Soluble and Insoluble Pollutants in Fog and Rime Water Samples

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Abstract: Fog and rime water samples were collected at the meteorological observatory Milešovka in February and June 2006. In the samples, the soluble and insoluble pollutant concentrations were evaluated separately and the differences between the fog and rime water samples were studied. The comparison of the fog and rime water samples indicates that the mean soluble component concentrations in the air appear to be higher during the rime events than during the fog events at Milešovka. We recorded a larger mean particle size of the insoluble compounds in the fog water samples than in those of rime water. Some elements contained in the insoluble particles like Ca, Cl, C, Cu, Ag, were present largely in fog whole others, like Fe, Al, Si, Ti, prevailed in rime. In addition to the overall evaluation, the backward air trajectories were determined for each fog/rime event and the concentrations are presented as depending on the direction of the air particle transfer.

Keywords: fog water; rime water; pollutant concentration

The fog/rime water is one of the components of the wet deposition that enter the catchments. Fog/rime water contains both soluble and insoluble components. The latter ones deposit in soil. Some of the insoluble chemical compounds can become soluble under suitable conditions and can be gradually washed out.

The interactions of fogs and clouds with the air pollution have been studied at various locations around the world (e.g., Facchini *et al*. 1992; Wobrock *et al.* 1994; Minďáš & Škvarenina 1995, 2001; Borys *et al*. 2000; Fišák & Řezáčová 2001; Škvarenina & Minďáš 2001; Acker *et al*. 2002; Fišák *et al.* 2002; Fuzzi *et al.* 2002; Herckes *et al.* 2007; Igawa *et al.* 2002; Aikawa *et al*. 2005).

In our study, fog and rime water samples were collected in February and June 2006 at the meteorological observatory Milešovka, which is a part of the IAP ASCR. In total, 6 rime water- and 5 fog water-samples were obtained. Prior to chemical analyses, the samples were filtered using nitrocellulose membrane filters in order to separate the insoluble components (IC). The liquid samples and the dry filters with the stuck IC were studied separately. The liquid samples were analysed in the laboratory of the Czech Geological Survey, the conductivity and pH values were determined directly at the sampling site. The dry filters were analysed in the laboratory of the IPC BAS.

Supported by the Czech Science Foundation, Grant No. 205/09/1918, by the AS CR, Project No. 1QS200420562, and by the IRP, Project No. AV0Z30420517.

We attempted at finding the differences between the concentrations of soluble and insoluble pollutants, and between the fog and rime water samples. In addition to the overall evaluation, backward air trajectories were determined for each event in order to study the concentrations as depending on the direction of the air particle transfer.

Site description

The Milešovka Mt. (50°33'17"N, 13°55'57"E, 837 m a.s.l.) is located in the immediate vicinity of the north Bohemian brown coalfield. The coalfield region belongs to the most polluted areas in the Czech Republic (CR). Several thermal power stations are situated close to the massif of the Krušné hory Mts that forms a north-western border line of the coalfield. Chemical industry is concentrated in this area and large conurbations (Ústí nad Labem, Most, Litvínov, Chomutov, Teplice, Litoměřice, Lovosice, Prague) can also contribute to the air pollution in the Milešovka region.

From the geological viewpoint, the close Milešovka surroundings are formed primarily by igneous rocks with claystone, coal, and calcite fillings in several places. The mountain top itself consists of phonolite.

Methods and fog/rime water samples collection

In this paper, we deal with the concentrations of the selected components that were identified in the fog and rime water samples. It is well known that the most frequent rime development occurs during the fog events at the temperature bellow 0°C. In this paper, we will use the expression fog water in the case that the liquid water sample is collected at a temperature above 0°C.

Even if the rime originated in fog, the rime water samples were obtained after melting the solid rime that was collected by a passive collector. The fog water samples were collected by an active collector described e.g. by Daube *et al.* (1987) and Tesař *et al.* (1995).

The liquid water content (LWC) values were calculated from horizontal visibility values (VIS) by the relationship:

$$LWC = 0.0152 \times VIS^{-0.8582} \quad (1)$$

Where VIS marks the horizontal visibility as measured by the PWD 21 device (Present Weather Detector). Relationship (1) was derived by regression from the Milešovka data (Fišák *et al.* 2006).

The fog/rime water samples were filtered before chemical analysis. We used nitrocellulose filters with the pore size of 0.45 μm. The filters were dried up and covered with a thin carbon layer. They were analysed using JEOL JSM-6390 with EDS of Oxford Instruments. About 50–60 particles in the fog samples and 80–120 particles in the rime samples were chosen taking into account their sizes, shapes, and positions in the sample area. They were investigated with respect to their sizes, shapes and chemical compositions.

The transport direction as characterised by the transfer sector was determined for each fog/rime event. Because of the limited number of samples, we considered only the four main sectors N (NW–NE), E (NE–SE), S (SE–SW), and W (SW–NW) in the evaluation of the transfer direction. In order to classify the samples by sectors, we used the backward trajectory. We applied the NOAA HYSPLIT model to calculate the backward trajectory for each sample (Draxler & Rolph 2003; Rolph 2003; http://www.arl.noaa.gov/ready.html). The trajectories were calculated for a period of 24 h, the trajectory ending time corresponding to the middle of the sampling time period.

Table 1 brings the overview of the fog/rime samples. The volume of samples, start and end times of sampling, LWC, and sector of transfer are shown in Table 1. The table shows that the sector N occurs three times in the case of rime, the other sectors occurring only once. The sectors N and W occur two times, and the S sector once in the case of fog. The transport direction from the E sector was not registered with fog.

RESULTS AND DISCUSION

Soluble pollutants

In this subsection, the soluble pollutant concentrations are presented as related to the fog/rime samples, and the pollutant concentrations in fog/rime water as related to the air volume. The knowledge of the sample concentrations is important if we assess the impact on the vegetation cover, and the knowledge of the pollutant concentration in the air is relevant for the deposition calculation and the check of the relationship to the hygienic limits.

Table 2 shows the results of chemical analysis of the selected soluble pollutants (SP) in the fog/rime water samples. The table shows that the mean SP concentration values in rime water are higher than the corresponding values in fog water with the exception of NH_4^+ a NO_3^-. Rime water also shows a higher conductivity. In addition, rime water had a lover pH than fog water.

Essential information on the pollutant amounts in the air can be obtained by taking into account the fog LWC. Table 3 shows the concentrations of the selected soluble components (SC) in fog/rime water in the air calculated from the SP values and the corresponding mean LWC. Obviously, the mean SC concentrations in fog/rime water in the air were higher during the rime events than in the fog events except NH_4^+, NO_3^-, and SO_4^{2-}. We can also observe that the mean fog/rime Na^+, Mg^{2+}, and Cl^- concentrations, were close one another in the air during fog and rime events.

Table 1. An overview of samples (LWC – liquid water content, UTC – universal time coordinated)

Type	Designation	Volume (ml)	LWC (g/m^3)	Sampling start (UTC)	Sampling end (UTC)	Transport direction (sector)
Rime	R-06	70	0.036	9. 2. 06 19:40	11. 2. 06 09:00	N
	R-07	85	0.051	15. 2. 06 00:00	16. 2. 06 10:00	S
	R-08	85	0.095	20. 2. 06 20:30	24. 2. 06 09:00	N
	R-09	95	0.132	9. 3. 06 04:30	9. 3. 06 14:00	W
	R-10	70	0.068	11. 3. 06 15:30	12. 3. 06 15:45	N
	R-11	110	0.061	12. 3. 06 15:45	15. 3. 06 12:00	E
Fog	F-01	100	0.130	3. 5. 06 03:30	3. 5. 06 08:30	S
	F-02	100	0.100	26. 5. 06 18:15	27. 5. 06 00:00	W
	F-03	95	0.079	3. 6. 06 21:15	4. 6. 06 05:30	N
	F-04	90	0.107	17. 6. 06 01:15	17. 6. 06 08:50	W
	F-05	80	0.062	30. 6. 06 02:20	30. 6. 06 10:20	N

Table 2. Soluble pollutant concentrations in fog/rime water samples

Sample	NH_4^+	Na^+	Mg^{2+}	K^+	Ca^{2+}	Mn^{2+}	NO_3^-	SO_4^{2-}	Cl^-	pH	Cond.
					(mg/l)					(–)	(µS/cm)
R-06	12.90	1.46	0.27	0.76	1.08	20.0	25.85	20.03	2.06	4.2	133.0
R-07	26.28	7.97	1.35	1.72	4.99	70.0	74.32	45.98	10.71	3.6	381.0
R-08	9.87	0.43	0.12	0.67	0.70	22.0	15.66	16.40	0.70	4.3	104.6
R-09	6.75	0.25	0.05	0.37	0.34	6.0	11.92	10.51	0.29	4.3	80.1
R-10	8.21	0.58	0.13	0.44	1.95	18.0	19.19	22.76	0.79	3.7	173.0
R-11	15.50	2.68	0.19	1.59	2.47	234.0	27.77	48.40	3.79	3.5	272.0
Mean	13.25	2.23	0.35	0.93	1.92	61.7	29.12	27.35	3.06	3.8	190.6
F-01	24.49	1.66	0.32	0.63	1.50	68.0	50.88	32.17	3.66	4.7	214.0
F-02	7.45	1.48	0.23	0.61	0.80	19.0	15.93	15.62	1.95	4.0	119.5
F-03	14.05	2.37	0.32	0.47	0.71	13.0	29.88	21.72	2.83	3.8	184.0
F-04	18.39	1.58	0.33	1.00	1.03	33.0	35.84	25.50	1.32	4.2	176.0
F-05	9.88	0.41	0.08	0.32	0.46	12.0	13.54	16.30	0.67	4.4	93.1
Mean	14.85	1.50	0.26	0.61	0.90	29.0	29.21	22.26	2.09	4.1	157.3

Table 3. Soluble pollutant concentrations in fog/rime water in the air (LWC – liquid water content)

Sample	LWC	NH_4^+	Na^+	Mg^{2+}	K^+	Ca^{2+}	Mn^{2+}	NO_3^-	SO_4^{2-}	Cl^-	pH	Cond.
	(g/m^3)			($\mu g/m^3$)			(ng/m^3)		($\mu g/m^3$)		(–)	($\mu S/cm$)
R-06	0.036	0.464	0.053	0.010	0.027	0.039	0.720	0.931	0.721	0.074	4.2	133.0
R-07	0.051	1.341	0.406	0.069	0.088	0.254	3.570	3.790	2.345	0.546	3.6	381.0
R-08	0.095	0.938	0.041	0.011	0.064	0.066	2.090	1.488	1.558	0.066	4.3	104.6
R-09	0.132	0.891	0.033	0.007	0.049	0.045	0.792	1.573	1.387	0.038	4.3	80.1
R-10	0.068	0.558	0.039	0.009	0.030	0.133	1.224	1.305	1.548	0.054	3.7	173.0
R-11	0.061	0.946	0.163	0.012	0.097	0.151	14.274	1.694	2.952	0.231	3.5	272.0
Mean	0.074	0.979	0.165	0.026	0.068	0.142	4.553	2.150	2.019	0.226	3.8	190.6
F-01	0.130	3.184	0.216	0.042	0.082	0.195	8.840	6.614	4.182	0.476	4.7	214.0
F-02	0.100	0.745	0.148	0.023	0.061	0.080	1.900	1.593	1.562	0.195	4.0	119.5
F-03	0.079	1.110	0.187	0.025	0.037	0.056	1.027	2.361	1.716	0.224	3.8	184.0
F-04	0.107	1.968	0.169	0.035	0.107	0.110	3.531	3.835	2.728	0.142	4.2	176.0
F-05	0.062	0.613	0.025	0.005	0.020	0.029	0.744	0.839	1.010	0.041	4.4	93.1
Mean	0.096	1.420	0.143	0.024	0.058	0.086	2.772	2.793	2.128	0.200	4.1	157.3

We compared SC concentrations in fog/rime in the air as obtained for different transport sectors. The samples were separated according to the transport sectors which were computed by NOAA HYSPLIT model. If more samples were collected in one sector, we computed the mean SC concentration. The results are summarised in Table 4. From Table 4 is it apparent that the mean SC concentrations in the air and in the transport sectors were higher in fog than in rime with the exception of the sector S. However, the total mean SC concentrations in the air were higher in rime than in fog (Table 3). This indicates the importance of the transfer from the sector S.

Maximum fog/rime SC concentrations were recorded at the air transfer from the S sector. It is valid for both fog and rime events (see Table 4). Minimum rime SC concentrations were recorded in the air transported the sectors N and W. All minima of SC concentrations in fog were found in the air flow from the N sector. The air with high SC concentrations in rime was also transported from the sector E. It is presumable that similar results can be reached with fog. However, no sample was collected from sector E. The concentrations of NH_4^+, NO_3^-, and SO_4^{2-} in the air in fogs were higher in all transport sectors (Table 4). The concentration of Ca^{2+} was higher in the rime- than in the fog samples in all transport sectors except the sector W (Table 4).

Insoluble pollutants

Chemical composition of the insoluble pollutants (IP) in fog/rime water was determined statistically. Maximum, minimum, and the mean IP concentrations relative to the amount in wt% were also determined statistically. The results are presented in Tables 5 and 6. The mean relative IP concentrations in the transport sectors are given in Table 7. The mean particle size is indicated in Tables 5 and 7. The insoluble particles in fog water reached a higher size than those in rime water as can be see in Table 5. The mean size of the insoluble particles was the highest in the air flow from the transport sector S. IP contained primary chemical compounds of phonolite. Tables 5, 6, and 7 relate to the components with wt% higher than 0.1 in at least one sample type (fog or rime). Apart from the components of phonolite, which prevails in the vicinity of the sampling place, the occurrence of Ag, Cr, Gd, and Tl was recorded. The metals, which do not originate in phonolite, were present only in trace amounts and are probably of anthropogenic origin. Table 6 shows the concentrations of IP in the air, which can give better evidence for respecting the hygienic limits.

Table 4. Soluble pollutant concentrations in fog/rime water samples and distributed according to the transport sector

Sector		NH_4^+	Na^+	Mg^{2+}	K^+	Ca^{2+}	Mn^{2+}	NO_3^-	SO_4^{2-}	Cl^-	pH	Cond.
		($\mu g/m^3$)					(ng/m^3)	($\mu g/m^3$)			(–)	($\mu S/cm$)
Rime	N	0.764	0.042	0.009	0.047	0.050	1.201	1.331	1.222	0.060	4.0	105.9
	E	0.946	0.163	0.012	0.097	0.151	14.274	1.694	2.952	0.231	3.5	272.0
	S	1.341	0.406	0.069	0.088	0.254	3.570	3.790	2.345	0.546	3.6	381.0
	W	0.891	0.033	0.007	0.049	0.045	0.792	1.573	1.387	0.038	4.3	80.1
Fog	N	0.861	0.106	0.015	0.028	0.042	0.886	1.600	1.363	0.133	4.0	138.5
	E	–	–	–	–	–	–	–	–	–	–	–
	S	3.184	0.216	0.042	0.082	0.195	8.840	6.614	4.182	0.476	4.7	214.0
	W	1.356	0.159	0.029	0.084	0.095	2.716	2.714	2.145	0.168	4.1	147.8

Table 5. Relative insoluble pollutant concentrations in fog/rime water (in wt%); the MS marks the particle mean size (in μm)

Element	Rime			Fog		
	mean	max	min	mean	max	min
C	46.94	48.43	46.27	51.40	57.79	46.19
N				0.29	0.29	0.29
O	42.26	44.40	40.20	41.05	44.26	38.45
Na	0.11	0.19	0.05	0.24	0.50	0.08
Mg	0.14	0.27	0.09	0.11	0.29	0.02
Al	2.15	3.27	1.26	1.54	2.34	0.19
Si	4.20	4.93	3.60	2.36	4.80	0.71
S	0.09	0.28	0.02	0.17	0.29	0.08
Cl	0.02	0.06	0.00	0.22	0.73	0.01
K	0.21	0.35	0.13	0.18	0.40	0.04
Ca	0.17	0.32	0.03	0.80	1.63	0.02
Ti	0.24	0.38	0.10	0.03	0.09	0.01
Cr	0.65	2.47	0.00	0.21	0.38	0.03
Mn	0.04	0.11	0.01	0.01	0.02	0.01
Fe	2.68	4.80	1.11	0.99	3.16	0.14
Ni	0.16	0.16	0.16	0.02	0.02	0.02
Cu	0.01	0.01	0.01	0.26	0.51	0.03
Zn	0.07	0.15	0.01	0.07	0.14	0.02
Zr	0.67	0.67	0.67			
Ag	0.03	0.03	0.03	0.83	1.63	0.03
Sn	0.27	0.27	0.27			
Gd				0.49	0.49	0.49
Ta	0.01	0.01	0.01			
Tl				0.43	0.43	0.43
Pb	0.26	0.26	0.26			
MS	3.72	4.81	2.85	6.27	10.55	3.80

Table 6. Insoluble pollutant concentrations in fog/rime water in the air (in pg/m^3); the missing values indicate that the pollutant was not detected in the corresponding sample type

Element	Rime			Fog		
	mean	min	max	mean	min	max
C	0.019218	0.006581	0.027528	5.734347	1.465976	15.637488
N				0.000011	0.000000	0.000054
O	0.000011	0.000004	0.000015	0.003032	0.000703	0.008273
Na	0.000018	0.000011	0.000031	0.012882	0.001290	0.041631
Mg	0.000047	0.000012	0.000124	0.017335	0.000594	0.076754
Al	0.000955	0.000551	0.001271	0.285546	0.008831	0.950009
Si	0.001778	0.000705	0.002443	0.446877	0.028883	1.733070
P	0.000009	0.000005	0.000016	0.006368	0.000278	0.018464
S	0.000034	0.000005	0.000100	0.023025	0.000000	0.090414
Cl	0.000000	0.000000	0.000000	0.000010	0.000000	0.000029
K	0.000034	0.000008	0.000077	0.013080	0.000635	0.051280
Ca	0.000051	0.000008	0.000132	0.033030	0.001013	0.062578
Ti	0.000190	0.000069	0.000315	0.013467	0.000000	0.063107
Cr	0.000512	0.000000	0.002860	0.083275	0.000000	0.413730
Mn	0.000057	0.000010	0.000216	0.000817	0.000000	0.002198
Fe	0.004436	0.000876	0.009945	0.850449	0.018306	3.746108
Ni	0.000037	0.000000	0.000224	0.000433	0.000000	0.002163
Cu	0.000004	0.000000	0.000012	0.153849	0.000000	0.686999
Zn	0.000100	0.000016	0.000278	0.033552	0.000000	0.151388
As	0.000004	0.000000	0.000026			
Zr	0.000190	0.000000	0.001140			
Mo	0.000002	0.000000	0.000011			
Ag	0.000008	0.000000	0.000047	0.517501	0.000000	2.582619
Sn	0.000067	0.000000	0.000404			
Gd				0.116319	0.000000	0.581594
Ta	0.000008	0.000000	0.000051			
Tl				0.012452	0.000000	0.062260
Pb	0.000080	0.000000	0.000483			

CONCLUSIONS

Mean fog/rime SC concentrations in the air appear higher during the rime events than during the warm fog events at the Milešovka station. This can be a consequence of different sampling techniques. An active sampling device was used for the fog water sampling, and a passive apparatus was employed for the rime water sampling. The duration of the sampling time was also different. Sampling time was of the order of hours in the case of fog and of the order of days in that of rime. The rime samples can contain dust particles. Ice sublimation and thickening of the samples may have also represented relevant factors. This idea can be supported by the results acquired from IP.

The mean IP concentrations in fog were markedly higher than those in rime. Some IP were trans-

Table 7. Insoluble pollutant concentrations in fog/rime water in the air distributed according to the transport sectors (in pg/m^3); the MS marks the mean particle size (in μm); the missing values indicate that a pollutant was not detected in the type of sample

Element	Rime				Fog			
	N	E	S	W	N	E	S	W
C	0.021453	0.016837	0.027528	0.006581	3.486727		15.637488	3.030397
N							0.000054	
O	0.000012	0.000010	0.000015	0.000004	0.001911		0.008273	0.001532
Na	0.000013	0.000030	0.000031	0.000011	0.008700		0.041631	0.002689
Mg	0.000034	0.000040	0.000124	0.000012	0.001737		0.076754	0.003224
Al	0.001138	0.000872	0.000894	0.000551	0.137416		0.950009	0.101445
Si	0.001997	0.001526	0.002443	0.000705	0.141495		1.733070	0.109164
P	0.000008	0.000011	0.000016	0.000005	0.004546		0.018464	0.002141
S	0.000046	0.000019	0.000040	0.000005	0.010947		0.090414	0.001409
Cl	0.000000	0.000000	0.000000		0.000021		0.000003	0.000002
K	0.000049	0.000022	0.000028	0.000008	0.004673		0.051280	0.002387
Ca	0.000030	0.000078	0.000132	0.000009	0.044473		0.032540	0.021833
Ti	0.000229	0.000069	0.000278	0.000107	0.000914		0.063107	0.001199
Cr	0.000002	0.002860	0.000207		0.001323		0.413730	
Mn	0.000034	0.000010	0.000216	0.000016	0.000943			0.001099
Fe	0.004695	0.001709	0.009945	0.000876	0.170581		3.746108	0.082487
Ni		0.000224			0.001082			
Cu	0.000004	0.000009		0.000005	0.005772		0.686999	0.035351
Zn	0.000087	0.000022	0.000278	0.000037			0.151388	0.008186
As		0.000026						
Zr			0.001140					
Mo	0.000004							
Ag	0.000016						2.582619	0.002443
Sn			0.000404					
Gd							0.581594	
Ta			0.000051					
Tl					0.031130			
Pb		0.000483						
MS	3.6	2.8	4.8	3.9	5.4		10.5	5.0

ported to the sampling site by the air flow from only one transport sector or they occurred in only one type of sample (fog or rime). The IP like Zr, Sn, and Ta occurred in the rime water samples in the air flow from the S sector. The occurrence of As and Pb was registered in rime water in the air flow from the E sector, and of Mo and Ag in the air flow from the N sector only. The IP like N and Gd were registered in fog water at the air transport from the S sector, and Tl at the air transport from the N sector only.

The markedly lower content of IP in rime water can be explained by the snow cover occurring in the winter, which reduced the dustiness. The frozen soil could have had the same effect.

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