Table 1 Physical and biogeochemical parameters at the different stations where vertical profiles were carried out during both campaigns (M2-0407 and M3-0510), SPM: suspended particulate matter, POC: particulate organic carbon.

Stations and locations	Date	Depth	Daily River discharge	Water Temperature	Daily Radiation	Salinity	Turbidity	SPM	POC	Chlorophyll a	Phaeo-pigment	Nitrates	Phos-phates	Silicic acid
		m	$m^3\ s^{-1}$	°C	$Wh\ m^{-2}$		NTU	mgL^{-1}	%	μgL^{-1}	μgL^{-1}	µM	µM	µM
Metadour 2 (April 2007) M2-0407														
IE-2 (43°30.2'N, - 01°29.5'W)	11.04.2007	0.5	383.7	14.3	292	0.9	10.8	11.5	4.7	3.6	9.4	115.3	0.1	58.9
M2-A (43°31.7'N, -01°32.2'W)	08.04.2007	0.5	393.5	12.7	550	25.2	n/a	2.7	15.2	4.1	7.6	36.1	nd	8.2
		4		12.6		30.4	n/a	2.9	13.1	12.1	30.8	5.5	nd	0.9
		12		12.5		34.6	n/a	3.2	8.5	3.8	11.6	1.2	nd	1.1
M2-B (43°31.9'N, -01°37.1'W)	10.04.2007	0.5	393.9	13.5	409	25.3	1.3	3.6	12.7	5.5	14.3	20.4	nd	5.3
		2		13.1		30.2	0.8	3.2	15.2	6.5	18.6	40.8	nd	11.2
		4		12.7		32.5	0.2	1.7	19.8	9.6	29.2	6.1	0.1	1.4
		8		12.2		33.2	nd	1.4	21.3	9.3	22.0	2.4	nd	1.2
		20		12.5		35.0	nd	0.4	20.6	0.1	2.3	nd	nd	0.2
		40		12.9		35.4	nd	0.5	15.2	0.4	1.0	2.0	0.1	1.3
M2-C (43°33.5'N, -01°45.3'W)	09.04.2007	0.5	468.6	13.5	438	32.2	nd	4.6	9.9	5.1	14.2	n/a	nd	11.8
		4		13.4		34.2	nd	0.5	18.8	0.7	2.0	0.4	nd	1.1
		12		12.6		34.6	nd	0.2	36.7	0.8	2.4	0.4	0.1	1.6
		20		12.7		35.0	nd	0.3	18.8	0.7	n/a	1.0	nd	2.0
		30		12.8		35.3	nd	0.2	32.3	0.7	1.6	1.4	0.1	2.3
		50		13.0		35.6	nd	0.3	24.5	0.6	1.5	2.4	0.1	2.2
Metadour 3 (May 2010) M3-0510														
IE-3 (43°30.2'N, -01°29.5'W)	15.05.2010	0.5	476.8	12.3	235	0.2	n/a	18.9	4.6	0.5	4.2	114.1	0.2	7.3
		6		12.3		0.2	n/a	20.9	4.4	n/a	n/a	145.7	0.2	41.9
M3-D (43°31.1'N, -01°34.3'W)	16.05.2010	0.5	573.0	14.3	520	27.7	4.9	4.6	6.7	0.9	2.7	12.6	0.1	4.7
		1	452.1	14.3		27.1	4.6	4.3	n/a	0.5	2.2	15.3	0.1	7.5
		5		14.5		35.0	1.0	0.7	n/a	0.8	1.9	3.4	nd	0.5
		24		14.2		35.4	0.4	0.1	n/a	0.3	0.8	1.3	0.1	n/a
M3-E (43°33.3'N, -01°45.2'W)	17.05.2010	0.5	389.4	15.5	411	34.6	1.4	1.1	9.3	0.4	1.2	2.3	0.1	0.3
		1	452.1	15.3		31.5	1.5	1.2	n/a	1.3	2.8	1.0	0.1	nd
		4		14.8		35.2	0.4	0.1	n/a	1.3	2.8	n/a	n/a	n/a
		13		14.4		35.2	0.3	nd	n/a	0.5	1.2	0.4	0.1	nd
		20		14.0		35.6	0.1	nd	n/a	0.5	1.2	n/a	n/a	n/a
		55		13.7		35.6	0.1	nd	n/a	0.1	0.5	3.4	0.2	nd
		80		13.4		35.6	0.3	nd	n/a	0.1	0.5	n/a	n/a	n/a
M3-F (43°36.1'N, -01°47.8'W)	18.05.2010	5	412.3	14.7	724	35.2	0.3	0.7	10.0	0.4	1.2	2.4	0.1	nd
		22		14.1		35.4	0.3	0.9	10.8	0.8	2.3	7.8	0.1	nd
		40		12.5		35.6	0.1	0.8	10.4	n/a	n/a	n/a	n/a	n/a
		80		12.3		35.7	0.1	nd	n/a	n/a	n/a	n/a	n/a	n/a
		240		12.0		35.7	0.4	0.1	n/a	0.2	0.3	10.0	0.1	0.3
		800		10.7		35.7	0.5	0.1	n/a	0.1	0.2	4.4	0.2	0.1

(n/a: not available, nd: not detected)

Table 2 Summary of Hg species concentration detected in the various vertical profiles carried out during both campaigns, Hg_T = DGM + $MeHg_T$ + $Hg(II)_T$. RSD for all species are < 10%.

Station	Depth	DGM	$MeHg_D$	$Hg(II)_D$	$MeHg_P$	$Hg(II)_P$	$MeHg_T$	$Hg(II)_T$	Hg_T	Hg_P/ Hg_T	$Hg(II)_T$/ Hg_T	$MeHg_T$/ Hg_T
	m	pM			nmol g^{-1}		pM			%	%	%
Metadour 2 (April 2007) M2-0407												
IE-2 (43°30.2'N, -01°29.5W)	0.5	n/a	0.16	1.70	0.08	11.54	0.24	13.24	13.48	86.2	98.2	1.8
M2-A (43°31.7'N, -01°32.2'W)	0.5	0.26	0.11	1.66	0.03	2.96	0.14	5.47	5.93	59.6	92.0	2.8
	4	0.19	0.10	2.22	0.05	2.81	0.15	4.23	4.43	53.2	93.5	2.9
	12	0.11	0.13	5.78	0.02	1.97	0.14	4.29	4.34	25.1	98.2	1.8
M2-B (43°31.9'N, -01°37.1'W)	0.5	0.28	0.17	1.66	0.02	1.50	0.19	3.16	3.63	41.9	87.0	5.2
	2	0.25	0.06	1.05	0.01	0.85	0.07	1.89	2.22	38.6	85.4	3.4
	4	0.29	0.11	1.73	0.01	0.72	0.12	2.45	2.87	25.4	85.5	4.3
	8	0.25	0.13	0.77	0.01	0.89	0.14	1.65	2.04	43.9	80.8	7.1
	20	0.23	0.06	0.67	0.01	0.88	0.07	1.55	1.85	48.2	83.9	3.7
	40	0.31	0.07	0.92	0.01	0.78	0.08	1.70	2.09	37.5	81.5	3.7
M2-C (43°33.5'N, -01°45.3'W)	0.5	0.23	0.09	2.94	0.04	2.16	0.13	5.09	5.46	40.3	93.3	2.5
	4	0.33	0.06	2.16	<d.l.	0.49	0.06	2.64	3.04	16.1	87.1	2.0
	12	0.25	0.11	0.65	0.01	0.33	0.11	0.97	1.33	25.0	72.8	8.5
	20	0.33	0.10	0.84	0.01	4.26	0.11	5.10	5.54	77.1	92.2	1.9
	30	0.27	0.05	1.96	0.01	0.81	0.06	2.77	3.10	26.5	89.5	1.9
	50	0.29	0.09	1.03	0.01	1.85	0.09	2.88	3.26	56.9	88.4	2.8
Metadour 3 (May 2010) M3-0510												
IE-3 (43°30.2'N, -01°29.5'W)	0.5	0.12	0.27	1.41	0.35	5.41	0.62	6.83	7.57	76.2	90.2	8.2
	6	0.54	0.22	1.32	0.26	1.17	0.47	2.49	3.50	40.7	71.2	13.4
M3-D (43°31.1'N, -01°34.3'W)	0.5	0.14	0.11	1.62	0.06	0.89	0.17	2.51	2.82	32.5	88.4	6.3
	1	0.26	0.09	1.38	0.02	0.52	0.11	1.90	2.28	23.9	83.6	4.8
	5	0.13	0.04	1.10	0.01	0.07	0.04	1.17	1.35	5.7	86.8	3.2
	24	0.19	0.05	0.49	0.01	0.09	0.05	0.58	0.82	11.2	70.5	6.4
M3-E (43°33.3'N, -01°45.2'W)	0.5	0.20	0.07	1.14	0.04	0.90	0.09	2.00	2.29	37.0	87.4	4.6
	1	0.27	0.09	0.95	0.14	2.37	0.33	3.84	4.44	56.7	86.5	7.5
	4	0.26	0.02	0.61	0.16	2.17	0.18	2.78	3.22	72.1	86.3	5.5
	13	0.31	0.04	1.08	0.07	0.96	0.11	2.04	2.47	42.0	82.9	4.6
	20	0.23	0.08	1.06	0.10	1.63	0.18	2.69	3.09	55.8	86.9	5.7
	55	0.25	0.04	1.06	0.15	1.77	0.19	2.84	3.27	58.7	86.6	5.8
	80	0.28	0.12	2.33	0.08	0.54	0.20	2.86	3.35	18.4	85.6	6.0
M3-F (43°36.1'N, -01°47.8'W)	5	0.21	0.06	1.64	0.10	1.19	0.16	2.83	3.20	40.2	88.3	5.1
	22	0.25	0.10	1.69	0.07	1.21	0.16	2.56	2.97	43.1	86.2	5.3
	40	0.29	0.11	1.82	0.05	1.23	0.15	3.06	3.50	36.5	87.3	4.4
	80	0.18	0.10	1.62	0.06	2.22	0.16	3.83	4.18	54.5	91.7	3.9
	240	0.17	0.12	1.26	0.05	3.99	0.16	5.25	5.58	72.3	94.1	2.9
	800	0.34	0.13	0.95	0.30	1.92	0.43	2.87	3.64	61.1	78.9	11.8

Table 3 Methylation, Demethylation and Reduction potentials (mean ± SD, n = 3) in filtered and unfiltered surface waters performed under light and dark conditions for both campaigns: M2-0407 (M2-A, M2-B, M2-C) and M3-0510 (M3-D, M3-E, M3-F). Detection limits are 0.01, 2.0 and 0.3 % for methylation, demethylation and reduction yields, respectively.

	Hg(II) Methylation (% day $^{-1}$)				**MeHg Demethylation** (% day $^{-1}$)				**Hg Reduction** (% day $^{-1}$)			
	Unfiltered waters		Filtered waters		Unfiltered waters		Filtered waters		Unfiltered waters		Filtered waters	
Stations	Diurnal	Dark	Diurnal	Dark	Diurnal	Dark	Diurnal	Dark	Diurnal	Dark	Diurnal	Dark
M2-A	< 0.01	0.1 ± 0.1	< 0.01	< 0.01	50.2 ± 13.5	20.5 ± 12.2	25.3 ± 11.4	9.6 ± 3.3	6.5 ± 0.2	8.2 ± 0.8	4.3 ± 2.3	1.6 ± 0.4
M2-B	0.4 ± 0.1	0.1 ± 0.1	0.3 ± 0.1	< 0.01	18.8 ± 4.0	18.0 ± 7.3	15.4 ± 2.5	6.1 ± 0.2	4.5 ± 0.5	1.2 ± 0.5	10.5 ±1.4	0.3 ± 1.4
M2-C	0.1 ± 0.1	< 0.01	< 0.01	< 0.01	31.7 ± 6.1	20.8 ± 9.3	28.5 ± 13.8	13.2 ± 6.1	7.2 ± 0.4	8.5 ± 2.2	9.0 ± 0.5	7.3 ± 1.3
M3-D	< 0.01	< 0.01	< 0.01	< 0.01	55.3 ± 35.0	22.1 ± 4.6	45.5 ± 12.1	2.9 ± 1.4	16.8 ± 3.4	14.7 ± 2.9	43.5 ± 8.7	7.2 ± 1.4
M3-E	< 0.01	< 0.01	< 0.01	< 0.01	23.5 ± 5.8	6.8 ± 0.9	8.0 ± 1.2	2.3 ± 3.0	19.7 ± 3.9	10.5 ± 2.1	25.0 ± 5.0	9.0 ± 1.8
M3-F	< 0.01	< 0.01	< 0.01	< 0.01	6.6 ± 3.5	10.9 ± 3.6	24.3 ± 9.4	< 2.0	29.8 ± 6.0	5.0 ± 1.0	10.2 ± 2.0	2.8 ± 0.6

Table 4 Potential daily variations of MeHg and gaseous Hg concentrations in the Adour estuarine plume, calculated using models described in section 3.2.2 and 3.2.3

Stations	Δ[MeHg]		Δ[Hg^0]*	
	pmol m^{-3} d^{-1}	ng m^{-3} d^{-1}	pmol m^{-3} d^{-1}	ng m^{-3} d^{-1}
M2-A concentrated plume	-64 ± 51	-13 ± 10	-55 ± 10	-11 ± 2
M2-B concentrated plume	16 ± 1	3 ± 0	-254 ± 0	-51 ± 0
M2-C diluted plume	-26 ± 7	-5 ± 1	-140 ±20	-28 ± 4
M3-D concentrated plume	-91 ± 39	-18 ± 8	-174 ±74	-35 ± 15
M3-E diluted plume	-27 ± 20	-5 ± 4	-261 ±96	-52 ± 19
M3-F marine water	-21 ± 1	-4 ± 0	-180 ± 19	-36 ±4

*Clark's model (Clark et al., 1995) for gas exchange and Plume mixed layer depth of 1m used for Δ[Hg°] calculations

Table 5 Comparisons between estuarine inputs of $MeHg_T$ and Hg_T and integrated demethylation in plume waters and Hg^0 evasion from the plume to the atmosphere. Values in brackets were reported by Point, (2004) for the same site.

		Plume surface area	Estuarine input $MeHg_T$	Net MeHg demethylation*	Demethylation loss of $MeHg_T$	Estuarine input Hg_T	Hg^0 Flux density#	Atmospheric evasion of Hg^0
		km^2	(g d^{-1})	ng m^{-2} d^{-1}	g d^{-1}	g d^{-1}	ng m^{-2} d^{-1}	g d^{-1}
M2-0407	Concentrated plume	274		11.2 ± 0.1	3.1 ± 0.04		78.6 ± 48.3	21.6 ± 13.2
	Diluted plume	119		5.2 ± 1.4	0.6 ± 0.2		45.0 ± 1.4	5.4 ± 0.2
	Total plume	**393**	**0.9 ± 0.1 (3.3 ± 5.2)**		**3.7 ± 0.2**	**69.1 ± 53.6 (44.4 ± 28.1)**		**26.9 ± 13.4**
M3-0510	Concentrated plume	83		18.2 ± 7.9	1.5 ± 0.6		38.8 ± 19.7	3.2 ± 1.6
	Diluted plume	94		5.4 ± 3.9	0.5 ± 0.4		59.3 ± 22.2	5.6 ± 2.1
	Total plume	**177**	**3.5 ± 0.3 (3.3 ± 5.2)**		**2.0 ± 1.0**	**33.5 ± 17.0 (44.4 ± 28.1)**		**8.8 ± 3.7**

*Area normalized demethylation rate assuming an estuarine plume mixed layer depth of 1 m.
Hg° evasion calculated using gas exchange model (Clark et al., 1995)