0.007	0.003	0.027
2011		0.006	0.033	0.043	0.004	0.002	0.032
2012		0.014	0.030	0.088	0.006	0.004	0.132
Wadden Sea <i>Marsdiep</i>		2003	0.005	0.017	0.015	0.001	0.001
	2004	0.005	0.015	0.013	0.001	0.001	0.002
	2005	0.005	0.014	0.008	0.001	0.001	0.001
	2006	0.003	0.012	0.006	0.002	0.001	0.001
	2007	n.d.	0.009	0.005	0.002	0.001	n.d.
	2008	n.d.	0.008	0.016	0.002	0.001	n.d.
	2009	0.002	0.006	0.007	0.002	0.001	0.002
	2010	0.002	0.004	0.007	0.002	0.001	0.002
	2011	0.002	0.003	0.003	<LOD	<LOD	0.003
	2012	0.002	0.003	0.006	<LOD	<LOD	0.004
	North Sea <i>Noordwijk</i>	2003	0.011	0.023	0.014	0.002	0.002
2004		0.006	0.024	0.011	0.001	0.002	0.002
2005		0.006	0.036	0.013	0.002	0.002	0.002
2006		0.004	0.013	0.012	0.002	0.002	0.001
2007		n.d.	0.012	0.010	0.002	0.001	n.d.
2008		n.d.	0.013	0.013	0.003	0.001	n.d.
2009		0.003	0.008	0.010	0.003	0.002	0.005
2010		0.002	0.006	0.010	0.002	0.002	0.003
2011		0.002	0.006	0.007	0.006	0.001	0.003
2012		0.002	0.003	0.013	<LOD	<LOD	0.004

¹ Maximum allowable concentration (MAC-EQS) European Commission. Directive 2013/39/EU

² Maximum allowable risk level (RIVM, 2014)

n.d. no data available

< LOD: Chemical concentrations provided by the Dutch ministry of infrastructure were below the limit of detection (LOD). LODs were not provided

Table S5: Annual average TU EC10 of the identified contaminants from 2003 to 2012 at Ems-Dollard, Western Scheldt, Wadden Sea and North Sea.

Location	year	annual average TU EC10						
		atrazine (0.104) ¹	diuron (0.597) ¹	isoproturon (0.218) ¹	irgarol (0.005) ¹	terbutryn (0.102) ¹	Terbutylazine (0.422) ²	ΣTU EC10
Ems-Dollard <i>Groote Gat Noord</i>	2003	0.001	0.103	0.028	0.002	0.017	0.020	0.171
	2004	0.001	0.129	0.026	0.003	0.026	0.017	0.202
	2005	0.001	0.119	0.018	0.004	0.030	0.017	0.189
	2006	0.000	0.109	0.012	0.004	0.024	0.014	0.164
	2007	n.d.	0.079	0.023	0.003	0.022	n.d.	0.127
	2008	n.d.	0.058	0.012	0.004	0.022	n.d.	0.096
	2009	0.000	0.041	0.007	0.005	0.021	0.025	0.099
	2010	0.000	0.025	0.004	0.004	0.021	0.031	0.085
	2011	0.000	0.024	0.004	0.003	0.018	0.032	0.082
	2012	0.000	0.016	0.005	0.003	0.025	0.027	0.076
Western Scheldt <i>Hansweert</i>	2003	0.011	0.663	0.039	0.004	0.045	0.017	0.778
	2004	0.007	0.595	0.035	0.006	0.051	0.014	0.708
	2005	0.006	0.433	0.035	0.013	0.054	0.019	0.559
	2006	0.004	0.337	0.031	0.014	0.050	0.024	0.460
	2007	n.d.	0.246	0.021	0.014	0.041	n.d.	0.321
	2008	n.d.	0.251	0.031	0.018	0.043	n.d.	0.343
	2009	0.001	0.122	0.021	0.014	0.033	0.056	0.247
	2010	0.001	0.088	0.016	0.009	0.026	0.037	0.178
	2011	0.001	0.069	0.011	0.006	0.026	0.041	0.155
	2012	0.001	0.065	0.021	0.005	0.034	0.065	0.192
Wadden Sea <i>Marsdiep</i>	2003	0.001	0.043	0.004	0.001	0.013	0.003	0.065
	2004	0.001	0.043	0.004	0.001	0.014	0.003	0.065
	2005	0.001	0.034	0.003	0.002	0.015	0.002	0.057
	2006	0.000	0.027	0.003	0.002	0.012	0.002	0.047
	2007	n.d.	0.021	0.002	0.002	0.011	n.d.	0.036
	2008	n.d.	0.019	0.005	0.003	0.012	n.d.	0.038
	2009	0.000	0.014	0.003	0.003	0.013	0.003	0.036
	2010	0.000	0.009	0.002	0.002	0.009	0.004	0.026
	2011	0.000	0.007	0.001	0.000	0.000	0.003	0.012
	2012	0.000	0.007	0.002	0.000	0.000	0.003	0.012
North Sea <i>Noordwijk</i>	2003	0.001	0.044	0.004	0.001	0.017	0.003	0.070
	2004	0.001	0.051	0.004	0.002	0.017	0.002	0.077
	2005	0.001	0.048	0.004	0.003	0.020	0.002	0.078
	2006	0.001	0.030	0.005	0.002	0.016	0.002	0.055
	2007	n.d.	0.024	0.003	0.002	0.014	n.d.	0.043
	2008	n.d.	0.022	0.004	0.003	0.014	n.d.	0.043
	2009	0.000	0.016	0.003	0.003	0.014	0.004	0.040
	2010	0.000	0.010	0.002	0.001	0.006	0.002	0.022
	2011	0.000	0.010	0.002	0.001	0.005	0.003	0.020
	2012	0.000	0.008	0.002	0.000	0.000	0.002	0.012

¹ Calculated Annual average (AA-EQS) TU EC10 based on concentrations from European Commission. Directive 2013/39/EU

² Calculated maximum allowable risk TU EC10 based on the maximum allowable risk level (RIVM, 2014)

n.d. no data available

Table S6: Maximum TU EC10 of the identified contaminants from 2003 to 2012 at Ems-Dollard, Western Scheldt, Wadden Sea and North Sea.

Location	year	maximum TU EC10					Terbutylazine (0.422) ²	ΣTU EC10
		atrazine (0.347) ¹	diuron (5.375) ¹	isoproturon (0.729) ¹	irgarol (0.029) ¹	terbutryn (0.535) ¹		
Ems-Dollard Groote Gat Noord	2003	0.001	0.167	0.095	0.004	0.036	0.058	0.277
	2004	0.001	0.197	0.131	0.005	0.036	0.044	0.281
	2005	0.001	0.200	0.040	0.006	0.046	0.049	0.295
	2006	0.001	0.152	0.036	0.006	0.033	0.031	0.215
	2007	n.d.	0.146	0.124	0.005	0.042	n.d.	0.235
	2008	n.d.	0.087	0.024	0.007	0.025	n.d.	0.122
	2009	0.000	0.060	0.018	0.007	0.031	0.058	0.146
	2010	0.001	0.036	0.014	0.006	0.030	0.067	0.132
	2011	0.000	0.034	0.019	0.006	0.033	0.094	0.157
	2012	0.001	0.027	0.016	0.004	0.038	0.074	0.138
Western Scheldt Hansweert	2003	0.023	1.105	0.087	0.007	0.076	0.024	1.250
	2004	0.011	0.866	0.059	0.009	0.068	0.022	0.996
	2005	0.015	0.597	0.064	0.020	0.083	0.051	0.787
	2006	0.005	0.478	0.068	0.022	0.069	0.051	0.630
	2007	n.d.	0.358	0.048	0.020	0.054	n.d.	0.430
	2008	n.d.	0.388	0.064	0.029	0.069	n.d.	0.475
	2009	0.002	0.164	0.050	0.020	0.047	0.093	0.336
	2010	0.001	0.110	0.031	0.013	0.039	0.060	0.225
	2011	0.001	0.099	0.031	0.008	0.036	0.072	0.205
	2012	0.002	0.090	0.064	0.011	0.061	0.294	0.431
Wadden Sea Marsdiep	2003	0.001	0.051	0.011	0.001	0.022	0.005	0.071
	2004	0.001	0.045	0.009	0.001	0.017	0.004	0.077
	2005	0.001	0.042	0.005	0.003	0.019	0.003	0.065
	2006	0.001	0.036	0.005	0.003	0.017	0.003	0.063
	2007	n.d.	0.025	0.003	0.003	0.014	n.d.	0.042
	2008	n.d.	0.024	0.012	0.004	0.016	n.d.	0.041
	2009	0.000	0.018	0.005	0.004	0.022	0.005	0.050
	2010	0.000	0.012	0.005	0.003	0.022	0.005	0.043
	2011	0.000	0.009	0.002	0.000	0.000	0.006	0.015
	2012	0.000	0.008	0.005	0.000	0.000	0.009	0.017
North Sea Noordwijk	2003	0.002	0.069	0.010	0.003	0.035	0.006	0.122
	2004	0.001	0.072	0.008	0.003	0.028	0.004	0.103
	2005	0.001	0.108	0.009	0.004	0.036	0.004	0.161
	2006	0.001	0.039	0.009	0.003	0.027	0.003	0.075
	2007	n.d.	0.036	0.007	0.003	0.022	n.d.	0.064
	2008	n.d.	0.039	0.009	0.005	0.020	n.d.	0.071
	2009	0.001	0.024	0.007	0.005	0.024	0.010	0.060
	2010	0.000	0.018	0.007	0.004	0.031	0.006	0.061
	2011	0.000	0.019	0.005	0.010	0.023	0.007	0.047
	2012	0.000	0.010	0.009	0.000	0.000	0.008	0.021

¹ Calculated Maximum allowable concentration (MAC-EQS) TU EC10 based on concentrations from European Commission. Directive 2013/39/EU

² Calculated maximum allowable risk level TU EC10 based on the maximum allowable risk level (RIVM, 2014)

n.d. no data available