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Investigations of ground-based clouds at the Mt. Brocken

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Abstract Results of the 1996 field season (April-November) of the cloud chemistry program starting in 1992 at the Mt. Brocken are presented. The mountain site (1142 m a.s.l.) is located in the Harz Mountains/Germany. For a large part of the database (about 40%), both cloud water and drizzle and/or precipitation were present at the same time. A separation into two groups was done: non-precipitating clouds and mixed category of events. More than 2000 one hour samples were collected and analysed for major ions. The mean cloud water pH for the frostfree period in 1996 was 3.9. Histograms giving the distribution of pH values show that the pH values of samples from non-precipitating clouds ranges from 2.5 up to values > 8 and that the distribution is bimodale. For the 1996 field season period as a whole, pH values of cloud water were lower than those of mixed samples (cloud water, drizzle, precipitation) at the Brocken site. This was related to higher concentrations of all ions in cloud water. Cloud base measurements using a ceilometer confirmed the correlation between liquid water content and height above cloud base and consequently the influence of sampling height relative to the cloud base on the total ionic content of cloud water.

Introduction

The Brocken program was designed and implemented in 1990/91 to study physical and chemical processes occurring in ground-based clouds, to establish a cloud chemistry climatology (as a long term goal) and to study also the influence of the expected high decrease of emission in East Germany on the chemical composition of the atmosphere [17]. 30–50% of the total water budget of terrestrial ecosystems may be derived from clouds [1]. Results from several Mountain Cloud Chemistry Programs (e.g. Mt. Brocken, Germany; Mt. Mitchell and Whiteface Mt., USA) indicated that clouds covering these upland areas for much of the year were both more acidic and had higher concentrations of major ions than precipitation at the same site [2–5, 11, 15]. Cloud water deposition of ionic matter may be 2 to 5 times the deposition received due to precipitation.

The Mt. Brocken (51.80°N, 10.67°E) is located in the National Park Hochharz. Because of its exposed position (the highest Harz mountain, 1142 m a.s.l.) in the surrounding lowlands and the high occurrence of clouds (see Fig. 1a), the Brocken summit is an excellent platform for air and cloud chemical measurements. For this research program experiences from the Mountain Cloud Chemistry Program in the eastern United States [2–4] and from the cloud research program at mountain sites in the United Kingdom [5, 27] were adopted. Selected results of the year 1996 will be presented in this paper. The weather station is situated on the top of the Mt. Brocken, at a small plateau above the tree line with subalpine vegetation. Whereas the surface wind in the lower Harz region is strongly influenced by the mountains (air masses will be taken around the Harz massif), the wind field at the Brocken summit itself is relatively uninfluenced and represents the predominant low tropospheric wind. The air masses are streaming over the mountain top. So the station provides access to regional air flow. This was shown by simulations of different synoptic situations using the nonhydrostatic "Karlsruhe Atmospheric Mesoscale Model" [6]. The summit plateau of the Mt. Brocken is normally situated in the upper atmospheric boundary layer (ABL) or even in the free troposphere [7] due to the steep slopes at all of its edges. The wind field at the station is dominated by westerly/south-westerly winds.

Since 1895 a weather station has been working at the Brocken site. The highest annual mean temperature at the Brocken summit was determined in 1989 with 4.8 °C, the

Dedicated to Professor Dr. Günter Henrion on the occasion of his 65th birthday

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lowest annual mean temperature was observed with 1.3 °C in 1902 and 1922. The highest daily value was measured with 27.9 °C on August 9th 1992, the lowest with –28.4 °C on February 1st 1956. There could be snow until the beginning of June and from the end of September. The precipitation is between 948 mm (in 1953) and 2335 mm/year (in 1981). These and more long-time observation data are given in [8].

Experimental

The measurement station consists of a wooden house with a platform roof about five meter above ground where all meteorological, cloud physical, and sampling devices are installed. Meteorological standard parameters (wind direction and velocity, temperature and humidity, global radiation) are measured with commercial instruments (Thies company). At the Brocken station trace gases (O_3, NO_x, SO_2) are measured using commercial automatic analysers [7, 11]. The measurements are checked several times using a mobile "transfer" standard calibration system which is calibrated against primary standards (U.S. N.I.S.T.).

The liquid water content of clouds (LWC) has been measured continuously using the Gerber Particle Volume Monitor (PVM 100). This instrument has a very robust construction. The measurement principle is the forward scattering of laser light by the cloud water droplets in the open air along a 40 cm path. The PVM measures in situ and in real time the integrated volume of suspended water droplets with a stated precision of 0.002 g/m^3 [9]. In 1994 a field intercomparison of all Gerber instruments available in German atmospheric research groups was carried out at the Mt. Brocken and the results agreed very well [10]. Under field conditions the PVM is calibrated weekly by the light diffusion disk (span), zero adjustment is controlled daily by the site operator. The average uncertainty becomes no greater than 0.009 g/m^3. The registered on-line analogue signal from the measurement system (sampling rate 5 s) is used to calculate automatically 30-s, 10-min and 1-h averages of LWC. Cloud water is collected in 1-hour time resolution to be in coincidence with the time scale of synoptic observations (e.g., cloud type, cloud frequency, cloud base) and in the typical time scale of weather changes. This high time resolution requires an automated cloud water collection system, which was designed and implemented in 1993 by our group [11]. A passive collector (ASRC-type), which uses wind speed to effect cloud water impacting on 0.4 mm Teflon strings [12], is housed in a collector tower. The PVM 100 instrument is used too as a fog presence detector to initiate sampling. If LWC arises the 25 mg/m^3

Table 1 Results of the WMO-GAW Acid Rain Performance 1996 [14]

Component	Sample	Expected value	BTU Cottbus		All participants		
			Mean	Std. dev.[a]	N[b]	Mean	Std. dev.
pH [pH units]	1	4.26	4.24	0.04	58	4.29	0.17
	2	3.88	3.83	0.02	57	3.90	0.08
	3	3.53	3.49	0.02	57	3.56	0.06
Conductivity [µS/cm]	1	26.3	25.6	0.2	55	24.5	2.8
	2	65.7	63.8	0.1	54	61.7	3.8
	3	137.1	132.0	0.1	53	128.3	6.2
SO_4^{2-} [mg S/L]	1	0.919	0.91	0.02	57	0.94	0.11
	2	2.640	2.58	0.04	57	2.68	0.24
	3	3.810	3.56	0.12	57	3.90	0.35
NH_4^+ [mg N/L]	1	0.079	0.08	0.01	54	0.08	0.02
	2	0.624	0.61	0.02	52	0.63	0.07
	3	0.792	0.75	0.03	53	0.80	0.08
NO_3^- [mg N/L]	1	0.111	0.12	0.01	58	0.11	0.01
	2	0.116	0.12	0.01	58	0.12	0.01
	3	1.390	1.38	0.04	57	1.39	0.10
Cl^- [mg/L]	1	0.296	0.27	0.02	56	0.31	0.07
	2	0.640	0.60	0.01	56	0.62	0.07
	3	0.938	0.90	0.02	57	0.94	0.11
Ca^{2+} [mg/L]	1	0.055	0.03	0.01	54	0.06	0.02
	2	0.136	0.09	0.02	55	0.14	0.04
	3	0.006	0.01	0.01	52	0.02	0.02
K^+ [mg/L]	1	0.070	0.08	0.01	56	0.08	0.02
	2	0.085	0.09	0.01	55	0.09	0.02
	3	0.097	0.12	0.01	57	0.11	0.03
Mg^{2+} [mg/L]	1	0.041	0.04	0.01	55	0.04	0.01
	2	0.020	0.03	0.01	55	0.02	0.01
	3	0.081	0.08	0.01	55	0.08	0.01
Na^+ [mg/L]	1	0.185	0.19	0.01	56	0.20	0.03
	2	0.250	0.25	0.01	57	0.25	0.03
	3	0.490	0.49	0.02	56	0.49	0.05

[a] For 10–12 repeated measurements;
[b] Number of individual values/participants

threshold this "wet only" sampling unit will be opened and the string collector will be moved to the sampling position. The impacted cloud water flows through the collector funnel and a sampling tube in one of the 24 1-L polyethylene sample bottles of the ISCO 3700 sampler with time controlled position change (Dr. Staiger, Mohillo + Co. GmbH). The samples are stored at 4 °C. A rain detector is working parallel to differ between non-precipitating and precipitating cloud events. LWC measurements and cloud water sampling are carried out only during the frost-free periods, as a rule between middle/end of April and middle/end of November.

The samples are analysed for anions (Cl^-, NO_3^-, SO_4^{2-}) and cations (Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}) by ion chromatography and for pH (WTW pH196 plus Mettler Toledo InLab 422 electrode) and conductivity (WTW LF96 plus WTW TetraCon 96–1.5 or Philips PW 9513 electrode). The anion chromatographic system consists of a SYKAM pump, a separation column LCA A14, a micromembrane suppressor DIONEX ASRS-II, a SYKAM conductivity detector and a JASCO sampler; the eluent contains 7.5 mM/L sodium carbonate/2 ml/L modifier. For the analysis of the cations a SYKAM pump, a separation column Waters IC-Pak CM/D, a SYKAM conductivity detector, a JASCO sampler and the eluent 0.1 mM/L EDTA/3.0 ml/L HNO_3 are used. The data obtained with ion chromatography correspond to the dissolved fraction.

The acceptance criteria for ion balance and for conductivity (measured/calculated), respectively, are defined in the Reports 85 and 102 of the World Meteorological Organisation [13] for all activities within the Global Atmospheric Watch (GAW) Program and also we follow rigorously these standard practice. The whole measurement and analytical procedure is laid down in standard operating procedures. Annual samples of the WMO/GAW laboratory intercomparison program organised by the U.S. EPA and since 1996 by QA/SAC Americas are analysed. Results for the 1996 intercomparison are given in Table 1.

Results of the 1996 Mt. Brocken cloud chemistry program

The volume content of the liquid phase in clouds is very low, e.g. at the Mt. Brocken about 3×10^{-4} L/m^3, but the liquid water content of clouds (LWC) predominantly determines the total ionic content of clouds (TIC, sum of dissolved Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}) [11]. The LWC of ground-based clouds at the Mt.Brocken summit was measured from April 18th to November 27th, 1996 with a data completeness of 97%. Clouds were observed very often at the Brocken summit, occurring 56% of this time period. Figure 1a shows the monthly frequency distribution of "Brocken-station in cloud" for the year 1996. Cloud frequency is defined as the percent of 10 minutes periods having a cloud liquid water content of minimum 0.010 g/m^3. Cloud frequency varied from 22% in April 1996 (16% according to synoptic observations for whole April by the German Weather Service, station Brocken) to 86% in November 1996. In Fig. 1b the relative frequency distribution of the LWC of clouds at the Mt. Brocken is given for the year 1996 in comparison to the mean of the years 1991–1995, indicating a shift to cloud events with higher LWC values in 1996. The 10-min averages of LWC in 1996 ranged from 0.011 to 1.412 g/m^3. The average LWC of all cloud events in the 1996 field season was 0.328 g/m^3.

Cloud water sampling at the Mt. Brocken began in 1996 on April 23rd. The liquid water content of most of the clouds experienced at the Mt. Brocken was high, allowing sufficient hourly collection volumes. Over 2081 hourly samples were collected through the field season until November 6th. This means that about 88% of all clouds observed at the Mt. Brocken with a cloud base lower than 1142 m above sea level during this frost-free period were analysed.

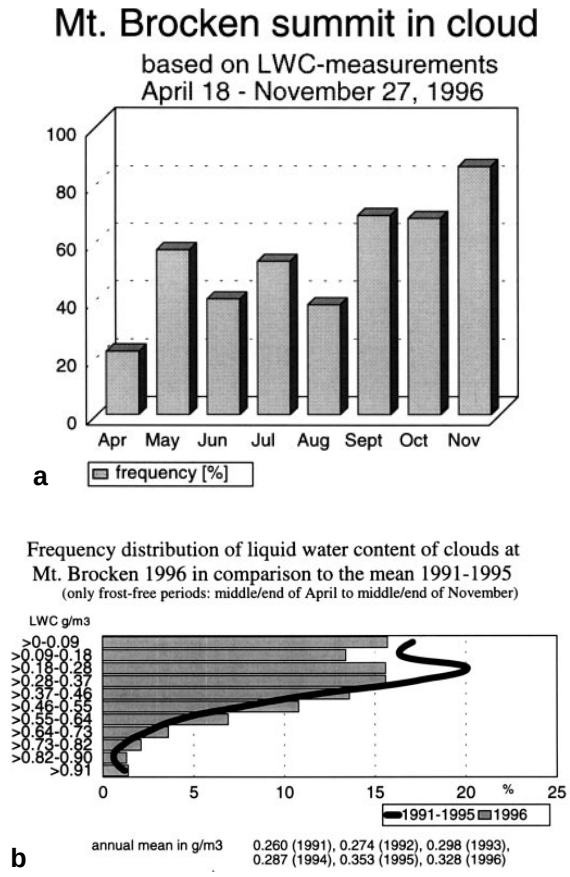

Fig. 1a–b Clouds at the Brocken summit and their liquid water content

Based on the check for ion balance (in equivalents):

$$\left|\frac{(\text{sum of cations} - \text{sum of anions})}{(\text{sum of cations} + \text{sum of anions})}\right|$$

98.5% of the samples fulfilled the criteria defined in the WMO – GAW program. The deviation was significantly below 10% for 95% of these atmospheric samples. Ions like sulphite, acetate, formiate, fluoride, nitrite, not detected within the monitoring program, can contribute to a few percent to the anion equivalent level [24, 25].

Cloud events at a mountain site are frequently accompanied by light precipitation in the form of drizzle. At the Brocken site this was observed mostly when the liquid water content of the cloud was about 600 mg/m^3 or higher, and the higher the LWC the higher was the amount of drizzle. Also more or less intense rain can occur together with the low clouds. If precipitation fell during the sampling period the dilution of cloud water ionic content must be taken into consideration. The data set is separated in non-precipitating and precipitating (drizzle and rain)

Table 2 Cloud water composition for clouds observed at the Brocken summit in the 1996 field season (Apr. 23rd to Nov. 6th) given as LWC weighted averages and as mean of dissolved pollutants per m^3 air volume

Component	Non-precipitating clouds, 1234 samples		precipitating clouds 811 samples	
	µeq/L	neq/m^3	µeq/L	neq/m^3
Cl^-	101	34.8	43	18.8
NO_3^-	360	123.7	111	47.4
SO_4^{2-}	312	107.1	122	52.4
Na^+	100	34.5	41	17.7
NH_4^+	472	162.1	161	69.1
K^+	1.3	0.4	0.6	0.3
Ca^{2+}	51	17.5	22	9.3
Mg^{2+}	24	8.3	9	3.8
H^+	131	45.0	45	19.4
LWC	343 mg/m^3		428 mg/m^3	

clouds. Characteristic compositions of both "cloud types" are given in Table 2.

Arithmetic averages are sensitive to the occurrence of even a few heavily concentrated samples. These samples can be characterised by either pollution events or very low liquid cloud water content [11]. To exclude the influence of such outliers, weighted averages were used. Therefore the following expression is applied:

$$\frac{\sum_{i}^{n}\left(LWC_i \times c_{i,j}\right)}{\sum_{i}^{n} LWC_i}$$

LWC – liquid water content of clouds during sampling time of the individual sample i (1-n),
c – concentration for a specific ion j of the individual sample i,
n – total number of samples available for averaging.

Because of an anti-correlation between ion concentration and liquid water content (collected cloud water amount) the LWC-weighted average concentration is about 13–37% lower than the unweighted average. The amount of dissolved pollutants per m^3 of air volume can be obtained by multiplication of the ion concentration in the collected cloud water sample with the liquid water content measured during the sampling period. As it was expected, the concentration of all species is significantly higher in non-precipitating clouds than in precipitating clouds, reflecting a dilution of the cloud water concentrations by precipitation. The dissolved total ionic content (TIC) of air masses has reached in non-precipitating clouds in 1996 values up to 6658 neq/m^3, whereas in precipitating clouds only values up to 2227 neq/m^3 were observed. In both classes the dominant cation was NH_4^+, the dominant anions were SO_4^{2-} and NO_3^-.

The pH values measured in the whole 1996 field season in samples of non-precipitating clouds show a distribution peaking in the pH class 3.75–4.0, with a long tail to higher values (up to pH values > 8.0), see Fig. 2. Besides, the distribution is bimodale, a second maximum is observed for pH values between 6.0 and 6.25. The distribution of pH values of precipitating clouds has only one significant peak in the pH class 4.25–4.50. The mean pH value is 0.5 pH units higher than the mean of cloud water alone, see also Fig. 2. This is in agreement with data reported 1992 by Aneja et al. [16] as a result of the Mt. Gibbs research program (NC, USA).

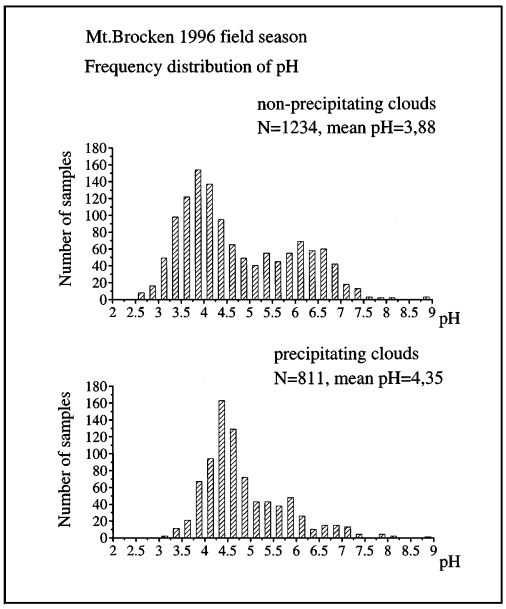

Fig. 2 Absolute frequency histograms of pH values of non-precipitating and precipitating clouds occurred at the Mt. Brocken between April 23rd and November 6th, 1996

Variations in the concentrations of ions in cloud water are in part linked to regional pollution climates [17]. Highly polluted air masses lead to larger TIC for given LWC and vice versa for less polluted air [18]. For example, at the Mt. Brocken the acid events and the largest concentrations of SO_4^{2-} are connected with E-SE trajectories (anthropogenic influence East Germany/East Europe). Fig. 3a–b illustrates the pollutant loading of two selected cloud events observed in September 1996. The dissolved total ionic contents per m^3 of air volume and the LWC are plotted vs. time. A high pressure system over northern Europe/ Norwegian Sea determined the weather in northern and eastern Germany on September 20th/21st with strong E-SE winds. The cloud water samples taken at the Mt. Brocken during these days were characterised by very high concentrations of SO_4^{2-} (up to 45% of TIC, see Fig. 3a), NH_4^+ (up to 45%), Ca^{2+} (up to 12%) and Mg^{2+} (up to 6%). The acidity was also high (pH 3.36–3.4 between 02:00 and 06:00 Central European Time (CET) on September 21st), but partly compensated by the high level of basic components. In the gas phase sulphur dioxide concentrations up to 75 µg/m^3 were observed. Later (7:20 CET) in connection with the influence of a low pressure system over France strong precipitation was observed

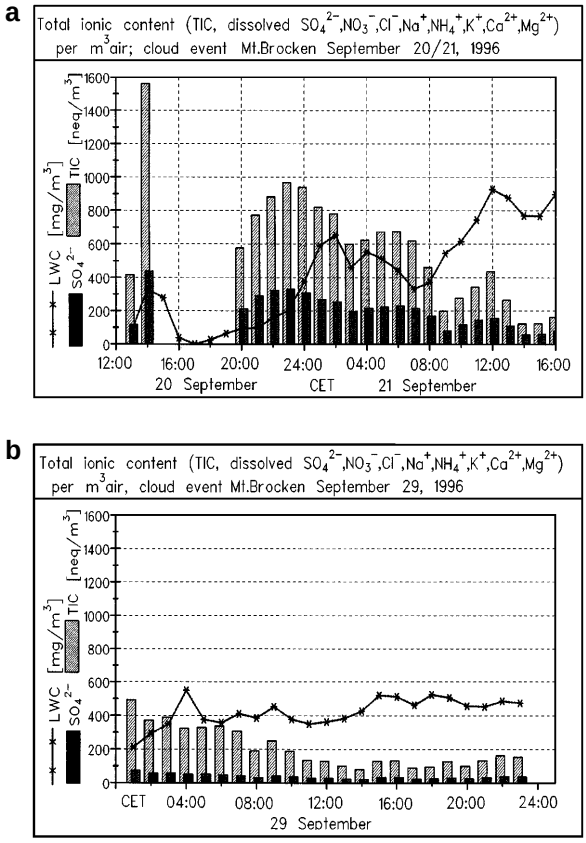

Fig. 3a–b Cloud water composition of selected Mt. Brocken cloud events in September 1996

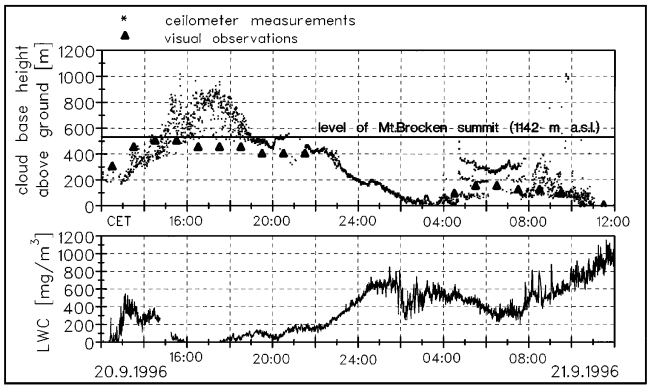

Fig. 4 Liquid water content measurements (Brocken summit) and cloud base measurements (Schierke, 610 m a.s.l.) during the cloud event September 20th/21st 1996. Visual observations of the cloud base height: German Weather Service, Braunlage (607 m a.s.l.)

from time to time also at the measurement site. This resulted in a substantial lower pollutant loading of the atmosphere, see Fig. 3a.

A marked low pressure system over Iceland determined the weather situation over Germany on September 29th with strong westerly winds and an extended cloud layer over NE Germany and Scandinavia. Air masses reached the Mt. Brocken on that day showed much lower concentrations of trace species in the gas phase (e.g. 2–5 µg/m^3 SO_2) and in the liquid phase, see Fig. 3b. Sulphate amounts to TIC only up to 28%. Partly also low pH-values (pH 4.3) were observed; in that case only 2% of TIC were Ca^{2+}. Rainfall was observed since about 7:00 CET with different intensity at the Mt. Brocken. The pollutant loading became lower because of an efficient wash-out of trace species.

Cloud droplets in the atmosphere develop on omnipresent aerosol particles which are not uniform in size and composition. As shown theoretically by Pruppacher and Klett [19] and others, for any given size, particles with a larger soluble fraction are more likely to act as cloud condensation nuclei [20]. The mass of the particulate material associated with droplets is determined by many microphysical and chemical processes like entrainment and mixing of sub-saturated air with cloudy air, coagulation of small particles with larger droplets and heterogeneous reactions. The number of cloud droplets depends on the cloud type and on the distance to the base of the cloud. Continental cumulus clouds have the highest droplet number, while marine cumulus clouds have lower numbers and stratus clouds the lowest [21]. The first ones have small droplet diameters, while droplets in stratus clouds can grow to large diameters, up to 30 µm. The droplet spectrum has a large influence on the formation of rain and on the optical characteristics of clouds. These differences in droplet spectrum, in LWC, in vertical motion and in time duration for the different cloud types cause differences in cloud water chemistry [15, 26–28]. Literature studies [29] and own results [22] show an influence of sampling height relative to the cloud base on LWC and also on TIC (Fig. 5).

For most cloud types the increase of LWC with altitude above cloud base follows approximately a linear function [11, 23, 29] and reaches a maximum at 80–90% of the total cloud depth. From the maximum to the cloud top the LWC decreases sharply. Near cloud base processes like droplet evaporation by mixing with subsaturated air and aerosol and gas scavenging disturb obviously the adiabatic conditions in the clouds.

Since September 1995 we have carried out direct optical cloud base measurements 532 m below the Brocken summit continuously (in Schierke; 610 m a.s.l.; 4.5 km in horizontal direction south/east of the Mt. Brocken) using a ceilometer additional to the visual observations of cloud types and cloud base altitude by the German Weather Service Braunlage. The ceilometer (Vaisala) operates on a LIDAR principle with a time resolution of 15 seconds and a spatial resolution of 30 m. The cloud base was found often between 900–1200 m a.s.l. for convective clouds (*cumulus*, *cumulonimbus*, *stratocumulus*, Sc/Cu) and 700–800 m a.s.l. for stratiform clouds (*stratus neb*, *stratus fractus*). Stratiform clouds amount to about 20% of the ground-based clouds. For the selected cloud event in Fig. 3a the position of the sampling site relative to the cloud base is shown in Fig. 4. The main cloud type during this cloud event on September 20th/21st 1996 was *stratus*, at the beginning (12:00–18:00 CET) *stratocumulus* was ob-

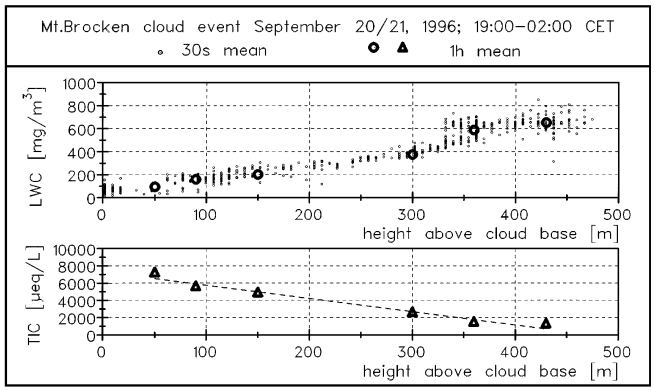

Fig. 5 Influence of the height above cloud base on the liquid water content (LWC) of the cloud and on the total ionic content (TIC) of the cloud water

served. The data shown in Fig. 5 relate to a period of several hours in the evening when the cloud base comes down to a lower level. A high correlation between the height above cloud base and both the liquid water content of cloud and the total ionic content of the cloud water was found. On September 29th 1996 the Brocken summit remained in stratus cloud all day with low variation in the height above cloud base (between 250–400 m) and liquid water content (mostly between 400 and 500 mg/m^3, see Fig. 3b).

Conclusions

The presented data reflect the liquid phase composition of ground-based clouds occurred at the Mt. Brocken between April 23rd and November 6th, 1996. The 10-min averages of liquid water content of all cloud events in the 1996 field season ranged from 0.011 to 1.412 g/m^3 (mean 0.328 g/m^3). Liquid phase samples were taken in 1 h resolution and analysed from about 88% of all cloud events. An influence of sampling height relative to the cloud base on the liquid water content of the cloud and also on the total ionic content of the cloud water could be shown. A separation in samples of non-precipitating clouds and precipitating clouds (drizzle and/or rain) was done. The latter have on the average by a factor 2–4 lower concentrations of all ionic species in comparison to clouds without precipitation. A bimodale distribution of pH values of samples from non-precipitating clouds was found, peaking in the pH classes 3.75–4.0 and 6.0–6.25. In cloud water samples connected with easterly/south-easterly air masses a substantial higher ionic content was observed than in clouds from westerly directions and the dissolved sulphate was the dominant ion.

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