Supplementary material

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

L. Deguillaume[1,2], T. Charbouillot[1,2,3,4], M. Joly[1,2,3,4], M. Vaïtilingom[1,2,3,4], M. Parazols[1,2,3,4], A. Marinoni[1,2,†], P. Amato[3,4], A.-M. Delort[3,4], V. Vinatier[3,4], A. Flossmann[1,2], N. Chaumerliac[1,2], J.M. Pichon[1,2], S. Houdier[5,6], P. Laj[1,2,‡], K. Sellegri[1,2], A. Colomb[1,2], M. Brigante[3,4], G. Mailhot[3,4]

[1] Clermont Université, Université Blaise Pascal, OPGC, Laboratoire de Météorologie Physique, BP 10448, F-63000 Clermont-Ferrand (France)

[2] CNRS, UMR 6016, LaMP/OPGC, BP80026, F-63177 Aubière

[3] Clermont Université, Université Blaise Pascal, Institut de Chimie de Clermont-Ferrand, BP 10448, F-63000 Clermont-Ferrand (France)

[4] CNRS, UMR 6296, ICCF, BP 80026, F-63177 Aubière

[5] Université Joseph Fourier-Grenoble 1, Laboratoire de Glaciologie et Géophysique de l'Environnement, F-38041 Grenoble (France)

[6] CNRS, UMR 5183, F-38041 Grenoble

[†] Now at the Institute for Atmospheric Science and Climate (ISAC), CNR, Bologna, Italy

[‡] Now at Laboratoire de Glaciologie et Géophysique de l'Environnement (LGGE), CNRS/University of Grenoble, Grenoble, France

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				Black forest, Germany	Le Donon, Vosges, France	Cheeka Peak Obs., USA	Mt. Broken, Germany	Rax, Austria	Vosges Moutains, France	Whiteface Mountain, USA
		Reference	This study				Lammel and Metzig, 1991		Vong et al., 1997	Acker et al., 1998	Löflund et al., 2002	Herckes et al., 2002	Moore et al., 2004
		Date	2001-2011				1988	1990	1993	1996	1999-2000	1998	1998
		Types	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud
		Categories	P	C	M	HM	P	P	M	C	C	C	P
	H_2O_2	Av	**4.9**	**9.9**	**6.2**	**11.2**							
		Min	1.9	1.0	0.1	0.8							
		Max	7.3	57.7	20.8	19.0							
	Fe(II)	Av	**2.7**	**1.1**	**0.4**	**0.7**							
µM		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							0.3
		Max	3.0	2.4	4.9	1.0							20.8
	Acetate	Av	**17.7**	**11.0**	**4.9**	**12.0**			**2.9**		**15.5**		
		Min	3.4	BDL	0.3	1.8					4.0		
		Max	50.7	41.6	22.2	57.6					37.8		
	Formate	Av	**5.5**	**13.5**	**6.3**	**13.0**			**6.4**		**13.3**		
		Min	2.7	0.3	0.8	2.3					1.3		
		Max	13.7	52.8	29.0	42.4					34.3		
	Succinate	Av	**1.6**	**1.8**	**0.6**	**1.1**					**2.5**		
		Min	0.8	0.1	BDL	BDL					1.7		
		Max	2.3	7.6	3.5	4.5					5.1		
	Malonate	Av	**1.0**	**1.6**	**0.7**	**1.1**					**3.8**		
		Min	0.7	0.3	0.3	0.3					1.5		
		Max	1.2	7.0	2.9	3.9					5.8		
	Oxalate	Av	**4.1**	**4.9**	**2.1**	**3.6**					**8.4**		
		Min	2.5	0.5	0.2	1.5					1.3		
		Max	6.9	19.4	7.5	12.0					25.3		
	Cl^-	Av	**69.4**	**35.0**	**25.3**	**238.2**	**90**	**115**	**432.9**	**101.0**	**16.1**	**143.0**	
		Min	3.9	4.8	0.5	81.4	20	50			0.8	17.0	
		Max	203.3	146.8	133.5	409.5	220	170			44.6	1078.0	
	NO_3^-	Av	**416.6**	**110.9**	**24.8**	**59.3**	**400**	**380**	**15.1**	**360.0**	**136.5**	**181.0**	
µeq L^{-1}		Min	310.0	31.9	0.8	9.7	240	230			16.3	5.1	66.3
		Max	516.5	297.9	93.2	231.8	680	950			319.4	555.0	1909.0
	SO_4^{2-}	Av	**119.8**	**97.9**	**28.3**	**79.3**	**500**	**355**	**63.5**	**312.0**	**163.4**	**298.0**	
		Min	70.4	26.4	3.9	18.8	240	240			22.9	29.0	180.6
		Max	171.0	218.4	77.2	261.6	1290	900			423.4	764.0	2193.0
	Na^+	Av	**44.4**	**34.4**	**25.7**	**286.3**	**70**	**180**	**290.6**	**100.0**	**15.7**	**175.0**	
		Min	1.0	5.6	0.4	110.8	10	70			2.6	10.1	
		Max	171.9	122.8	127.5	678.6	190	230			49.3	1657.0	
	NH_4^+	Av	**233.1**	**145.1**	**43.2**	**88.4**	**830**	**330**	**11.1**	**472.0**	**229.6**	**276.0**	
		Min	151.0	72.7	6.0	28.6	410	210			29.5	4.9	103.1
		Max	376.3	339.6	96.2	219.6	1310	430			490.8	852.0	1124.4
	K^+	Av	**18.2**	**5.0**	**3.0**	**19.9**	**60**	**35**	**8.1**	**1.3**	**7.2**	**57.2**	
		Min	2.1	BDL	BDL	5.2	30	20			1.3	6.9	
		Max	71.6	35.3	20.5	159.4	110	60			19.5	540.0	
	Mg^{2+}	Av	**7.6**	**13.2**	**7.8**	**46.9**			**66.4**	**24.0**	**21.4**	**25.9**	
		Min	0.7	0.0	0.0	8.4					1.6	2.5	2.9
		Max	27.0	78.4	64.9	95.8					82.3	142.0	241.5
	Ca^{2+}	Av	**105.1**	**29.0**	**17.2**	**59.4**			**36.1**	**51.0**	**21.5**	**120.0**	
		Min	0.8	BDL	BDL	5.5					4.0	24.0	7.4
		Max	519.4	155.5	123.2	110.3					77.0	470.0	1124.0
	pH	Av	**4.3**	**5.1**	**5.7**	**6.2**	**4.1**	**3.4**	**4.5**		**3.8**	**4.8**	
		Min	3.1	3.9	4.6	4.7	4.0	2.8			3.4	3.9	2.7
		Max	6.6	7.1	7.6	6.9	4.9	4.7			5.3	6.6	3.8
	E_h	Av	**122**	**60**	**22**	**4**							
mV		Min	35	-162	-164	-63							
		Max	230	133	166	50							
	C	Av	**169**	**35**	**18**	**57**							
µS cm^{-1}		Min	94	8	2	27							
		Max	348	74	155	134							
	LWC	Av	**0.41**	**0.29**	**0.28**	**0.27**	**0.04**	**0.03**	**0.30**	**0.34**	**0.28**	**0.13**	
g m^{-3}		Min	0.40	0.07	0.06	0.05	0.01	0.01			0.15	0.04	
		Max	0.41	0.92	0.58	0.48	0.10	0.05			0.59	1.60	
	TOC	Av	**12.4**	**5.5**	**3.3**	**4.8**							
mgC L^{-1}		Min	4.8	2.0	0.3	2.5							
		Max	25.0	15.5	15.5	10.3							

		Location	puy de Dôme, France				Mount Schmücke, Germany	Daekwanreung, South Korea	Stog Izerski, Poland	Szrenica, Poland	East Peak, Puerto Rico					Whiteface moutain, USA
		Reference	This study				Van Pinxteren et al., 2005	Kim et al., 2006	Blas et al., 2008		Reyes-Rodriguez et al., 2009	Gioda et al., 2009		Gioda et al., 2011		Aleksic et al., 2009
		Date	2001-2011				2001-2002	2002-2003	2003-2004		2007	2005	2005	2004-2007	2004-2007	1994-2006
		Types	Cloud	Cloud	Cloud	Cloud	Clooud	Cloud/fog	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud	Cloud
		Categories	P	C	M	HM	C	M/C/P	P	P	M	M	C	M	P	P
µM	H_2O_2	Av	**4.9**	**9.9**	**6.2**	**11.2**										
		Min	1.9	1.0	0.1	0.8										
		Max	7.3	57.7	20.8	19.0										
	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										
µeq L^{-1}	Acetate	Av	**17.7**	**11.0**	**4.9**	**12.0**								**3.8**	**11.6**	
		Min	3.4	BDL	0.3	1.8	1.6									
		Max	50.7	41.6	22.2	57.6	41.4									
	Formate	Av	**5.5**	**13.5**	**6.3**	**13.0**								**3.0**	**2.0**	
		Min	2.7	0.3	0.8	2.3	4.9									
		Max	13.7	52.8	29.0	42.4	39.1									
	Succinate	Av	**1.6**	**1.8**	**0.6**	**1.1**										
		Min	0.8	0.1	BDL	BDL										
		Max	2.3	7.6	3.5	4.5	1.8									
	Malonate	Av	**1.0**	**1.6**	**0.7**	**1.1**										
		Min	0.7	0.3	0.3	0.3	0.4									
		Max	1.2	7.0	2.9	3.9	1.8									
	Oxalate	Av	**4.1**	**4.9**	**2.1**	**3.6**								**1.4**	**7.4**	
		Min	2.5	0.5	0.2	1.5	2.4									
		Max	6.9	19.4	7.5	12.0	11.6									
	Cl^-	Av	**69.4**	**35.0**	**25.3**	**238.2**		**166.1**	**65.6**	**96.7**	**384.0**	**446.1**	**395.8**	**473.0**	**388.0**	**5.6**
		Min	3.9	4.8	0.5	81.4		6.2			151.0	166.3	235.4			1.0
		Max	203.3	146.8	133.5	409.5		668.0			841.0	681.8	556.3			7.7
	NO_3^-	Av	**416.6**	**110.9**	**24.8**	**59.3**		**493.8**	**172.6**	**177.4**	**28.0**	**11.2**	**66.2**	**16.0**	**40.0**	**79.9**
		Min	310.0	31.9	0.8	9.7		7.2			19.0	2.9	46.9			39.4
		Max	516.5	297.9	93.2	231.8		2729.3			37.0	29.1	85.6			99.1
	SO_4^{2-}	Av	**119.8**	**97.9**	**28.3**	**79.3**		**489.3**	**133.4**	**136.6**	**68.0**	**54.6**	**79.3**	**54.0**	**89.0**	**200.0**
		Min	70.4	26.4	3.9	18.8		34.0			46.0	29.0	59.3			116.0
		Max	171.0	218.4	77.2	261.6		1838.6			101.0	84.1	99.2			293.8
	Na^+	Av	**44.4**	**34.4**	**25.7**	**286.3**		**166.2**	**67.3**	**100.9**	**362.0**	**476.8**	**428.7**	**532.0**	**370.0**	**3.1**
		Min	1.0	5.6	0.4	110.8		1.2			158.0	174.6	255.5			1.2
		Max	171.9	122.8	127.5	678.6		532.8			738.0	685.0	601.9			6.0
	NH_4^+	Av	**233.1**	**145.1**	**43.2**	**88.4**		**610.7**	**166.6**	**189.5**	**7.0**	**5.3**	**6.5**	**5.8**	**24.0**	**117.4**
		Min	151.0	72.7	6.0	28.6		55.8			1.0	3.4	6.1			66.6
		Max	376.3	339.6	96.2	219.6		2078.4			13.0	7.5	6.9			175.1
	K^+	Av	**18.2**	**5.0**	**3.0**	**19.9**		**28.4**	**5.7**	**20.7**	**56.0**	**10.4**	**8.7**	**30.0**	**20.0**	**1.6**
		Min	2.1	BDL	BDL	5.2		0.8			27.0	7.2	4.8			0.8
		Max	71.6	35.3	20.5	159.4		86.1			116.0	14.7	12.6			2.8
	Mg^{2+}	Av	**7.6**	**13.2**	**7.8**	**46.9**		**68.1**	**20.6**	**27.2**	**10.0**	**109.0**	**93.0**	**89.0**	**68.0**	**5.0**
		Min	0.7	0.0	0.0	8.4		0.8			6.0	39.7	54.2			2.2
		Max	27.0	78.4	64.9	95.8		180.5			17.0	155.4	131.8			10.3
	Ca^{2+}	Av	**105.1**	**29.0**	**17.2**	**59.4**		**197.6**	**52.4**	**64.8**	**50.0**	**24.9**	**22.2**	**60.0**	**23.0**	**17.8**
		Min	0.8	BDL	BDL	5.5		6.6			44.0	13.7	19.6			4.5
		Max	519.4	155.5	123.2	110.3		761.8			61.0	33.6	24.7			40.7
	pH	Av	**4.3**	**5.1**	**5.7**	**6.2**		**4.4**	**4.3**	**4.4**	**6.0**	**6.1**	**4.9**	**5.8**	**4.5**	**3.8**
		Min	3.1	3.9	4.6	4.7		0.7			5.2	5.8	4.8			3.7
		Max	6.6	7.1	7.6	6.9		6.5			7.4	6.4	4.9			4.0
mV	E_h	Av	**122**	**60**	**22**	**4**										
		Min	35	-162	-164	-63										
		Max	230	133	166	50										
µS cm^{-1}	C	Av	**169**	**35**	**18**	**57**		**149**	**66**	**69**						**80**
		Min	94	8	2	27		14								43
		Max	348	74	155	134		530								121
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**					**0.5**			**0.7**	**1.3**	
		Min	4.8	2.0	0.3	2.5					0.2					
		Max	25.0	15.5	15.5	10.3					0.7					

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

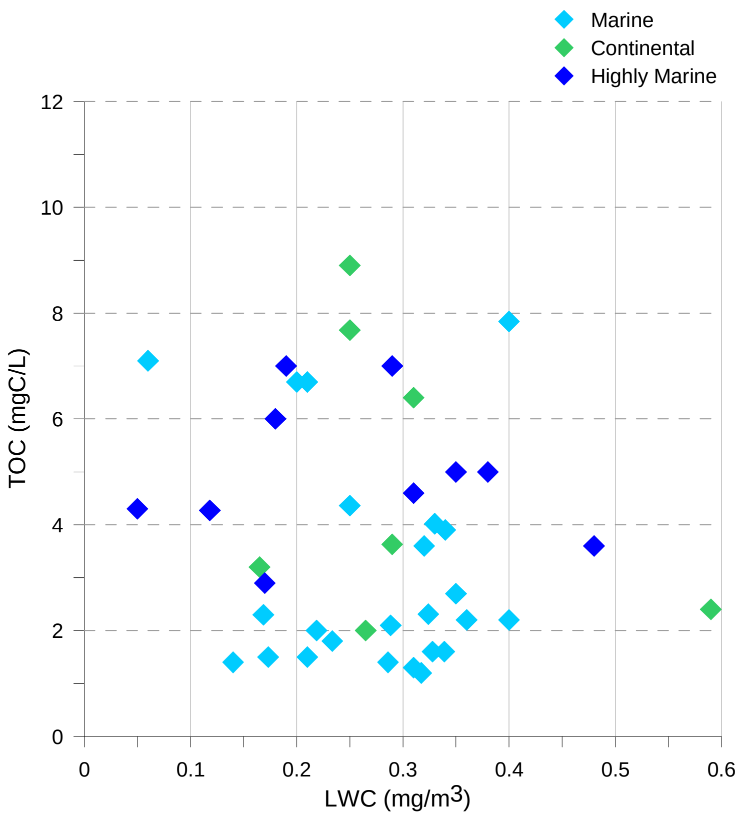
Marine
Continental
Highly Marine
12
10
8
6
4
2
0
TOC (mgC/L)
0
0.1
0.2
0.3
0.4
0.5
0.6
LWC (mg/m3)

Figure S1.

References

Acker, K., Möller, D., Wieprecht, W., Kalaß, D., and Auel, R.: Investigations of ground-based clouds at the Mt. Brocken, Fresenius' Journal of Analytical Chemistry, 361, 59-64, 10.1007/s002160050834, 1998.

Aleksic, N., Roya, K., Sistla, G., Dukett, J., Houck, N., and P. Casson: Analysis of cloud and precipitation chemistry at Whiteface Mountain, NY, Atmospheric Environment, 43, 2709-2716, 10.1016/j.atmosenv.2009.02.053, 2009.

Anastasio, C., Faust, B. C., and Allen, J. M.: Aqueous phase photochemical formation of hydrogen peroxide in authentic cloud waters, Journal of Geophysical Research, 99, 8231-8248, 10.1029/94jd00085, 1994.

Arakaki, T., and Faust, B. C.: Sources, sinks, and mechanisms of hydroxyl radical (OH) photoproduction and consumption in authentic acidic continental cloud waters from Whiteface Mountain, New York: The role of the Fe(r) (r = II, III) photochemical cycle, Journal of Geophysical Research, 103, 3487-3504, 10.1029/97jd02795, 1998.

Blas, M., Sobik, M., and Twarowski, R.: Changes of cloud water chemical composition in the Western Sudety Mountains, Poland, Atmospheric Research, 87, 224-231, 10.1016/j.atmosres.2007.11.004, 2008.

Deguillaume, L., Leriche, M., Monod, A., and Chaumerliac, N.: The role of transition metal ions on HO_x radicals in clouds: a numerical evaluation of its impact on multiphase chemistry, Atmospheric Chemistry and Physics, 4, 95-110, 10.5194/acp-4-95-2004, 2004.

Desyaterik, Y., Sun, Y., Shen, X., Lee, T., Wang, X., Wang, T., and Collett, J. L.: Speciation of “brown” carbon in cloud water impacted by agricultural biomass burning in eastern China, Journal of Geophysical Research: Atmospheres, 10.1002/jgrd.50561, 2013.

Ervens, B., Wang, Y., Eagar, J., Leaitch, W. R., Macdonald, A. M., Valsaraj, K. T., and Herckes, P.: Dissolved organic carbon (DOC) and select aldehydes in cloud and fog water: the role of the aqueous phase in impacting trace gas budgets, Atmospheric Chemistry and Physics, 13, 5117-5135, 10.5194/acp-13-5117-2013, 2013.

Freney, E. J., Sellegri, K., Canonaco, F., Boulon, J., Hervo, M., Weigel, R., Pichon, J. M., Colomb, A., Prévôt, A. S. H., and Laj, P.: Seasonal variations in aerosol particle composition at the puy de Dôme research station in France, Atmospheric Chemistry and Physics, 11, 13047-13059, 10.5194/acp-11-13047-2011, 2011.

Gelencsér, A., May, B., Simpson, D., Sánchez-Ochoa, A., Kasper-Giebl, A., Puxbaum, H., Caseiro, A., Pio, C., and Legrand, M.: Source apportionment of PM2.5 organic aerosol over Europe: Primary/secondary, natural/anthropogenic, and fossil/biogenic origin, Journal of Geophysical Research, 112, D23S04, 10.1029/2006jd008094, 2007.

Gioda, A., Mayol-Bracero, O., Morales-García, F., Collett, J., Decesari, S., Emblico, L., Facchini, M., Morales-De Jesús, R., Mertes, S., Borrmann, S., Walter, S., and Schneider, J.: Chemical composition of cloud water in the Puerto Rican tropical trade wind cumuli, Water Air and Soil Pollution, 200, 3-14, 10.1007/s11270-008-9888-4, 2009.

Gioda, A., Reyes-Rodríguez, G. J., Santos-Figueroa, G., Collett, J. L., Jr., Decesari, S., Ramos, M. d. C. K. V., Bezerra Netto, H. J. C., de Aquino Neto, F. R., and Mayol-Bracero, O. L.: Speciation of water-soluble inorganic, organic, and total nitrogen in a background marine environment: Cloud water, rainwater, and aerosol particles, Journal of Geophysical Research, 116, D05203, 10.1029/2010jd015010, 2011.

Herckes, P., Wendling, R., Sauret, N., Mirabel, Ph., and Wortham, H.: Cloudwater studies at a high elevation site in the Vosges Mountains (France), Environmental Pollution, 117, 169-177, 10.1016/S0048-9697(02)00037-2, 2002.

Herckes, P., Valsaraj, K. T., and Collett Jr, J. L.: A review of observations of organic matter in fogs and clouds: Origin, processing and fate, Atmospheric Research, 132-133, 434-449, 10.1016/j.atmosres.2013.06.005, 2013.

Hutchings, J., Robinson, M., McIlwraith, H., Triplett Kingston, J., and Herckes, P.: The chemistry of intercepted clouds in Northern Arizona during the North American monsoon season, Water Air and Soil Pollution, 199, 191-202, 10.1007/s11270-008-9871-0, 2009.

Khwaja, H. A.: Atmospheric concentrations of carboxylic acids and related compounds at a semiurban site, Atmospheric Environment, 29, 127-139, 10.1016/1352-2310(94)00211-3, 1995.

Kim, M.-G., Lee, B.-K., and Kim, H.-J.: Cloud/fog water chemistry at a high elevation site in South Korea, Journal of Atmospheric Chemistry, 55, 13-29, 10.1007/s10874-005-9004-8, 2006.

Lammel, G., and Metzig, G.: Multiphase chemistry of orographic clouds: Observations at subalpine mountain stations, Fresenius' Journal of Analytical Chemistry, 340, 564-574, 1991.

Leriche, M., Chaumerliac, N., and Monod, A.: Coupling quasi-spectral microphysics with multiphase chemistry: a case study of a polluted air mass at the top of the puy de Dôme mountain (France), Atmospheric Environment, 35, 5411-5423, 10.1016/S1352-2310(01)00300-4, 2001.

Leriche, M., Curier, R., Deguillaume, L., Caro, D., Sellegri, K., and Chaumerliac, N.: Numerical quantification of sources and phase partitioning of chemical species in cloud: application to wintertime anthropogenic air masses at the puy de Dôme station, Journal of Atmospheric Chemistry, 57, 281-297, 10.1007/s10874-007-9073-y, 2007.

Löflund, M., Kasper-Giebl, A., Schuster, B., Giebl, H., Hitzenberger, R., and Puxbaum, H.: Formic, acetic, oxalic, malonic and succinic acid concentrations and their contribution to organic carbon in cloud water, Atmospheric Environment, 36, 1553-1558, 10.1016/S1352-2310(01)00573-8, 2002.

Marinoni, A., Laj, P., Sellegri, K., and Mailhot, G.: Cloud chemistry at the puy de Dôme: variability and relationships with environmental factors, Atmospheric Chemistry and Physics, 4, 715-728, 10.5194/acp-4-715-2004, 2004.

Marinoni, A., Parazols, M., Brigante, M., Deguillaume, L., Amato, P., Delort, A.-M., Laj, P., and Mailhot, G.: Hydrogen peroxide in natural cloud water: Sources and photoreactivity, Atmospheric Research, 101, 256-263, 10.1016/j.atmosres.2011.02.013, 2011.

Moore, K. F., Sherman, D. E., Reilly, J. E., and Collett, J. L.: Drop size-dependent chemical composition in clouds and fogs. Part I. Observations, Atmospheric Environment, 38, 1389-1402, 10.1016/j.atmosenv.2003.12.013, 2004.

Parazols, M., Marinoni, A., Amato, P., Abida, O., Laj, P., and Mailhot, G.: Speciation and role of iron in cloud droplets at the puy de Dôme station, Journal of Atmospheric Chemistry, 54, 267-281, 10.1007/s10874-007-9069-7, 2006.

Pio, C. A., Legrand, M., Oliveira, T., Afonso, J., Santos, C., Caseiro, A., Fialho, P., Barata, F., Puxbaum, H., Sanchez-Ochoa, A., Kasper-Giebl, A., Gelencsér, A., Preunkert, S., and Schock, M.: Climatology of aerosol composition (organic versus inorganic) at nonurban sites on a west-east transect across Europe, Journal of Geophysical Research, 112, D23S02, 10.1029/2006jd008038, 2007.

Reyes-Rodríguez, G. J., Gioda, A., Mayol-Bracero, O. L., and Collett Jr, J.: Organic carbon, total nitrogen, and water-soluble ions in clouds from a tropical montane cloud forest in Puerto Rico, Atmospheric Environment, 43, 4171-4177, 10.1016/j.atmosenv.2009.05.049, 2009.

Sellegri, K., Laj, P., Marinoni, A., Dupuy, R., Legrand, M., and Preunkert, S.: Contribution of gaseous and particulate species to droplet solute composition at the puy de Dôme, France, Atmospheric Chemistry and Physics, 3, 1509-1522, 10.5194/acp-3-1509-2003, 2003a.

Sellegri, K., Laj, P., Peron, F., Dupuy, R., Legrand, M., Preunkert, S., Putaud, J. P., Cachier, H., and Ghermandi, G.: Mass balance of free tropospheric aerosol at the puy de Dôme (France) in winter, Journal of Geophysical Research, 108, 4333, 10.1029/2002jd002747, 2003b.

Venzac, H., Sellegri, K., Villani, P., Picard, D., and Laj, P.: Seasonal variation of aerosol size distributions in the free troposphere and residual layer at the puy de Dôme station, France, Atmospheric Chemistry and Physics, 9, 1465-1478, 10.5194/acp-9-1465-2009, 2009.

Voisin, D., Legrand, M., Chaumerliac, and N.: Scavenging of acidic gases (HCOOH, CH_3COOH, HNO_3, HCl, and SO_2) and ammonia in mixed liquid-solid water clouds at the puy de Dôme mountain (France), Journal of Geophysical Research, 105, 6817-6835, 10.1029/1999JD900983, 2000.

Vong, R. J., Baker, B. M., Brechtel, F. J., Collier, R. T., Harris, J. M., Kowalski, A. S., McDonald, N. C., and McInnes, L. M.: Ionic and trace element composition of cloud water collected on the Olympic Peninsula of Washington State, Atmospheric Environment, 31, 1991-2001, 10.1016/S1352-2310(96)00337-8, 1997.

Wang, Y., Chiu, C.-A., Westerhoff, P., Valsaraj, K. T., and Herckes, P.: Characterization of atmospheric organic matter using size-exclusion chromatography with inline organic carbon detection, Atmospheric Environment, 68, 326-332, 10.1016/j.atmosenv.2012.11.049, 2013.

Wang, Y., Guo, J., Wang, T., Ding, A., Gao, J., Zhou, Y., Collett Jr, J. L., and Wang, W.: Influence of regional pollution and sandstorms on the chemical composition of cloud/fog at the summit of Mt. Taishan in northern China, Atmospheric Research, 99, 434-442, 10.1016/j.atmosres.2010.11.010, 2011.