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CLOUD WATER CHEMISTRY AND ITS EFFECT ON PRECIPITATION COMPOSITION

The occurrence of clouds at ground level over hills is particularly difficult to categorize and quantify in mountainous terrain and constitutes a basic part of our studies. Precipitation/deposition samples (8 rain and 3 cloud collectors) in the Karkonosze Mountains over the period of 2 years (1994–1995) were collected in order to examine chemical interactions between constituents taking place in aqueous medium. An elevational gradient in chemical composition of cloud water and bulk deposition has been investigated. The results show no evidence of significant elevational variations of most ion concentrations when averaged over a month. Concurrent measurements of cloud water chemistry at three sites (1300, 1200, 1000 m asl) have revealed that all ion concentrations were greater at the lowest elevation site.

1. INTRODUCTION

It is well known that low clouds are commonly formed in mountainous regions. The formation of such cap clouds takes place due to the condensation of humid air which occurs when the air is forced to ascend over the mountain barrier. The analyses of water samples from cap clouds have shown that they are often heavily polluted. There is no rain from such clouds but their droplets can be deposited in the ground either directly through turbulent deposition or by being washed out by raindrops falling from the upper layer of clouds. A process is known as the seeder-feeder effect [1]. Dore et al. [2]-[3] have proved that the seeder-feeder effect results in heavy pollution deposition in the mountainous areas along the west coast of Britain, being three-five times higher than in the nearby coastal areas.

In winter months at the Karkonosze Mountains, south-western Poland, the formation of cold air lakes, nearby the polluted valley basin, commonly occurs in the lower part of the ridge being enshrouded in clouds. Since low clouds are observed

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along the Karkonosze ridge for approximately 240 days in the year, then the process of turbulent cloud water deposition and the seeder-feeder effect are expected to be essential for determining the pollution deposition in the Karkonosze Mountains.

The development of this phenomenon was confirmed by the preliminary results obtained within the framework of a joint Polish project [4], [5]. The project was aimed at investigating the pollution and its effects on the forest ecosystem. Bulk precipitation was monitored monthly based on the samples taken at over 20 sites in the Karkonosze National Park during the period of 1992–1994. The results obtained proved that the seasonal deposition of sulphur and nitrogen compounds increased visibly between 1300 and 1500 m asl by a factor of 1.2 and 1.5, respectively. The investigations, however, did not take account of the so-called occult deposition from clouds/fogs. Their drops of water settle on the surface of soil in the form of either liquid deposit at temperatures higher than 0 °C or rime at temperatures lower than 0 °C.

This work is aimed at examining chemical interactions between constituents in precipitation water samples. For this purose the samples of rime, snow and cloud water were gathered during field campaigns. Furthermore, monthly rain and cloud water samples were also collected.

2. DESIGN OF FIELD INVESTIGATIONS

2.1. SAMPLING SITES AND SAMPLING PROCEDURES

Precipitation samples (bulk month precipitation or two-week deposition) were collected at 7 sampling sites located on the western slope of the Mount Szrenica at the altitudes of 1315, 1270, 1220, 1200, 1100, 1000 and 840 m asl from July to October, 1994. There are also 3 cloud water collectors placed at the altitudes of 1315, 1000 and 840 m asl. During the field campaigns in February, 1994 and 1995, July, 1994 and October, 1994 precipitation samples (rain water, cloud/fog water and rime) were collected every day at three measuring points. Each rain collector consisted of a polyethylene bottle with a funnel of 15.2 cm in diameter, placed 1.5 m above the ground. Cloud water was collected using passive cloud collectors. Cloud samples, collected on teflon, V-shaped strings (0.5 mm in dia), are fed into a sample catchpot. A lid of 0.8 m in diameter prevents precipitation from entering.

2.2. ANALYTICAL PROCEDURES

Wet deposition samples (from rain water and cloud water collectors) were collected directly in individual polyethylene bottles (1 or 2 dm³ of volume) with funnels. Measurement of the pH of both cloud water and precipitation were made

directly after sample collection using pH-meter equipped with a combined electrode calibrated using the buffers of pH 4 and 7.

The trace metals in the precipitation samples were stabilized due to addition of suprapure HNO₃. Each sample was stored in a polyethylene bottle and refrigerated during the transport to laboratory. The ICP technique (PU 7000 Philips Scientific) was used to quantify the elemental content of the liquid samples without doing any additional sample preparation. Detection limits calculated for three blanks were extremely low (table 1). Ion-selective method was used to determine the inorganic ions such as chloride and nitrate. Sulphate ions were measured by the thorin method [6], and ammonium ions – by the Nessler method [7]. Individual ion detection limits ranged from approximately 0.5 to 2 µmol/dm³.

Table 1

Detection limits for single element evaluation of an ISP spectrum from water samples

Element	Water [mg/dm ³]
Al	0.0002
Ca	0.0001
Cd	0.00001
Cr	0.0005
Cu	0.00006
Fe	0.0002
K	0.01
Mg	0.00003
Mn	0.00003
Na	0.0004
Ni	0.0008
Pb	0.001
Zn	0.00007

Note: The detection limit is calculated as 3σ for blanks inserted in a normal run. For water samples the ultrasonic nebulizer (U-5000 AT) is used.

3. RESULTS AND DISCUSSION

3.1. COMPARISON OF RIME-SNOW COMPOSITION

The concentrations of inorganic species in snow and rime were measured simultaneously at three different sites along a west-east transect with elevations of 1315, 1200, 1000 m asl, respectively. Investigation of winter precipitations was conducted in February 1994, 1995 (two field campaigns). During the major sampling

events 27 fresh snow samples and 20 rime samples were carried out (12 snow and rime samples collected simultaneously). The variations in the concentrations of the major

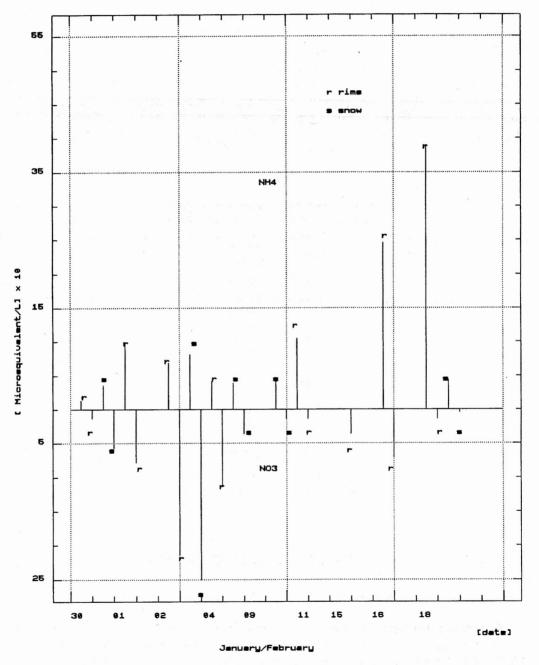
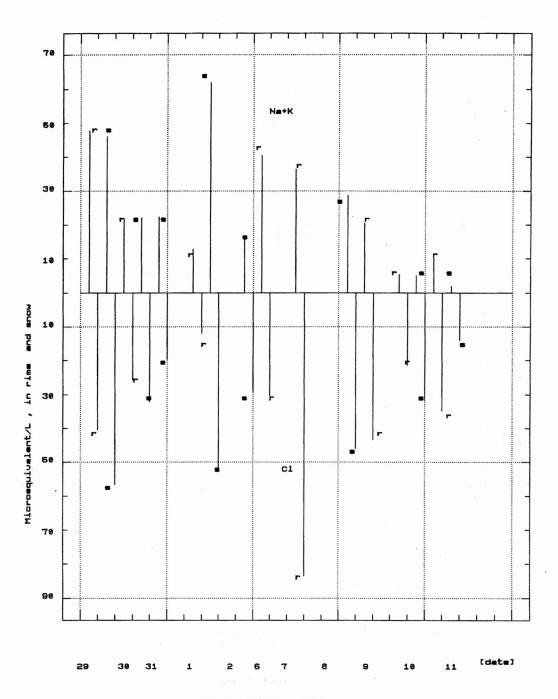


Fig. 1. Ammonium and nitrate concentrations in rime and snow for the Mt. Szrenica (30.01-10.02.95)



January/February 1994

Fig. 2. Sodium, potassium, chloride concentrations in snow (s) and rime (r) for the Mt. Szrenica (29.01-11.02.94)

ions in snow and rime are illustrated in figures 1-4. Tables 2 and 3 provide summary of statistics for the observations. The dual rime/snow sampling was designed to

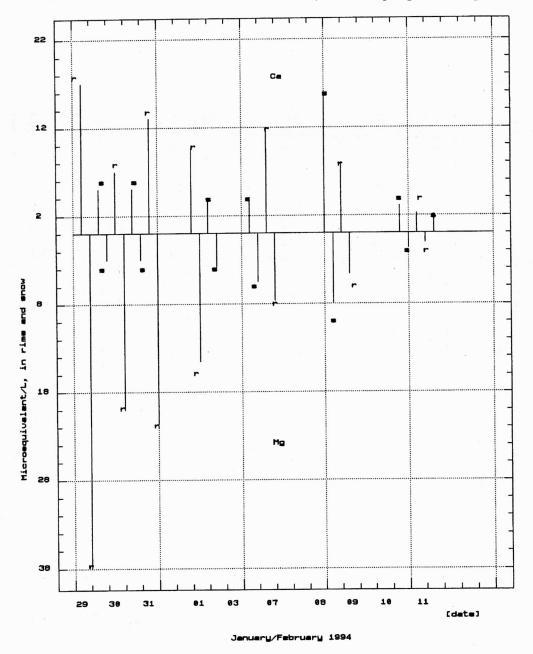


Fig. 3. Calcium and magnesium concentrations in rime (r) and snow (s) for the Mt. Szrenica (29.0.1-11.02.94)

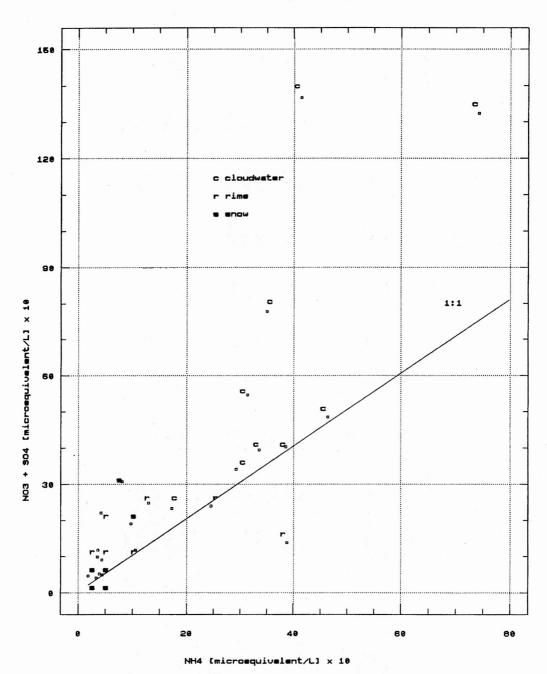


Fig. 4. Acid-base balance in rime, cloud water and snow for the Mt. Szrenica. The line represents the dependence of NH_4 concentration on $SO_4 + NO_3$ concentration. Points below and to the right of this line represent alkaline reaction, while the points above and to the left – acidic reaction

Table 2

Statistical data on snow and rime samples at the Mt. Szrenica (1315 m asl)

Constituent [mg/dm³]	pН	N(NH ₄)	Na	K	Ca	Mg	Cl	SO ₄	Pb	Zn	Fe
28.01-18.02.94	P			Main co	mponents				Tra	ce element	ts
RIME											
Sample size	10	10	10	10	10	10	10	10	10	10	10
Avg conc.	3.55	1.31	3.81	13.25	0.33	0.28	15.23	0.12	0.022	0.021	0.089
Min conc.	3.3	0.1	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Mediane	3.42	1.22	3.5	1.02	0.17	0.13	9.04	0.09	0.013	0.015	0.07
Max conc.	4.08	3.25	10.71	82.5	1.93	1.05	69.7	0.32	0.064	0.093	0.324
Var.	0.085	1.217	1.529	80.73	0.329	0.129	51.9	0.006	0.0004	4×10^{-4}	8×10^{-3}
SNOW											
Sample size	8	8	8	8	8	8	8	8	8	8	8
Avg conc.	3.79	0.83	0.46	0.07	0.14	0.04	1.21	0.041	0.013	0.026	0.018
Min conc.	3.36	bdl	bdl	bdl	0.03	bdl	0.5	bdl	bdl	bdl	bdl
Mediane	3.73	0.53	0.3	0.03	0.06	0.037	1.12	0.045	0.01	0.016	0.013
Max conc.	4.68	2.42	1.32	0.2	0.42	0.09	1.98	0.11	0.032	0.093	0.053
Var.	0.166	0.682	0.231	5×10^{-3}	0.021	8×10^{-4}	0.312	1.4×10^{-3}		8×10^{-4}	3×10^{-4}

evaluate spatial variations in the chemical composition of rime and snow from the same event. The pH ranges between 3.3–5.9 for rime and 3.4–7.1 for snow samples reflect some spatial and temporal tendencies. Depending on the wind direction, contributions of maritime aerosols to the overall chemical budget in precipitation/deposition can be found. Sodium, potassium and chloride (figure 2) clearly reflect the influence of sea salt species in samples collected in January/February (an obvious maritime trajectory).

Table 3
Statistical data on snow and rime samples at the Mt. Szrenica (1315 m asl)

Constituent	pН	N(NH ₄)	N(NO ₃)	CI	SO ₄
		30.01–18.0	2.95		
RIME					
Sample size	9	9	9	9	9
Avg conc.	4.9	1.9	5.8	4.1	6.7
Min conc.	4.4	bdl	bdl	bdl	bdl
Mediane	4.8	1.4	5.2	1	6.1
Max conc.	5.9	5.4	14	30	9.9
Var.	0.28	3.1	15.5	94.8	7.8
SNOW					
Sample size	7	6	7	7	7
Avg conc.	5.57	0.64	4.05	0.82	1.95
Min conc.	3.6	bdl	bdl	bdl	bdl
Mediane	5.44	0.58	1.3	0.5	2.04
Max conc.	7.1	1.13	19	3.5	3.8
Var.	1.2	0.059	44.6	1.45	1.33

3.2. CLOUD WATER VS PRECIPITATION (RAIN) COMPOSITION IN MONTHLY SAMPLE

During the measurement period in July-October 1994, rain (n = 56) and cloud water (n = 10) samples were collected in a cycle of 14 days by means of rain and cloud collectors, respectively. The compositions of rain and cloud water samples (presented as monthly mean ion concentrations $-\mu M/dm^3$) are shown in tables 6-9.

The pH values of the rain water at seven sites ranged between 3.4 and 7.0 (mediane 4.5). The pH ranges found most frequently were as follows: 3.5 < pH < 4.0 (19%), 4.0 < pH < 4.5 (24%), 4.5 < pH < 5.0 (26%). The chemical composition of rain water samples collected at seven various elevations was examined for pH ranges 4.0-4.5 and 4.5-5.0. Precipitation samples with pH values from 4.0 to 4.5 were observed in July, August and October at three elevations: 1000 m, 1200 m, 1220 m asl. The concentrations of $N(NH_4^+)$, $N(NO_3^-)$, Cl^- , Na^+ , K^+ , Ca^{++} , (Pb^{++}, Zn^{++}) in rain water were higher; however, the concentrations of SO_4^- and $Mg^{++}(Ca^{++})$ were lower (mainly in July and October) than those stated at a lower location. For the pH range between 4.5

Table 4

The average ion concentrations in water samples collected in cloud collector at three elevations on the western slope of the Mt. Szrenica

Da	ate	19.10.94	20.10.94	21.10.94	22.10.94		24.10.94		25.10.94	26.10.94	2	7.10.94		28.10.94	29.10.94	30.10.94
Elevation	m asl	1315	1315	1315	1315	1315	1200	1000	1315	1315	1315	1200	1000	1315	1315	1315
\overline{v}	cm ³	300	200	50	50	600	45	24	300	630	900	70	60	225	550	120
pH		5.9	3.8	5.7	5.5	4.5	4.5	4.3	3.5	4.1	4.2	4	3.9	7.2	4.1	4.4
$N(NH_4)$	mg/dm3	2.3	10.2	4.9	6.5	4.1	4.5	13.9	4.9	5.8	2.42	1.99	5.92	4.7	4.4	5.4
SO ₄	mg/dm ³	9.7	8.9	9.9	9.1	6.9	6.3	16.3	10.1	7.5	5.5	10.4	7.3	9.2	6	8.4
Cl	mg/dm ³	5	4.6	3.8	3.6	0.8	4.6	15.8	2.4	2.1	0.6	5.8	6.2	1.3	1.8	1.6
N(NO ₃)	mg/dm3	0.8	16	bld	4.2	2.8	15.2	14.6	8	17	1.7	2.6	14.6	2.9	5.95	5.22
Na	mg/dm3	2.3	10	3.6	1.5	0.29	2.3	7	0.91	0.9	0.18	1.2	7.5	3.9	1	0.9
K	mg/dm ³	0.5	2.3	6.5	2.5	0.24	2.1	5.5	0.47	0.4	0.001	0.4	1.7	0.65	0.34	0.001
Ca	mg/dm ³	3.7	9.5	2.9	6.3	0.24	12.5	2.4	0.35	0.54	0.71	0.73	0.55	0.71	0.73	0.55
Mg	mg/dm3	1.62	8.6	0.5	0.98	0.15	1.1	1.39	0.59	0.55	0.17	0.42	0.63	0.65	0.78	0.65
Pb	mg/dm3	0.38	0.12	bld	0.2	0.06	bld	0.28	0.18	1.09	0.15	bld	0.87	0.05	0.09	0.16
Zn	mg/dm ³	0.21	bld	0.05	0.1	0.47	5	0.9	0.64	0.21	0.12	0.3	0.45	0.37	0.12	0.12
Mn	mg/dm ³	0.27	1.6	0.1	0.1	0.05	0.1	2.9	0.05	0.06	0.03	0.05	0.68	0.06	0.06	0.07
Fe	mg/dm ³	0.64	0.9	0.2	0.3	0.15	0.12	4.24	0.59	0.78	0.59	0.1	4.4	0.37	0.63	0.58
Al	mg/dm ³	0.6	bld	bld	bld	0.05	bld	1.99	0.33	0.23	0.14	bld	2.66	0.19	0.2	0.18
Cu	mg/dm ³	0.19	bld	bld	bld	0.04	bld	0.18	0.035	0.039	0.006	bld	0.19	0.024	0.001	0.0003
$Cd \times 0.01$		0.7	bld	bld	bld	1.07	bld	4.7	1.7	2.33	1.13	bld	2.63	0.8	1.1	0.5

Table 5

Statistical data on cloud water samples at the Mt. Szrenica (1315 m asl) in October 1994

Constituent [mg/dm ³]	pН	Cl	N(NO ₃)	SO ₄	N(NH ₄)	Na	K	Mg	Ca	Pb	Zn	Fe	Mn
Sample size	11	11	11	11	11	11	11	11	11	11	11	11	11
Avg conc.	4.80	2.50	5.90	8.30	5.00	2.30	1.30	1.40	2.40	0.22	0.22	0.52	0.22
Min conc.	3.50	0.60	0.10	5.50	2.30	0.20	bdl	0.15	bdl	bdl	bdl	0.15	bdl
Mediane	4.40	2.10	4.20	8.90	4.90	1.00	0.50	0.65	0.71	0.15	0.12	0.59	0.06
Max conc.	7.20	5.00	17.00	10.10	10.20	10.00	6.50	8.60	9.50	1.09	0.64	0.90	1.60
Var.	1.25	2.34	32.90	2.50	4.50	8.00	3.70	5.90	9.20	0.09	0.04	0.06	0.21

Table~6 Month mean precipitation concentrations [$\mu M/dm^3$] for seven sampling sites, July 1994

***************************************			A	ltitude [m a	ısl]		
a	1 1315	2 1270	3 1220	4 1200	5 1100	6 1000	7 840
Precip. amount	107.0	45.0	67.5	95.5 [mm]	105.0	107.5	92.5
pН	3.8-6.4	6.1	3.8	3.6-4.6	3.9-4.6	4.0-4.8	5.2-6.5
N(NH ₄)	93.2	195.4	53.6	41.6	53.6	62.9	154.9
SO	54.9	83.5	87.2	65.3	71.5	39.4	71.0
Cl	73.2	77.1	28.5	233.6	63.5	89.8	100.0
$N(NO_3)$	104.9	100.0	78.5	160.8	107.1	136.1	85.1
Na	bdl	8.6	bdl	4.7	4.1	10.5	41.8
K	bdl	17.9	7.9	23.8	20.4	13.2	67.4
Ca	38.1	22.6	12.5	24.5	36.3	45.5	23.9
Mg	4.1	5.0	3.3	5.4	6.3	4.8	8.1
Mn	bdl	bdl	bdl	0.9	bdl	bdl	0.4
Fe	bdl	bdl	bdl	bdl	bdl	bdl	0.3
Pb							

1-7 - rain collectors.

bdl - below detection limit.

and 5.0 recorded in August and September at 1000 m, 1200 m, 1220 m and 1270 m asl, ion concentrations (SO_4^- , $N(NO_3^-)$, Na^+ , Ca^{++} , Mg^{++}) showed an upward tendency with the decrease of site elevation. Concentrations of Cl^- , $N(NH_4^+)$, K^+ were higher at lower altitude.

In tables 6–9, there are presented the ion concentrations in cloud water samples collected at the elevations of 1315 m and 1000 m asl, which confirm the mentioned above variability in the concentrations of major ions with altitude for the following pH ranges: 3.1–4.4 (1315 m) and 3.8–6.2 (1000 m). In the most cases, major ion concentrations were by a factor 5–7 or more higher than those found in samples of rain water.

Table 7 Month mean concentrations in rain and cloud water samples $[\mu M/dm^3]$ for seven sampling sites, August 1994

	Altitude [m asl]								
	1	2	3	4	5	6	7	8	9
	1315	1270	1220	1200	1100	1000	840	1315	1000
Precip.	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
amount	[mm]	[mm]	[mm]	[mm]	[mm]	[mm]	[mm]	[cm ³]	[cm ³]
pН	4.4	4.5	4.7	4.5	4.7	4.5	4.6	4.2	6.2
$N(NH_4)$	10.4	9.5	24.7	61.9	31.2	3.5	176.4	153.4	237.9
SO_4	20.6	7.9	27.8	41.2	12.2	35.9	27.8	182.5	99.7
Cl	60.0	54.2	28.5	28.5	57.0	62.8	28.5	25.7	246.8
$N(NO_3)$	85.7	78.5	71.5	71.4	78.5	85.7	92.8	56.2	239.2
Na	4.3	4.3	bdl	4.3	bld	bdl	4.3	43.4	117.3
K	bdl	7.6	7.6	7.6	7.6	bdl	17.9	27.6	61.5
Ca	25.0	22.5	12.5	12.5	30.0	30.0	15.0	65.0	82.3
Mg	2.5	2.5	2.5	4.1	13.3	2.0	2.0	11.2	52.0
Mn	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	0.9
Fe	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl

1-7 - rain collectors, 8-9 - cloud collectors, bdl - below detection limit.

 $\begin{tabular}{ll} Table 8 \\ Month mean concentrations in rain and cloud water samples $[\mu M/dm^3]$ for seven sampling sites, \\ September 1994 \\ \end{tabular}$

	Altitude [m asl]									
	1									
	1315	1270	1220	1200	1100	1000	840	1315	1000	
Precip.	97.5	80.0	102.2	88.5	94.5	67.5	82.5	600	480	
amount	[mm]	[mm]	[mm]	[mm]	[mm]	[mm]	[mm]	[cm ³]	[cm ³]	
pН	5.6-6.4	4.5-4.9	4.6-4.9	4.8-4.9	5.2-5.3	4.6-5.9	4.7-7.0	3.1-4.2	3.9-6.2	
$N(NH_4)$	77.8	39.4	29.5	31.3	62.5	40.8	85.4	82.0	206.4	
SO ₄	41.4	36.4	39.3	45.3	31.7	30.9	39.1	119.6	90.6	
Cl	39.0	35.3	30.6	27.5	35.5	42.8	29.7	78.8	81.7	
N(NO ₃)	47.5	53.6	48.4	60.1	39.7	26.0	33.9	114.3	74.5	
Na	17.4	24.7	20.7	26.2	16.3	14.5	16.4	100.0	139.0	
K	6.8	7.6	6.6	5.1	5.6	23.1	25.5	23.1	28.2	
Ca	131.2	82.5	57.7	131.7	143.6	105.0	54.3	250.0	162.5	
Mg	6.4	4.7	5.4	5.4	5.4	5.0	4.6	50.0	39.6	
Mn	bdl	bdl	bdl	bdl	bdl	bdl	bdl	0.9	1.5	
Fe	bdl	bdl	bdl	bdl	bdl	bdl	bdl	10.7	7.1	
Pb	bdl	0.74	0.20	0.17	0.15	0.09	0.19	0.7	0.19	

1-7 - rain collectors, 8-9 - cloud collectors, bdl - below detection limit.

Table~~9 Month mean concentrations in rain and cloud water samples [$\mu M/dm^3$] for seven sampling sites, October 1994

7 840 26.0	9 1000
840	-
26.0	
[mm]	480 [cm ³]
5.6	3.8-6.2
118.8	133.4
55.6	60.6
25.3	32.5
42.8	75.2
13.0	108.3
33.3	15.9
17.2	51.8
8.3	28.2
bdl	bdl
bdl	20.4
0.10	1.6
bdl	0.58
	5.6 118.8 55.6 25.3 42.8 13.0 33.3 17.2 8.3 bdl bdl 0.10

1-7 - rain collectors, 9 - cloud collectors, bdl - below detection limit.

Direct comparison of the cloud water at three sites of various elevation was limited by fact that cloud interception generally did not occur simultaneously at all sites.

4. CONCLUSIONS

Data on chemical composition of monthly bulk deposition along elevational gradient (840–1315 m) was assembled and used to define their relationships with altitudes for the Mount Szrenica. Our preliminary results show no evidence of significant elevational variations in most ion concentrations when averaged over a month. The investigations did not take account of the so-called occult deposition from clouds/fogs. The results prove that in most cases, the ion concentrations in cloud water ($\mu M/dm^3$) were several times higher than those found in rain water samples.

Concurrent measurements of cloud water chemistry in the event represented by four samples at the Szrenica summit and at 1200 and 1000 m asl have revealed that all ion concentrations were greater at low elevated sites. A comparison of the acidity of cloud water sampled at the Mount Szrenica with that at other sites

showed that the pH was quite low. pH values of cloud water were less acidic at the higher altitudes. This difference occurs despite the maximum concentrations of NO_3^- , SO_4^- , Cl^- . The key reason for the pH difference between the sites is the amount of NH_4^+ in the cloud water. More detailed research, however, is needed to improve our understanding of variations in cloud water with altitude, LWC and chemistry in order to assess elevational gradient in cloud water chemistry in the region under study. These processes will be considered in the future within the EASE (Emission Abatement Strategies and the Environment) project.

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CHEMIA WODY TWORZĄCEJ CHMURY I JEJ WPŁYW NA SKŁAD OPADU

W terenie górzystym skład opadu szczególnie trudno jest sklasyfikować i określić ilościowo chmury występujące przy powierzchni ziemi i nad wzniesieniami. W okresie dwóch lat (1994–1995) w Karkonoszach pobierano próbki padającej i osadzającej się wody (8 zbieraczy deszczu i 3 pary wodnej z chmur), aby zbadać interakcje chemiczne pomiędzy jej składnikami. Przeanalizowano także wpływ wysokości na skład chemiczny wody deszczowej i osadu. Okazało się, że nie ma znaczących zmian w stężeniach większości badanych jonów, gdy wzięto pod uwagę wartości średnie dla miesiąca. Równoważne pomiary dla wody z chmur w trzech miejscach (1300, 1200 i 1000 m npm) wykazały, że wszystkie stężenia badanych jonów były największe na najniżej położonym miejscu.

химия воды, образующей туманы и ее влияние на состав осадка

В гористой местности состав осадка особенно трудно классифицировать и определить по количеству туманы, выступающие при поверхности земли и над возвышенностями. В течение двух годов (1994—1995) в Крконошах брали пробы падающей и осаждающейся воды (8 подборщиков воды и 3 водяного пара из туманов), чтобы исследовать химические взаимодействия между его компонентами. Проведен также анализ влияния высоты на химический состав дождевой воды и осадка. Оказалось, что нет существенных изменений в концентрациях большинства исследуемых ионов, когда брали во внимание средние месячные значения. Эквивалентные измерения для воды из туманов в трех местах (1300, 1200 и 1000 м) обнаружили, что все концентрации исследуемых ионов были самыми большими на наиболее низко расположенном месте.