Supplementary material

Classification of clouds sampled at the puy de Dôme (France) from 10-year monitoring: Mean features of their physico-chemical properties

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Short summary of previous studies performed at the pdD station

During the past ten years, many studies have been done at the pdD station by scientists from various research fields such as atmospheric physic and chemistry, photochemistry and microbiology. Monitoring of the gas phase and aerosol particles was performed and several investigations were conducted on the physico-chemical processes occurring in the cloud medium. Major conclusions are briefly recalled below.

The physico-chemical characterization of aerosol particles was studied together with their ability to act as cloud condensation nuclei (CCN). A strong seasonal variability in aerosol concentrations overlapping with diurnal variations was highlighted, with maxima during summer and daytime and minima during winter and nighttime (Venzac et al., 2009). This diurnal increase was mainly attributed to the variation of the boundary layer height, which reaches the pdD summit more frequently during the warm season and under sunny conditions. The link between the chemical composition of aerosols and the air mass origin was also investigated, showing that higher SO_4^{2-}, NO_3^- and NH_4^+ mass contents were well correlated with an anthropogenic influence (Sellegri et al., 2003b). This was confirmed by a recent study using an Aerosol Mass Spectrometer that followed the particle chemical composition. It is strongly influenced by both the season and the origin of the air mass (Freney et al., 2011). This monitoring of aerosol particles at the pdD site contributed to the CARBOSOL campaign that focused on the variability of aerosol concentrations and sources over Europe (Pio et al., 2007). It showed that the organic carbon (from 50 to 80 %) contained in the solid particle was water-soluble, and 63 to 78 % was produced from secondary processes (Gelencsér et al., 2007).

Cloud water has been sampled and analyzed in the past at the pdD mountain. Particularly, the chemical composition of the cloud aqueous phase was characterized and a strong variability in the ionic species composition between cloud events with different regional influences (like anthropogenic or marine) was reported (Marinoni et al., 2004). Parazols *et al.* observed higher aqueous concentrations of dissolved iron (Fe) in anthropogenic air masses compared to marine ones (Parazols et al., 2006). The cloud water pH seemed to be the most significant factor controlling the total dissolved iron content. H_2O_2, another strong oxidant, was analyzed (Marinoni et al., 2011): higher differences between day and night of H_2O_2 concentrations were observed for air masses influenced by anthropogenic activities (mainly Northern directions) compared to those from remote areas (oceanic). The mass transfer from the gas phase seemed to prevail its photo-production in the aqueous phase.

Finally, cloud process models were developed and used to interpret these *in situ* measurements (Deguillaume et al., 2004; Leriche et al., 2001; Leriche et al., 2007). The partitioning of different species between interstitial air and condensed phases in clouds was followed at the pdD mountain (Voisin et al., 2000; Sellegri et al., 2003a). For specific cloud events, SO_2/SO_4^{2-}, HNO_3/NO_3^- and NH_3/NH_4^+ were mainly found in the liquid phase while acetic and formic acids were predominant in the interstitial gas phase. A cloud chemistry model was applied to simulate clouds forming at the pdD (Leriche et al., 2007). Numerical results were in good agreement with measurements regarding the contribution of gas and aerosol phases to the concentrations of aqueous chemical species. The model allowed quantifying the contribution of in-cloud chemical reactivity to the concentrations of targeted chemical compounds that are significant for sulfate.

Table captions

- **Table S1.** Description of the physico-chemical analysis performed for each cloud event. The total number of samples is 199 corresponding to 73 cloud events. (IC: Ionic Chromatography: concentrations of organic and inorganic compounds). Air masses are divided into four different back-trajectory sectors: West (W), Northwest/North (NW/N), Northeast (NE) and South/Southwest (S/SW) based on their geographical origins. Cloud events are classified following the Principal Component Analysis (PCA) as Polluted (P), Continental (C), Marine (M) and Highly Marine (HM) and this classification is indicated in the column named "Categories".

- **Table S2.** Minimum, maximum and average value for parameters measured in the present study (chemical concentrations in μeqL^{-1} and physico-chemical data such as pH, liquid water content, conductivity, redox potential). The categories are defined as: P = polluted, C = continental, M = marine and HM = Highly Marine. Values determined in recent studies on the cloud water chemical composition are also reported for comparison. BDL: Below Detection Limit.

- **Table S3.** Total Organic Carbon (TOC) measurements (min, max, average) in cloud water performed during field campaigns at various sites. Air mass origins of the samples are also indicated ("Influence").

- **Figure S1.** TOC concentrations as a function of Liquid Water Content (LWC) for the categories (Continental, Marine, Highly Marine) following the ACP analysis.

Event number	Date	Number of sampling	Origin	Categories	pH	Conductivity	Redox potential	Iron speciation	H_2O_2	TOC	Ionic chromatography
1	08/02/2001	6	SW/W/NW	HM/M		■		■			■
2	12/02/2001	9	W/NW/N	M/C				■			■
3	17/02/2001	11	N/NE	M/C/P	■			■			■
4	22/02/2001	18	N	M/C	■			■			■
5	28/02/2001	12	W/NW	HM/M/C	■			■			■
6	02/03/2001	8	SW	M	■			■		■	■
7	08/03/2001	2	W	M/C	■			■			■
8	05/04/2001	1	SW	M				■		■	■
9	14/02/2002	1	SW	C	■			■	■	■	■
10	22/02/2002	1	SW	C	■			■			■
11	27/02/2002	1	SW	M	■			■	■	■	■
12	28/02/2002	1	W	HM	■			■	■	■	■
13	07/03/2002	1	NW	M	■			■	■	■	■
14	13/04/2002	1	N	C	■			■	■	■	
15	26/04/2002	1	W	M	■			■		■	
16	03/05/2002	1	W	C	■			■	■	■	■
17	11/05/2002	1	NW	C	■			■	■	■	■
18	30/04/2003	1	W	C	■			■	■		
19	06/05/2003	1	S	S	■			■	■		
20	10/05/2003	1	W	M	■			■	■		
21	14/01/2004	9	W	M	■			■	■	■	
22	20/01/2004	10	NW	m	■	■	■	■	■	■	
23	16/02/2004	2	NE	P	■	■	■	■	■	■	
24	17/02/2004	4	N/NE	C/P	■	■	■	■	■	■	
25	08/03/2004	5	NE	P	■	■	■	■	■	■	
26	06/04/2004	5	W	HM	■	■	■	■	■	■	
27	22/04/2004	3	SW	S	■	■	■	■	■	■	
28	25/06/2004	2	W	HM	■	■	■	■	■	■	
29	08/07/2004	1	W	M	■	■	■	■	■	■	
30	23/09/2004	4	NW	M	■	■	■	■	■	■	■
31	17/11/2004	3	NW	M	■	■	■	■	■	■	■
32	16/12/2004	4	SW/W	M/C	■	■	■	■	■	■	■
33	19/01/2005	4	NW	HM	■	■	■	■	■	■	■
34	18/04/2005	3	SW	M	■	■	■		■		■
35	22/05/2005	6	SW/W	M	■	■	■			■	■
36	06/06/2005	5	W/NW	M	■	■	■		■		■
37	28/09/2005	4	NW	HM	■	■	■		■	■	■
38	03/10/2005	6	NE	M/C	■	■	■		■	■	■
39	18/10/2005	2	S	M/C	■	■	■		■	■	■
40	18/09/2006	2	W	M	■		■			■	■
41	04/10/2006	4	W	HM/M	■		■		■	■	■
42	13/12/2007	1	NE	C	■		■		■		■
43	17/01/2008	1	W	HM	■		■				■
44	29/02/2008	1	W	M	■		■		■		■
45	21/04/2008	1	SW	M	■		■		■		■
46	09/10/2008	1	SW	M	■						■
47	17/11/2008	1	N	M	■						■
48	01/12/2008	1	NW	HM	■		■			■	■
49	27/01/2009	1	N	M						■	■
50	09/02/2009	1	W	M			■		■	■	■
51	17/02/2009	1	NW	M	■		■		■	■	■
52	24/02/2009	1	N	C	■		■		■	■	■
53	30/03/2009	1	NE	P	■				■		■
54	03/11/2009	1	W	M	■	■	■	■	■	■	■
55	23/11/2009	1	W	M	■	■	■	■	■	■	■
56	08/01/2010	1	NE	C	■	■	■	■		■	■
57	26/01/2010	1	NE	P	■	■	■	■		■	■
58	19/02/2010	1	NW	HM							■
59	04/03/2010	1	NE	C	■	■	■	■		■	■
60	31/03/2010	1	W	HM	■	■	■	■		■	■
61	31/05/2010	1	NW	M	■	■		■	■	■	■
62	01/06/2010	1	W	M	■	■		■	■		■
63	08/06/2010	1	S	C	■	■		■	■		■
64	16/06/2010	1	NE	M	■	■		■	■	■	■
65	18/06/2010	1	NE	C	■	■		■	■	■	■
66	16/09/2010	1	W	M	■	■			■		■
67	19/01/2011	1	N	C	■	■	■	■	■	■	■
68	02/02/2011	1	W	M	■	■	■	■	■	■	■
69	24/02/2011	1	W	M	■	■	■	■	■	■	■
70	28/03/2011	1	SW	M	■	■	■	■	■	■	■
71	31/03/2011	1	W	M	■	■	■	■	■	■	■
72	12/04/2011	1	NW	HM	■	■	■	■	■	■	■
73	28/04/2011	1	N	P	■	■	■	■	■	■	■
	Total number of samples	**199**									

Table S1

		Location	puy de Dôme, France				Cheeka Peak Obs., USA	Mt. Broken, Germany	Rax, Austria	Whiteface Mountain, USA	Mount Schmücke, Germany
		Reference	This study				Vong et al., 1997	Acker et al., 1998	Löflund et al., 2002	Moore et al., 2004	Van Pinxteren et al., 2005
		Date	2001-2011				1993	1996	1999-2000	1998	2001-2002
		Types	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Clooud
		Categories	P	C	M	HM	M	C	C	P	C
		Av	4.87	9.90	6.15	11.21					
	H_2O_2	Min	1.90	1.03	0.10	0.80					
		Max	7.30	57.70	20.80	19.00					
		Av	2.7	1.1	0.4	0.7					
uM	Fe(II)	Min	1.1	0.1	0.1	0.3					
		Max	5.3	6.8	4.0	1.6					
		Av	0.6	0.5	0.4	0.2					
	Fe(III)	Min	BDL	BDL	BDL	BDL				0.27	
		Max	3.0	2.4	4.9	1.0				20.75	
		Av	17.74	10.97	4.94	12.02	2.90		15.49		
	Acetate	Min	3.42	BDL	0.31	1.80			4.00		1.60
		Max	50.74	41.58	22.24	57.61			37.80		41.40
		Av	5.53	13.46	6.32	13.01	6.40		13.25		
	Formate	Min	2.66	0.25	0.80	2.26			1.30		4.90
		Max	13.67	52.79	28.97	42.38			34.33		39.10
		Av	1.58	1.81	0.60	1.09			2.54		
	Succinate	Min	0.85	0.14	BDL	BDL			1.70		
		Max	2.31	7.60	3.50	4.48			5.08		1.80
		Av	0.98	1.62	0.74	1.10			3.84		
	Malonate	Min	0.71	0.34	0.31	0.32			1.54		0.40
		Max	1.24	7.00	2.86	3.88			5.76		1.80
		Av	4.13	4.92	2.07	3.61			8.44		
	Oxalate	Min	2.54	0.51	0.18	1.51			1.34		2.40
		Max	6.89	19.40	7.52	12.02			25.32		11.60
		Av	69.41	34.95	25.28	238.20	432.90	101	16.08		
	Cl^-	Min	3.85	4.76	0.49	81.37			0.85		
		Max	203.31	146.82	133.52	409.52			44.57		
		Av	416.65	110.88	24.84	59.30	15.10	360	136.45		
$\mu eq\ L^{-1}$	NO_3^-	Min	309.98	31.89	0.80	9.71			16.29	66.30	
		Max	516.51	297.86	93.17	231.80			319.35	1909.00	
		Av	119.77	97.93	28.27	79.30	63.50	312	163.40		
	SO_4^{2-}	Min	70.43	26.42	3.88	18.76			22.90	180.60	
		Max	170.98	218.40	77.24	261.64			423.40	2193.00	
		Av	44.38	34.43	25.75	286.31	290.60	100	15.69		
	Na^+	Min	0.96	5.64	0.37	110.76			2.62		
		Max	171.86	122.83	127.50	678.56			49.26		
		Av	233.06	145.07	43.23	88.45	11.10	472	229.57		
	NH_4^+	Min	151.01	72.69	5.99	28.55			29.46	103.10	
		Max	376.29	339.64	96.17	219.63			490.83	1124.40	
		Av	18.25	5.02	2.96	19.89	8.10	1.3	7.17		
	K^+	Min	2.15	BDL	BDL	5.23			1.28		
		Max	71.57	35.26	20.51	159.42			19.46		
		Av	7.62	13.18	7.82	46.92	66.40	24	21.40		
	Mg^{2+}	Min	0.68	0.00	0.00	8.43			1.64	2.90	
		Max	27.01	78.40	64.89	95.84			82.28	241.50	
		Av	105.11	29.03	17.17	59.40	36.10	51	21.48		
	Ca^{2+}	Min	0.82	BDL	BDL	5.46			4.00	7.40	
		Max	519.37	155.47	123.20	110.32			76.96	1124.00	
		Av	4.3	5.1	5.7	6.2	4.45		3.84		
	pH	Min	3.1	3.9	4.6	4.7			3.36	2.73	
		Max	6.6	7.1	7.6	6.9			5.26	3.77	
		Av	122.3	59.8	22.0	4.3					
mV	E_h	Min	35.0	-162.0	-164.0	-63.0					
		Max	230.0	133.0	166.0	50.0					
		Av	168.6	34.5	17.8	56.6					
$\mu S\ cm^{-1}$	C	Min	93.7	7.5	2.0	27.0					
		Max	348.0	74.0	155.0	134.0					
		Av	0.41	0.29	0.28	0.27	0.30	0.34	0.28		
$g\ m^{-3}$	LWC	Min	0.40	0.07	0.06	0.05			0.15		
		Max	0.41	0.92	0.58	0.48			0.59		
		Av	12.4	5.5	3.3	4.8					
$mgC\ L^{-1}$	TOC	Min	4.8	2.0	0.3	2.5					
		Max	25.0	15.5	15.5	10.3					

		Location	puy de Dôme, France				Daekwanreung, South Korea	Stog Izerski, Poland	Szrenica, Poland	East Peak, Puerto Rico				
		Reference	This study				Kim et al., 2006	Blas et al., 2008		Reyes-Rodriguez et al., 2009	Gioda et al., 2009		Gioda et al., 2011	
		Date	2001-2011				2002-2003	2003-2004		2007	2005	2005	2004-2007	2004-2007
		Types	Cloud	Cloud	Cloud	Cloud	Cloud/fog	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud
		Categories	P	C	M	HM	M/C	P	P	M	M	C	M	P
uM	H_2O_2	Av	**4.87**	**9.90**	**6.15**	**11.21**								
		Min	1.90	1.03	0.10	0.80								
		Max	7.30	57.70	20.80	19.00								
	Fe(II)	Av	**2.7**	**1.1**	**0.4**	**0.7**								
		Min	1.1	0.1	0.1	0.3								
		Max	5.3	6.8	4.0	1.6								
	Fe(III)	Av	**0.6**	**0.5**	**0.4**	**0.2**								
		Min	BDL	BDL	BDL	BDL								
		Max	3.0	2.4	4.9	1.0								
$\mu eq\ L^{-1}$	Acetate	Av	**17.74**	**10.97**	**4.94**	**12.02**							**3.80**	**11.60**
		Min	3.42	BDL	0.31	1.80								
		Max	50.74	41.58	22.24	57.61								
	Formate	Av	**5.53**	**13.46**	**6.32**	**13.01**							**3.00**	**2.00**
		Min	2.66	0.25	0.80	2.26								
		Max	13.67	52.79	28.97	42.38								
	Succinate	Av	**1.58**	**1.81**	**0.60**	**1.09**								
		Min	0.85	0.14	BDL	BDL								
		Max	2.31	7.60	3.50	4.48								
	Malonate	Av	**0.98**	**1.62**	**0.74**	**1.10**								
		Min	0.71	0.34	0.31	0.32								
		Max	1.24	7.00	2.86	3.88								
	Oxalate	Av	**4.13**	**4.92**	**2.07**	**3.61**							**1.40**	**7.40**
		Min	2.54	0.51	0.18	1.51								
		Max	6.89	19.40	7.52	12.02								
	Cl^-	Av	**69.41**	**34.95**	**25.28**	**238.20**	**166.10**	**65.60**	**96.70**	**384.00**	**446.14**	**395.84**	**473.00**	**388.00**
		Min	3.85	4.76	0.49	81.37	6.20			151.00	166.29	235.35		
		Max	203.31	146.82	133.52	409.52	668.00			841.00	681.76	556.32		
	NO_3^-	Av	**416.65**	**110.88**	**24.84**	**59.30**	**493.80**	**172.60**	**177.40**	**28.00**	**11.16**	**66.24**	**16.00**	**40.00**
		Min	309.98	31.89	0.80	9.71	7.20			19.00	2.88	46.92		
		Max	516.51	297.86	93.17	231.80	2729.30			37.00	29.05	85.55		
	SO_4^{2-}	Av	**119.77**	**97.93**	**28.27**	**79.30**	**489.30**	**133.40**	**136.60**	**68.00**	**54.57**	**79.25**	**54.00**	**89.00**
		Min	70.43	26.42	3.88	18.76	34.00			46.00	28.95	59.34		
		Max	170.98	218.40	77.24	261.64	1838.60			101.00	84.14	99.16		
	Na^+	Av	**44.38**	**34.43**	**25.75**	**286.31**	**166.20**	**67.30**	**100.90**	**362.00**	**476.76**	**428.67**	**532.00**	**370.00**
		Min	0.96	5.64	0.37	110.76	1.20			158.00	174.56	255.46		
		Max	171.86	122.83	127.50	678.56	532.80			738.00	685.00	601.88		
	NH_4^+	Av	**233.06**	**145.07**	**43.23**	**88.45**	**610.70**	**166.60**	**189.50**	**7.00**	**5.29**	**6.49**	**5.80**	**24.00**
		Min	151.01	72.69	5.99	28.55	55.80			1.00	3.37	6.05		
		Max	376.29	339.64	96.17	219.63	2078.40			13.00	7.45	6.93		
	K^+	Av	**18.25**	**5.02**	**2.96**	**19.89**	**28.40**	**5.70**	**20.70**	**56.00**	**10.39**	**8.72**	**30.00**	**20.00**
		Min	2.15	BDL	BDL	5.23	0.80			27.00	7.22	4.84		
		Max	71.57	35.26	20.51	159.42	86.10			116.00	14.67	12.60		
	Mg^{2+}	Av	**7.62**	**13.18**	**7.82**	**46.92**	**68.10**	**20.60**	**27.20**	**10.00**	**109.04**	**92.99**	**89.00**	**68.00**
		Min	0.68	0.00	0.00	8.43	0.80			6.00	39.73	54.15		
		Max	27.01	78.40	64.89	95.84	180.50			17.00	155.40	131.82		
	Ca^{2+}	Av	**105.11**	**29.03**	**17.17**	**59.40**	**197.60**	**52.40**	**64.80**	**50.00**	**24.88**	**22.16**	**60.00**	**23.00**
		Min	0.82	BDL	BDL	5.46	6.60			44.00	13.66	19.64		
		Max	519.37	155.47	123.20	110.32	761.80			61.00	33.59	24.68		
	pH	Av	**4.3**	**5.1**	**5.7**	**6.2**	**4.40**	**4.25**	**4.44**	**6.00**	**6.14**	**4.85**	**5.80**	**4.50**
		Min	3.1	3.9	4.6	4.7	0.70			5.15	5.80	4.76		
		Max	6.6	7.1	7.6	6.9	6.50			7.37	6.39	4.94		
mV	E_h	Av	**122.3**	**59.8**	**22.0**	**4.3**								
		Min	35.0	-162.0	-164.0	-63.0								
		Max	230.0	133.0	166.0	50.0								
$\mu S\ cm^{-1}$	C	Av	**168.6**	**34.5**	**17.8**	**56.6**	**149.00**	**66.00**	**69.00**					
		Min	93.7	7.5	2.0	27.0	13.50							
		Max	348.0	74.0	155.0	134.0	530.00							
$g\ m^{-3}$	LWC	Av	**0.41**	**0.29**	**0.28**	**0.27**				**0.14**	**0.35**	**0.09**	**0.20**	**0.10**
		Min	0.40	0.07	0.06	0.05				0.06	0.13	0.07		
		Max	0.41	0.92	0.58	0.48				0.28	0.60	0.11		
$mgC\ L^{-1}$	TOC	Av	**12.4**	**5.5**	**3.3**	**4.8**				**0.45**			**0.70**	**1.30**
		Min	4.8	2.0	0.3	2.5				0.15				
		Max	25.0	15.5	15.5	10.3				0.66				

Table S2

		TOC (mgC L^{-1})		
	Influence	Av	Min	Max
Various sites, US (Anastasio et al., 1994) *	Continental	12.0	3.0	18.0
Whiteface moutain, US (Khwaja et al., 1995) *	Continental	7.6	5.1	11.0
Whiteface moutain, US (Arakaki and Faust, 1998)	Continental		2.4	26.8
Rax mountain, Austria (Löflund et al., 2002)	Continental	5.7	1.0	14.0
Schmücke, Germany (Brüggemann et al., 2005)	Continental	5.8		
Arizona, US (Hutchings et al., 2009)	Continental	9.2	2.9	18.6
East Peak, Puerto Rico (Reyes-Rodríguez et al., 2009)	Marine	0.4	0.1	0.7
Mount Tai, China (Wang et al., 2011)	Polluted	18.5	1.8	153.1
Mount Tai (Desyaterik et al., 2013 & Herckes et al., 2013)	Highly polluted		100.0	200.0
Whistler mountain, US Fresno, US (Wang et al., 2013)	Continental Polluted		2.0 12.0	11.0 35.0
Raven's nest, US Whistler Peak, US (Ervens et al., 2013) *	Continental Continental		1.8 3.1	5.8 8.1
PdD moutain, France (Marinoni et al., 2004) *	Marine/Continental	3.6	1.2	15.5
This study, pdD moutain, France	Polluted Continental Marine Highly marine	12.4 5.5 3.3 4.8	4.8 2.0 0.3 2.5	25.0 15.5 15.5 10.3

*This value corresponds to the Dissolved Organic Carbon (DOC).

Table S3

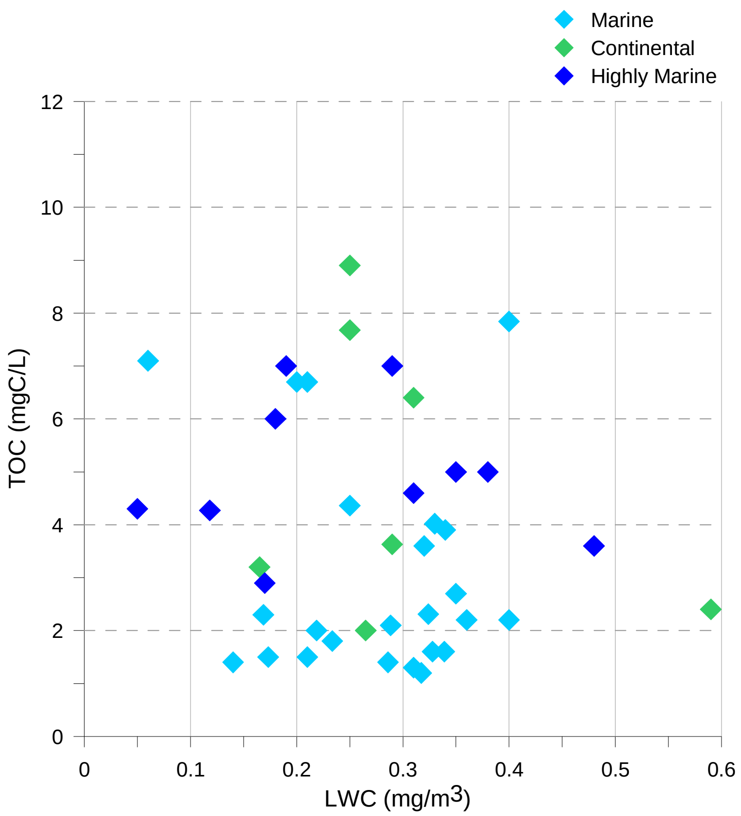
Marine
Continental
Highly Marine
TOC (mgC/L)
12
10
8
6
4
2
0
0
0.1
0.2
0.3
0.4
0.5
0.6
LWC (mg/m3)

Figure S1.

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