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Intercomparison of fog water samplers

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ABSTRACT

During the Po Valley Fog Experiment 1989, 2 fogwater collectors were operated simultaneously at the ground and the results were compared to each other. The chemical analyses of the samples as well as the collection efficiencies showed remarkable differences between both collectors. Some differences in the solute concentrations in the samples of both collectors could be expected due to small differences in the 50% cut-off diameters ($5 < d_{50} < 7$ μm). The large differences in the collection efficiencies however cannot be explained by these small variations of d_{50}, because normally only a small fraction of the water mass is concentrated in the size range of 5–7 μm droplets. It will be shown, that it is not sufficient to characterize a fogwater collector only by its cut-off diameter. The results of several wind tunnel calibration tests show, that the collection efficiencies of the fogwater collectors are a function of windspeed and shape of the droplet spectra. Calculations to simulate the performance of each collector give an idea of the shape of the collection efficiency curves, especially for the larger droplets. Differences in fogwater concentrations sampled simultaneously in 2 levels of the FISBAT-tower (Fuzzi et al., 1992) by identical collectors can occur due to changing collection efficiencies caused by different windspeeds. Since it is also possible to simulate the time evolution of the solute concentration in the fogwater samples, it will be shown, that changes in the solute concentration can be explained to a large extent by changing microphysical conditions.

1. Introduction

Based on the experience gained during an intercomparison study dealing with chemistry and microphysics of fogwater collection (Enderle and Jaeschke, 1988), it was decided to use impactors for the sampling of fogwater during the joint field campaigns of the EUROTRAC subproject GCE.

Two single-stage fogwater impactors of different design were operated side by side during the Po Valley Fog Experiment 1989 by the Institute for Experimental Physics (IEP) of the Vienna University (Berner, 1988) and the Meteorological Observatory of the German Weather Service (MOH) (Winkler, 1986). Additionally one set of each collector was operated alternatively at four levels of the FISBAT-Tower to obtain vertical gradients of fogwater solute concentrations (Fuzzi et al., 1992).

Since the physical- (sampled volume) as well as the chemical data show differences between these two collectors, it will be useful to look for reasons for this behaviour. Especially the investigation of vertical concentration gradients requires a good estimation of possible variations in fogwater solute concentrations due to the sampling characteristics of the used collectors. Differences in solute concentrations can occur, even when measured by identical collectors, due to different windspeeds in each level, which influence the size dependent inlet or

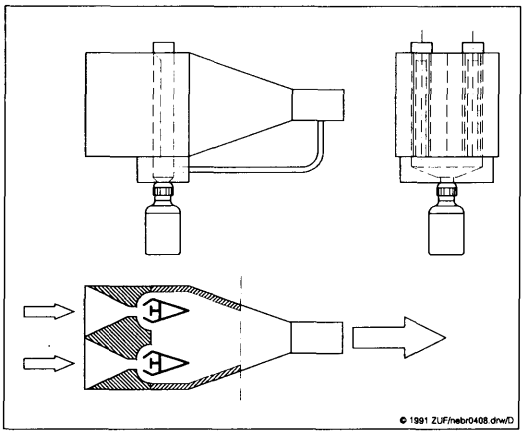

Fig. 1. Schematic of the design of the MOH-collector (with permission of ZUF-Verlag Frankfurt).

aspiration efficiency of each sampling device (see Subsection 5.1). Several tests were performed in the fog chamber CHIEF of the Netherlands Energy Research Foundation ECN to validate the characteristics of the collectors. Since the IEP-collector is relying on traditional single round jet impactor practice and theory, the calculated cut-off characteristics should be reliable and only few tests were performed. The MOH-collector uses a slit impactor technique with an u-shaped impaction surface. The impaction characteristics could then become to some extent different from that of traditional impactors (due to secondary impactions on the side walls of the impaction body). Therefore additional wind-tunnel tests were made to obtain the cut-off characteristics of this special design.

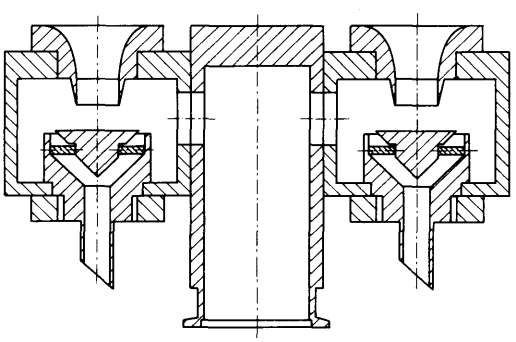

Fig. 2. Schematic of the design of the IEP-collector.

2. Instrumentation

Both fogwater collectors are single-stage inertial impactors based on the work of Marple and Willeke (1976). They operate under the principle that if a stream of particle-laden air is directed to a surface, particles of sufficient inertia will impact on the surface whereas smaller particles will follow the streamlines and will not be collected. The MOH-collector (Fig. 1) is designed as a slit impactor, while the IEP-collector (Fig. 2) uses round nozzles.

The cloudy air enters the MOH-collector via two rectangular inlets oriented vertically side by side, each 12 cm long and 6 cm wide (Fig. 1). The direction of aspiration is in the plane of the wind. The instrument was operated at a flow rate of 125 $m^3 h^{-1}$, thus resulting in a mean inlet velocity of 2.4 $m s^{-1}$. The air is accelerated through the conical nozzles and directed towards the u-shaped impaction plates. The instrument has a calculated cut-off diameter (diameter of 50% collection efficiency) of 5 μm. The droplets are collected on a plexiglass plate, which is bordered by side-walls to prevent the impacted droplets from blowing off the plate. The collected water enters the rear part of the collection bodies via two small slits between the impaction plates and the border walls. In this part of the impaction bodies, the pressure is slightly reduced by means of tapping the static pressure on a particular point with a reduced flow cross-section to support the water flow into the vial.

The IEP-collector consists of four identical jet impaction stages, arranged on the four corners of a square and connected to a common exhaust tube equipped with a throttle for limiting the flow. The fog enters the individual stages via an upward facing round nozzle with rounded edges (Fig. 2). The direction of aspiration is vertical to the wind. The cylindrical throat of the nozzle has a diameter of 16.2 mm and a length of about 16 mm. The circular collection disc is mounted at a distance of 16 mm from the mouth of the nozzle. The instrument was operated at a total flow rate of 76 $m^3 h^{-1}$ resulting in a calculated cut-off diameter of 7 μm. The deposited liquid is driven by the airflow to the edge of the collection plate and runs into a funnel. The nozzle, the disc and the funnel are made from PTFE (polytetrafluorethylene) and the vial from PE (polyethylene).

All other parts of the stage are made from PVC (polyvinylchloride). For further analysis the collected water has to be poured together from the separate vials sitting under each impaction stage.

3. Fogwater sampler intercomparison tests in CHIEF

3.1. Experimental setup

In September 1990, the IEP and MOH fog-samplers have been tested in CHIEF (*Ch*amber for *I*nvestigation with *E*quilibrated *F*og), operated by the Netherlands Energy Research Foundation ECN (Mallant, 1988). After the air was equilibrated in a large vessel, it entered the wind tunnel, first passing the beam of a PVM-100 (Particulate Volume Monitor) (Gerber, 1991; Arends et al., 1992) to determine the liquid water content. The PVM-100 is a forward scattering laser spectrometer in which a laser beam crosses the air over a length of 42 cm with an optical probing volume of 3 cm^3, and light scattered forward in a small angle (0.32–3.58°) is registered. Whenever it was possible, two instruments were tested simultaneously. The tests were performed in 21 runs at windspeeds of 0.8 to 15 m s^{-1} (Table 1). Mainly two different droplet spectra were used, one with a volume median diameter (VMD) varying between 8 and 10 μm and the other with a VMD of about 20 μm (Fig. 3). In the first experiments (runs 1 to 18) the fog was produced from one salt solution (KCl). After each run, the collected water was analyzed for K^+ and compared to the concentrations in the stock solution which was nebulized. The intercomparison was performed between 3 instruments:

(i) the IEP collector in the version used in the Po Valley Fog Experiment 1989;

(ii) the MOH collector in the version used during the Po Valley Fog Experiment 1989, named as MOH-old;

(iii) a new version of the MOH sampler, called MOH-new, which was constructed according to the old shape, but manufactured from one piece of perspex, so that the edges were worked out better and were sharper. A new design of the impaction body is used in the new type in order to improve the water transports into the vial.

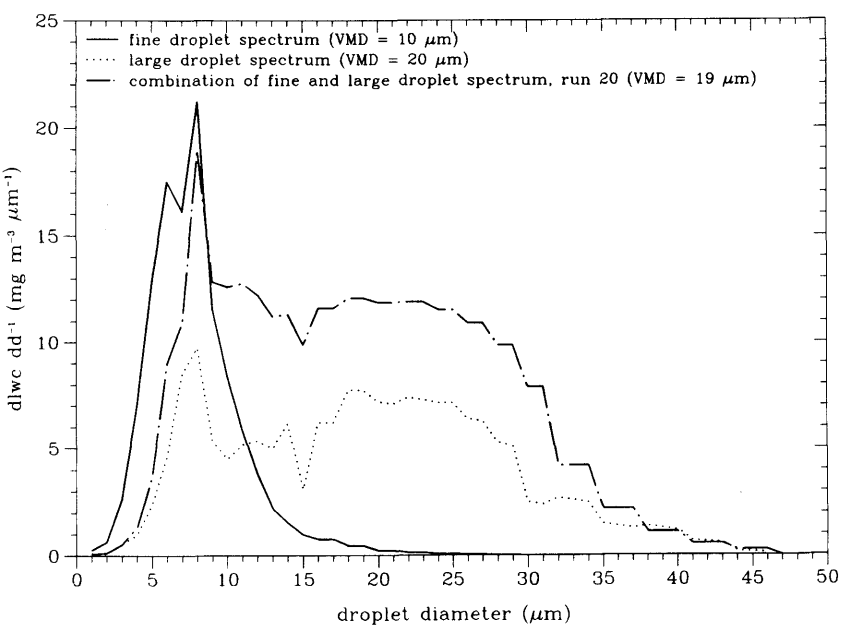

Fig. 3. Examples for the used droplet spectra in CHIEF. Fine droplet spectrum with a VMD of 10 μm, large droplet spectrum with a VMD of 20 μm and a combination of both spectra (run 20) with a VMD of 19 μm.

Table 1. *Comparison of sampled liquid water content measured by the fogwater collectors at different windspeeds and droplet spectra*

Run ID	1	2	4	5	6	7	8
windspeed ($m\ s^{-1}$)	2.1	3.6	2.1	2.1	2.1	2.1	5.4
VMD (μm)	8	8	20	20	20	10	8
LWC PVM ($mg\ m^{-3}$)	100.0	81.7	125.5	121.6	121.8	154.0	62.2
MOHnew ($mg\ m^{-3}$)	40.5	27.4	89.4	81.9	80.7	50.7	24.2
MOHold ($mg\ m^{-3}$)	26.7	21.5	76.5	75.2	—	—	—
IEP ($mg\ m^{-3}$)	—	—	—	—	42.7	84.6	23.1
MOHnew/PVM	0.41	0.34	0.71	0.67	0.66	0.33	0.39
MOHold/PVM	0.27	0.26	0.61	0.62	—	—	—
IEP/PVM	—	—	—	—	0.35	0.55	0.37
remarks							

Run ID	9	10	11	12	13	14	15
windspeed ($m\ s^{-1}$)	5.4	5.4	5.4	5.4	5.4	5.4	10.0
VMD (μm)	8	17	17	17	17	10	8
LWC PVM ($mg\ m^{-3}$)	59.3	44.6	81.4	85.8	77.8	64.3	116.0
MOHnew ($mg\ m^{-3}$)	20.7	20.5	55.3	31.9	—	—	11.0
MOHold ($mg\ m^{-3}$)	—	—	—	—	—	—	—
IEP ($mg\ m^{-3}$)	19.0	9.9	7.8	25.2	11.3	25.7	28.9
MOHnew/PVM	0.35	0.46	0.68	0.37	—	—	0.09
MOHold/PVM	—	—	—	—	—	—	—
IEP/PVM	0.32	0.22	0.1	0.29	0.15	0.4	0.25
remarks	a	a	c	a, b, c			a

Run ID	16	17	18	19	20	21
windspeed ($m\ s^{-1}$)	10.0	15.0	15.0	2.1	0.8	2.1
VMD (μm)	15	15	8	14	19	14
LWC PVM ($mg\ m^{-3}$)	130.0	154.0	118.0	175.0	328.0	162.0
MOHnew ($mg\ m^{-3}$)	44.0	53.2	28.2	88.3	161.0	85.0
MOHold ($mg\ m^{-3}$)	—	—	—	—	—	—
IEP ($mg\ m^{-3}$)	13.8	6.6	20.1	66.9	163.0	81.0
MOHnew/PVM	0.34	0.35	0.24	0.5	0.49	0.52
MOHold/PVM	—	—	—	—	—	—
IEP/PVM	0.11	0.04	0.17	0.38	0.5	0.5
remarks	a	a	a			

Flow rates: IEP: $67\ m^3\ h^{-1}$, MOH: $125\ m^3\ h^{-1}$. Remarks: (a) MOH sampler was operated with isokinetic inlet, (b) MOH sampler was turned 30° out of the wind, which possibly blocked the air in front of the IEP collector, (c) flow rate IEP: $71\ m^3\ h^{-1}$.

The 1st 4 tests were performed only with the 2 MOH impactors. The effect of an isokinetic inlet in front of MOH-new was tested as well as the effect of turning the instrument 30° out of the winddirection. In runs 19 to 21, 2 different salt solutions were nebulized. One solution, which was nebulized as small droplets contained K^+, the other one contained Na^+ and was nebulized as larger droplets. Samples were taken with the IEP and the MOH-new collector and the concentrations of the resulting samples were compared.

Additionally, filter measurements to determine the liquid water content (LWC) were carried out. The droplet spectra were measured using two FSSP-100 Forward Scattering Spectrometer Probes (Knollenberg, 1972; Baumgardner, 1983) mounted side by side. As the FSSPs were arranged in the rear of all other instruments in the tunnel,

some influence of the fog samplers on the measured spectra was possible. To generate windspeeds of 10 and 15 m s^{-1}, the cross section of the tunnel was reduced, so that an influence of one instrument to another could not be excluded.

The calibration of all instruments to measure the liquid water content and the droplet mass distribution is described by Arends et al. (1992). The FSSPs were calibrated with monodisperse glass beads, which led to a correct classification of the droplets into the corresponding size ranges. The number of droplets per range, however, could not be determined in this manner. Former measurements showed, that the FSSP overestimates LWC for droplets with diameters $>20\ \mu$m, which leads to an overestimation of the LWC for spectra with volume median diameters of $20\ \mu$m by about 30%. The PVM-100 was calibrated by comparison to light extinction measurements in the factory. An additional calibration was carried out by ECN, so that the PVM-100 turned out to perform the most reliable measurements of LWC. The amount of water in the individual droplet classes measured by FSSP-100 were therefore corrected by the data obtained by the PVM-100.

In a 2nd calibration procedure, the MOH-new collector was tested in CHIEF using a spinning-disk to generate a monodisperse aerosol to obtain information on the cut-off characteristics of the tested collector. In several runs, wet aerosol particles and small droplets of given diameters and K^+-concentrations were generated and collected on the impaction surfaces. The collection time was kept short enough to prevent any water from running off the surface, so that all of the collected particles remained on the plate. After each experiment, the deposited salt was washed off with deionized water and the solutions were analyzed. The K^+-concentrations in the air were determined by a filter method and were used as a reference for the concentrations found on the impaction surfaces. The concentration ratio represents the collection efficiency in the corresponding droplet size class.

3.2. *Results of the CHIEF experiments*

The results of the tests for the sampling volume can be taken from Table 1, where the absolute LWC determined by the PVM-100 is compared with the amount of water collected by the fog samplers. For a better comparison, the relative values of the fog collector data compared to the LWC in the tunnel are given also in this table. From this data it can be seen, that for low windspeeds the MOH collector samples more larger droplets, while the IEP collector samples the smaller droplets with a higher efficiency (Run 7). Run 6 with low windspeeds and a droplet spectrum with a volume median diameter (VMD) of $20\ \mu$m, as well as most of the other runs, showed a remarkable difference in the collection efficiency of both collectors with a lower efficiency for the IEP collector. This can be explained by the different inlet efficiencies due to the different geometries of the collectors. To enter the IEP-collector, the droplets have to be bent off by 90°, so that a combination of low windspeeds and small droplets will increase the collection efficiency (run 7). The MOH collector with its vertical inlet therefore has less problems if it is directly oriented into the wind. If not, the MOH collector also had problems to reach its optimal sampling efficiency. Run 12 was performed to verify the above hypothesis. The MOH collector was turned out of the wind with an angle of 30° resulting in a 30% lower collection efficiency compared to run 11, which was performed under the same conditions but with the inlet oriented to the wind. A summary of the results described above is given in Fig. 4, which shows the measured collection efficiencies for both collectors as a function of wind velocity and VMD. The areas of the circles represent the collection efficiencies.

As there were no direct intercomparisons performed between the collectors which were operated side by side during the field campaign (MOH-old and IEP), the first four runs give an idea of improvements of the new MOH collector. With MOH-new, collection efficiencies are about 9% higher compared to the old device, independent of the given droplet spectra or windspeeds. As described above, the changes of the MOH collector were mainly accomplished to improve the water flow from the impaction stage into the bottles. The detected losses of the old collector could then be identified as after stage losses concerning all droplet sizes in the same way.

One experiment was carried out to get a rough quantification of the mass of water which was already lost in the inlet and behind the stage of the MOH collector. Therefore a tissue was placed on

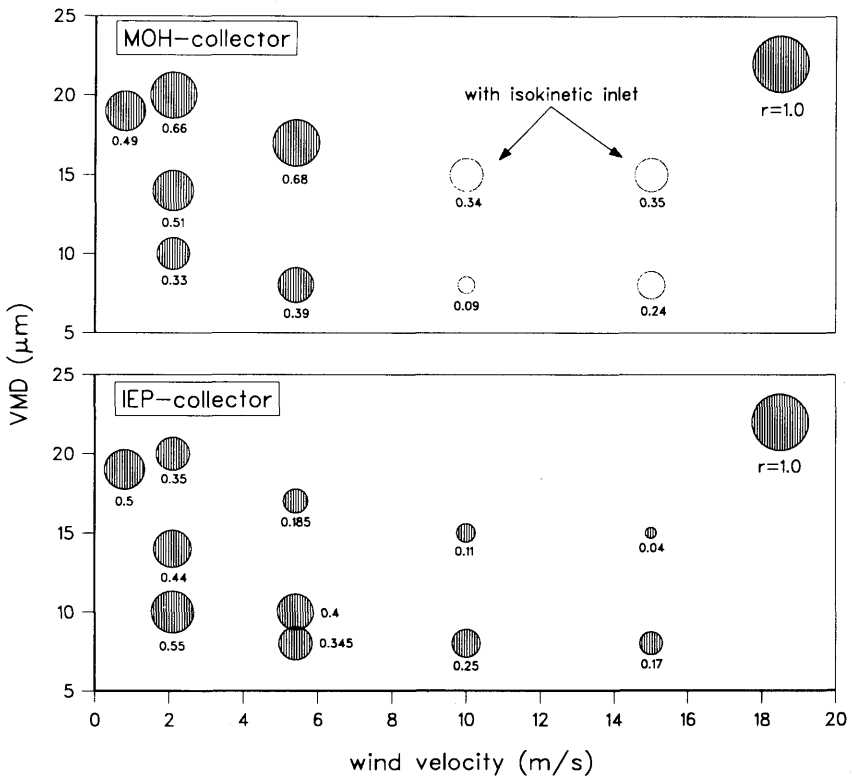

Fig. 4. Collection efficiencies as a function of wind velocity and VMD of both collectors. The areas of the circles represent the collection efficiencies.

the walls and weighed after collection. The results show, that a considerable amount of water can be found behind the impaction plate (15%) and a smaller part in the inlet (6%). 56% of the water was found in the sample vial and on the plate. The remainder of 23% must be attached to errors in the measurements, to droplets which passed the collector without impacting on any obstacle and to droplets which were blown off after collection. This experiment was done only with the large droplet spectrum (see Fig. 3).

Unfortunately, similar tests with the IEP collector were not performed, but the observations in CHIEF as well as under field conditions showed, that droplets also were impacted at the inlet walls. But it can be excluded, that these droplets could enter the sample vials, as they run along the nozzle walls before they were blown off without reaching the impaction plate.

Condensation or evaporation processes do not seem to be a problem for both collectors. Table 2 shows the comparison of potassium concentrations in the fog samples and the concentrations of K^+ before the solution was nebulized. No major changes in the concentrations could be observed. In most cases, the concentrations in the samples were lower than in the generated solutions. From that it could be expected, that condensation occurred during the collection of the droplets. The problem of condensation of supersaturated water vapour in an impaction stage was discussed in detail by Berner (1988). It was shown, that the residence time in an impaction stage is too short for large amounts of vapour to condense on particles or walls. More serious seems to be the problem of evaporation, if the water is removed not fast enough from the gas flow into the collection bottles. As the described experiment showed no higher concentrations in the samples of the collectors, evaporation seems not to occur during

Table 2. *Comparison of the K^+-concentrations in the samples compared to the nebulized solution in mg l^{-1} and the relative amounts of recovered K^+ in the samples*

Run ID	1	2	4	5	6	7	8	9
generator	107	113	113	113	104	108	108	109
MOHnew	81	91	87	101	98	84	93	101
MOHold	84	93	97	—	—	—	—	—
IEP	—	—	—	—	100	85	90	93
MOHnew/gen.	0.76	0.81	0.77	0.89	0.94	0.78	0.86	0.93
MOHold/gen.	0.79	0.82	0.86	—	—	—	—	—
IEP/gen.	—	—	—	—	0.96	0.79	0.83	0.85
Run ID	**10**	**11**	**12**	**13**	**14**	**15**	**16**	**17**
generator	106	105	106	105	105	105	108	104
MOHnew	99	96	102	99	—	100	96	95
MOHold	—	—	—	—	—	—	—	—
IEP	98	97	97	129	100	89	98	107
MOHnew/gen.	0.93	0.91	0.96	0.94	—	0.95	0.89	0.91
MOHold/gen.	—	—	—	—	—	—	—	—
IEP/gen.	0.92	0.92	0.92	1.23	0.95	0.85	0.91	1.03

the sampling of fog. The observed lower K^+ concentrations in the samples compared to the stock solution may then be caused by water droplets which escaped from the humidifier and diluted the samples.

Table 3 gives the results of the experiments (runs 19 to 21) when two different solutions have been nebulized. The K^+ solution was sprayed with a small droplet spectrum (VMD = 8–10 μm) whereas Na^+ was nebulized with a large droplet spectrum (VMD = 17–20 μm). The experiments were carried out at low windspeeds to simulate the conditions in the Po Valley fog. The comparison of the concentrations of K^+ and Na^+ between both collectors shows, that the IEP collector samples more small droplets compared to the MOH collector, which led to higher K^+ concentrations, and also sampled less large droplets. After run 21, also the inlets of both collectors were washed off and the salt solutions were analyzed. From the resulting data it can be suspected, that preferentially large droplets impact on the inlet walls of the MOH collector because the concentration was found to be twice the concentration of the stock solution (Table 3). Smaller droplets did not seem to suffer from inlet losses. The same experiments with the IEP-collector showed a higher amount of smaller droplets impacting on the inlet walls, whereas nearly the same amount of larger droplets compared to the MOH-collector was found there.

It should be mentioned, that this experiment only gives a rough idea of the characteristics of the collectors since a lot of problems due to coagulation processes or droplet losses in the wind tunnel can occur, which cannot be quantified. Therefore

Table 3. *Comparison of K^+- and Na^+-concentrations found at the impaction stages and the inlets of both collectors. K^+ was sprayed with a fine droplet spectrum, Na^+ with a coarse droplet spectrum*

Run ID		19	20	21
Solution 1:	K^+, ppm	94	95	104
fine droplets	Na^+, ppm	0.1	0.1	0.2
Solution 2:	K^+, ppm	0.2	<0.1	<0.1
coarse droplets	Na^+, ppm	31	31	32
concentration	K^+, ppm	47	48	52
of solution	Na^+, ppm	16	18	16
MOH:	K^+, ppm	23	19	17
	Na^+, ppm	19	20	22
IEP:	K^+, ppm	44	30	31
	Na^+, ppm	15	17	21
MOH inlet:	K^+, ppm	—	—	6
	Na^+, ppm	—	—	34
IEP inlet:	K^+, ppm	—	—	21
	Na^+, ppm	—	—	27

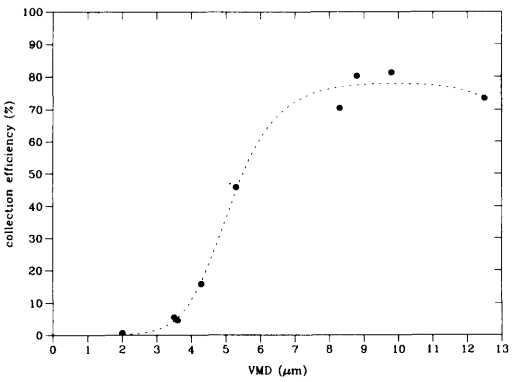

Fig. 5. Results of the calibration of the MOH collector with monodisperse aerosol.

care should be taken to calculate sampling efficiencies from this data.

Recent experiments in CHIEF, using a monodisperse aerosol as described in Subsection 3.1, gave much better information on the cut-off characteristics of the tested fogwater collectors. Fig. 5 shows the results of the experiments. From that, the MOH-new collector can be characterized by a 50% cut-off diameter of 6 μm, an 80%-efficiency for droplets in the size range of 9 to 11 μm, and possibly a decreasing efficiency for droplets >11 μm. These experiments were performed isokinetically.

4. Comparison of the results of the field measurements

4.1. Differences in measured liquid water content

The liquid water content, determined by the two collectors, showed remarkable differences compared to each other over the whole field campaign. The sampling time of the two collectors was set to 1 hour which led to sample volumes typically in the range between 3 and 12 ml. Fig. 6 shows the temporal variations of the LWC measured by the two fogwater collectors and the PVM-100 (1 hour mean values) from 11 to 14 November. Within the first half of the described period, the IEP-collector showed slightly higher sampled LWCs compared to the MOH-collector with short periods showing the opposite behaviour. This changed at 12 November, 2100. Beginning from that point, the IEP-collector had the tendency to collect more water. These 2 periods can mainly be characterized by the observed droplet spectra,

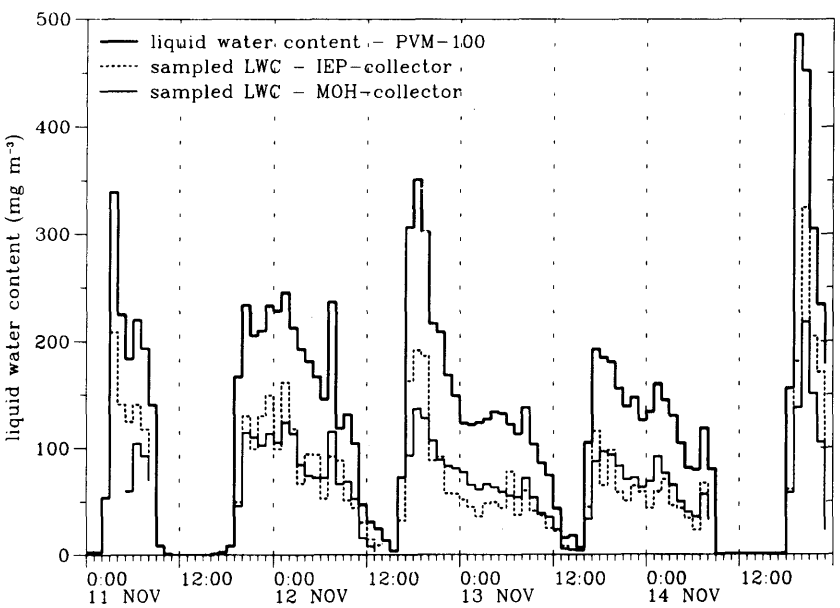

Fig. 6. Comparison of sampled liquid water content between the two fogwater collectors and LWC measured by PVM-100.

which were obtained by FSSP and the mean windspeed. In agreement to the results of the wind tunnel tests, the IEP collector had a higher collection efficiency, when the droplet spectra were shifted towards the smaller droplets and the windspeed was relatively low. A comparison of the collection efficiencies together with the one hour mean values of the volume median diameters is given in Fig. 7. The most remarkable period, which illustrates this behaviour very clearly, was from 1700 of 12 November to the morning of 13 November. The comparison of LWC and VMD shows an immense increase of the liquid water content for a period of 3 h, while the volume median diameter was relatively low, with the consequence, that a large number of small droplets was present in the fog of this period. Towards midnight, the liquid water content decreased, while the VMD continued to increase which means a decrease of droplet number concentrations but an increase of the number of larger droplets. This behaviour was reflected very clearly by the two fogwater collectors, showing contrary slopes of their collection efficiency curves in this period (Fig. 7). While a large amount of small droplets was present, the IEP-collector sampled with an efficiency of 60% (maximum), whereas the MOH collector only reached 40% in the same hour. During the presence of large droplets the MOH-collector reached a maximum collection efficiency of also about 60%, while the efficiency decrease of the IEP-collector stopped at about 30%.

Another remarkable difference between both collectors becomes noticeable while analyzing the variability of the collection efficiencies. The variations of collection efficiencies in two consecutive hours are much higher in the IEP-collector compared to the MOH-collector. Since the windspeed at the ground as well as the droplet mass distribution did not show such high variabilities, these large variations could either be explained by a very strong sensitivity of the IEP-collector due to small changes in the windspeed (see Subsection 5.1) or by possible errors originating in the handling of the collected samples.

4.2. *Differences in chemical data*

The chemical analyses of the samples of both collectors as well showed large differences for each chemical component analyzed during the field campaign. For the comparison, the mean values of the main components were calculated from the results of both collectors for a particular sampling period.

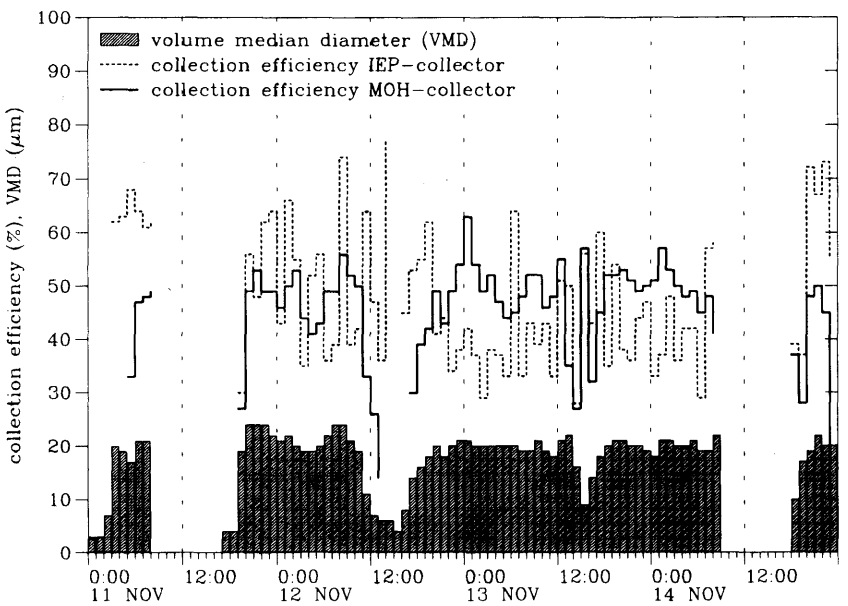

Fig. 7. Collection efficiencies of both collectors and volume median diameter of the droplet spectrum.

Subsequently the differences in the concentrations were related to the mean values (eq. (1)), so that positive values of D indicate higher concentrations in the samples of the IEP- and negative values higher concentrations in the samples of the MOH-collector.

$$D = (C_{IEP} - C_{MOH}) \times 100/C \quad (1)$$

with:

D: difference in solute concentrations (%)
C_{IEP}: concentration in the samples of IEP-collector
C_{MOH}: concentration in the samples of MOH-collector
C: mean concentration of both collectors.

Only those samples were taken for a comparison which satisfied the following restrictions:

(a) the chemical analyses of both collectors must be available for the collection intervals under comparison (necessarily);

(b) the percent differences (PD) of ionic balance, calculated as

$$PD = \frac{100 \times (\sum [A^-] - \sum [C^+])}{(\sum [A^-] + \sum [C^+])},$$

must be lower than 17% (Fuzzi et al., 1992).

These restrictions were fulfilled for the most components of 27 samples. Table 4 gives an overview of the calculated differences in the chemical analyses of the main components $SO_4^=$, NO_3^+, and NH_4^+ by comparing the mean values, maxima, minima and standard deviations. The values of the absolute concentrations averaged over the results of both collectors are given in brackets. The mean values of the absolute differences between the two collectors are 192, 284, and 566 μeq/l for $SO_4^=$, NO_3^-, and NH_4^+ respectively. $SO_4^=$ and NO_3^- were analyzed by ion chromatography, NH_4^+ by means of an ion-selective electrode. A more detailed description of the chemical analyses is given in Fuzzi et al. (1992).

The time series of the observed differences are shown in Fig. 8. It can be seen, that in most of the cases the samples of the IEP collector were higher concentrated. The largest difference with higher concentrations in the samples of the IEP-collector was found in the morning hours of 13 November. Due to the microphysical changes in the period described above, also the variations in the chemical data showed a remarkable behaviour since they confirm the assumption of a higher collection efficiency for smaller droplets of the IEP-collector.

The higher concentrations in the samples of the MOH-collector in the morning hours of 12 November could not be explained in a satisfactory way by changes in windspeed or changing volume median diameters. The comparison of the data with the measured liquid water contents in this period (Fig. 6) gives only little indication, that this behaviour could be governed by lower LWC eventually leading to some evaporation of the smaller droplets.

Table 4. *Differences in the chemical analyses of the two collectors for the main components sulfate, nitrate and ammonium (in percent), averaged over all observations; the mean values of the absolute concentrations in the samples of both collectors (in μeq/l), averaged over all observations, are given in brackets*

	$SO_4^=$		NO_3^-		NH_4^+	
mean	21.8	(882)	24.1	(1180)	21.4	(2647)
max	100.7	(3535)	123.9	(4407)	70.9	(9542)
min	−60.9	(208)	−80.9	(164)	−68.5	(756)
n	27	(54)	27	(54)	27	(54)
stddev	28.6	(560)	43.0	(974)	28.3	(1613)

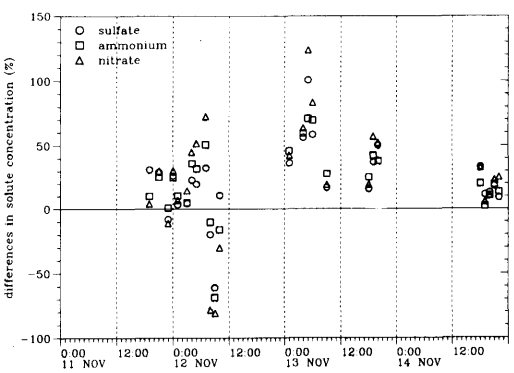

Fig. 8. Differences in the solute concentrations of the main components in the samples of both collectors, in percent.

5. Performance calculations for both collectors

The amount of liquid water collected by the two samplers can be simulated for every time step with the knowledge of the following parameters: droplet size distribution, liquid water content, windspeed, aspiration- or inlet efficiency and collection efficiency. A comparison of the results of the simulations with the field data can give useful hints to sensitivities of both collectors originating from environmental changes.

The total collection efficiencies (E_t) of fogwater collectors are first of all governed by their inlet efficiencies. Since it is known from the wind tunnel experiments, that the collection efficiencies of both collectors are dependent on the windspeed and VMD (Fig. 4), this effect has been quantified.

5.1. Calculation of aspiration efficiencies

The chief requirement for aerosol measurements is the representativeness of the sample, i.e., the identity of the sample with the aerosol from which it was taken, with respect to concentration, droplet size distribution, chemical composition etc. To obtain representative samples from a moving airstream, a number of conditions must be fulfilled (Fuchs, 1975):

(a) the axis of aspiration should be parallel to the wind direction (condition of isoaxiallity), otherwise the particle concentration C_i in the sample would be less than in the surrounding atmosphere C_a and the *aspiration coefficient* $A = C_i/C_a$ would be less than 1;

(b) the mean flow velocity in the probe U_i should be equal to the wind speed U_a (condition of isokinesis), otherwise, at $R = U_a/U_i > 1$, the flow lines diverge at the inlet and some large particles, that were not originally in the gas volume sampled, will travel into the probe and be included in the sample with the result of an overestimation of particle concentration ($A > 1$) (Hinds, 1982);

(c) the walls of the inlets should be infinitely thin, i.e., in practice the edges should be sharp or only very slightly rounded, otherwise droplets entering the stagnant zone in front of the edge under inertial force can penetrate into the probe leading to an increase of the aspiration coefficient A.

Strict isokinetic conditions are difficult to maintain in practice. Therefore it is necessary to know which error results from given deviations of R from unity and nonisoaxial sampling and/or using "blunt" samplers.

Based on the studies of Durham and Lundgren (1980), Vincent et al. (1986) developed a physical model which takes account of the fact, that the sampled air not only diverges or converges (depending on the relationship between wind speed and sampling velocity), but also turns to pass through the plane of the sampling orifice. Additional to their model investigations they carried out experiments in a large wind tunnel to investigate the aspiration efficiencies. Sampling conditions ranged from sub- to super-isokinetic at orientations with respect to the wind direction ranging from 0 to 180°. Finally they obtained the following empirical equation for the aspiration efficiency:

$$A = 1 + [1 - \{1/(1 + k\,St(\cos\theta + 4R^{1/2}\sin^{1/2}\theta))\}][R\cos\theta - 1] \quad (2)$$

with:

A: aspiration efficiency
St: Stokes number referred to air speed and probe diameter
θ: angle of probe orientation
R: ratio of velocities, $R = U_a/U_i$
U_i: sampling velocity
U_a: freestream air velocity
k: empirical coefficient, $k = 1.8$

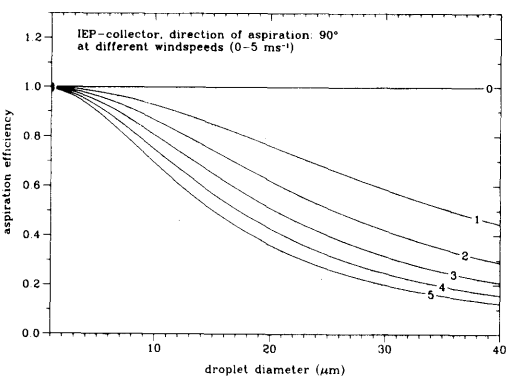

Fig. 9. Aspiration efficiency calculated for the IEP-collector for 6 different windspeeds from 0 to 5 m s^{-1}.

For $\theta = 0°$ and $R = 1$, eq. (2) yields the familiar theoretical result for isokinetic sampling, namely $A = 1$, independent of wind speed, sampling flow rate, probe dimensions and particle aerodynamic diameter.

For $\theta = 90°$, eq. (2) reduces to

$$A = 1/\{1 + 4k\, St\, R^{1/2}\}. \quad (3)$$

Applying these calculations, one has to take into account that in practice both collectors are far from having the ideal inlets, these calculations rely on. Nevertheless, an estimate of the importance of the effects which influence the total collection efficiencies is obtained.

Some simplifications were made describing the cross section of the inlets. Since the 2 collectors have 2 and 4 orifices, respectively, their opening areas were added which led to the assumption of having one inlet for each collector. Furthermore, no distinction has been made between rectangular and round inlets. Fig. 9 shows the aspiration efficiencies at different windspeeds from 0 to 5 m s^{-1} for the IEP-collector. When sampling from still air, there is no decrease of A towards larger droplets, whereas a windspeed of 1 m s^{-1} allows only 60% of droplets with diameters of 30 μm to enter the inlet. The MOH-collector does not show such drastic effect, since its direction of aspiration is in the plane of the wind. Nevertheless, also for this instrument the aspiration efficiencies are lower than unity for windspeeds below 2.4 m s^{-1} (Fig. 10), which is the inlet velocity of this device. Since the windspeeds at the ground were mostly below 2 m s^{-1} during the field experiment it can be stated, that both collectors sampled droplets with an aspiration efficiency less than 1. Fig. 11 shows the effect of turning the MOH-collector out of the wind. For windspeeds below 3 m s^{-1} the aspiration efficiencies decrease with increasing droplet diameters, whereas for higher windspeeds they remain as high as for a properly aligned instrument. The disagreement between calculation and the results of the wind tunnel experiments can be explained by the fact, that the amount of water, which first enters the cross-sectional area of the inlets and subsequently impacts on the walls of the inlet, is not quantified by the calculations.

5.2. *Simulation of LWC*

For the simulation of the amount of water sampled by the fogwater collectors during the field experiment, the following data were used:

(a) 1-h mean values of the windspeed to calculate the aspiration efficiency in each sampling period;

(b) 1-h mean values of the droplet size distributions which were measured with a FSSP-100 (Arends et al., 1992);

(c) 1-h mean values of the liquid water content measured with a PVM-100 (Arends et al., 1992);

(d) information on the collection efficiencies of the fogwater collectors (cut-off diameter, wall losses, after stage losses).

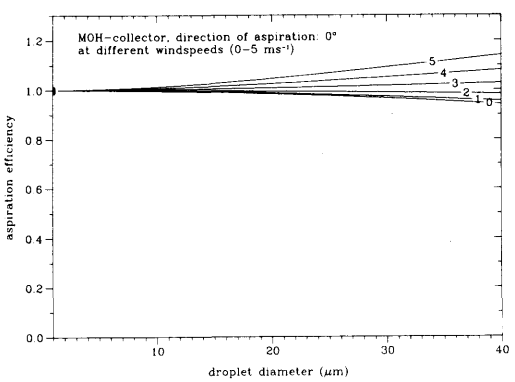

Fig. 10. Aspiration efficiency calculated for the MOH-collector for 6 different windspeeds from 0 to 5 m s^{-1}.

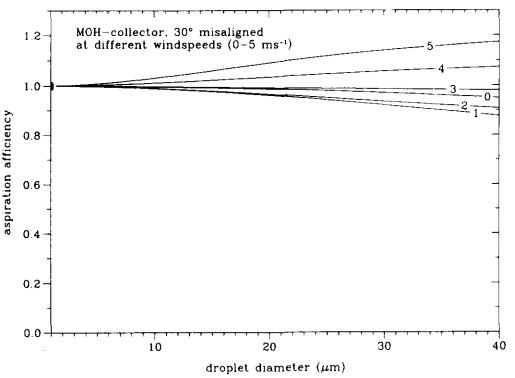

Fig. 11. Aspiration efficiency calculated for the MOH-collector assuming a misalignment of 30° for 6 different windspeeds from 0 to 5 m s^{-1}.

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The simulated liquid water content for each collector can be calculated as follows:

$$LWC_c = \sum_d (M(d)\, A(d, w, B)\, E(d)), \qquad (4)$$

with:

LWC_c: simulated liquid water content calculated for each fogwater collector in 1-h time steps;

$M(d)$: 1-h mean value of water mass as a function of droplet size d taken from FSSP and PVM-100 measurements;

$A(d, w, B)$: aspiration efficiency as a function of droplet size, 1-h mean values of the actual windspeed w, and the collector characteristics B

$E(d)$: collection efficiencies of the collectors.

The factor $E(d)$ in eq. (4) combines several effects which influence the amount of collected water in the samplers. First of all it represents the cut-off diameter, which can be calculated for each collector or determined by a calibration procedure (see Subsection 3.2). From this, however, possible losses on the walls and behind the stages of the impactors cannot be quantified analytically. To find out proper collection efficiencies as a function of droplet diameter $E(d)$, the simulations were started with an efficiency curve containing all available information on the cut-off characteristics of each collector, including the results of the wind-tunnel tests. By comparing the results of the simulation with the measured values, the efficiency curves were modified iteratively until a good fit between measured data and simulation was achieved.

Fig. 12 shows the resulting total collection efficiencies ($E_t = AE$) of both collectors as a function of droplet diameter and wind speed. This first result from the simulations confirms the interpretations of the results of the wind-tunnel tests and field measurements. The IEP-collector is very sensitive to changes in windspeed, since the total collection efficiency decreases very drastically for higher windspeeds. Additionally, this collector samples only a small band out of the droplet spectra with the maximum droplet diameter of 28 μm. The maximum collection efficiency of 90% is reached for droplets in the range between 12 and 18 μm. The MOH-collector is not that sensitive for changing conditions, but reaches for all windspeeds only a collection efficiency of about

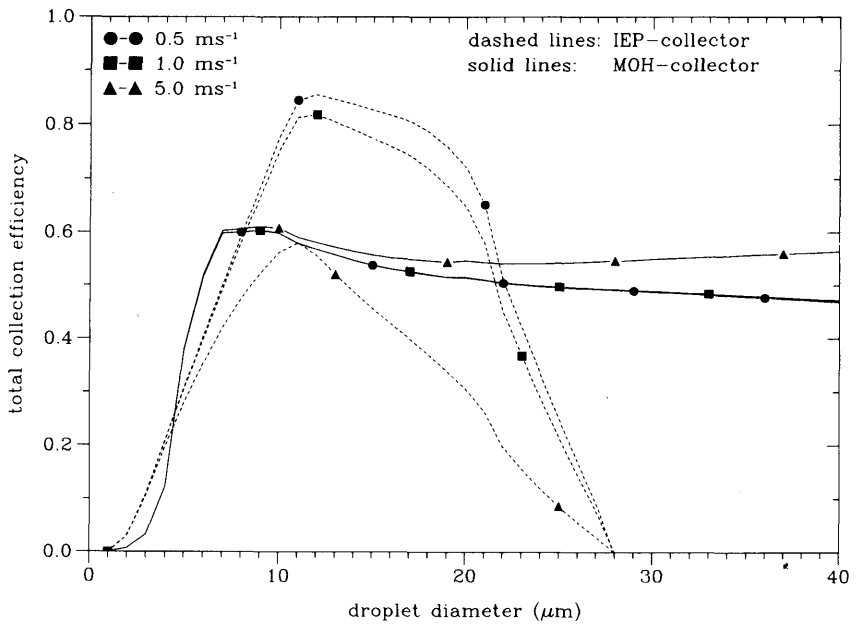

Fig. 12. Total collection efficiencies of both collectors for 3 different windspeeds.

60% for droplets in the range 7 to 10 μm. The decrease of the efficiency for larger droplets is very low and it can be stated, that this collector samples fog droplets over the whole spectrum with an efficiency of 50–60%. The 50% cut-off diameter of the MOH-collector is about 6 μm. The cut-off diameter of the IEP-collector varies between 7 and 8.5 μm, dependent on the windspeed.

Figs. 13 and 14 show the comparison of the results of the simulations with the measured values of the liquid water content for each collector. In general, the curves correspond to each other very well. If this is not given, the measured liquid water content must have been influenced by additional parameters which were not considered in the simulation. This could be possible errors in the handling or weighing of the probes as well as a summation of errors of each individual measurement. Since the MOH-collector has to be adjusted into the wind by hand, the instrument certainly was misaligned for shorter periods of time. Further sources of error could be the deterioration of the IEP samples by rain or drizzle, what however can be ruled out for the investigated fog events. Periods with higher LWC in the IEP collector could also result from increased settling of bigger droplets. This possible effect has not been accounted for the simulations. Evaporation of the droplets after impaction could also be a possible source of error. Since the liquid is pushed off the plates almost immediately after collection, this error was minimized as described in detail for the IEP collector by Berner (1988). The same argument holds for the MOH collector since the velocity in the nozzles is nearly the same and the collected water is also pushed off the plates immediately.

5.3. *Simulation of the chemical analyses*

Going one step further, the solute concentrations in the samples of each collector were simulated. It was shown in the previous section, that the 50% cut-off diameter of the MOH-collector is somewhat lower compared to the cut-off diameter of the IEP-collector. The shape of the supposed efficiency curve in this size region must rely on the calculations and calibrations for each collector since the contribution of the droplets up to a diameter of about 7 μm is too low to cause big changes in LWC. Changes in the efficiency curves in this size region will therefore cause only changes in the solute concentrations of the samples if a size dependency of the droplet composition is considered. Several theoretical and experimental studies were performed to examine this problem

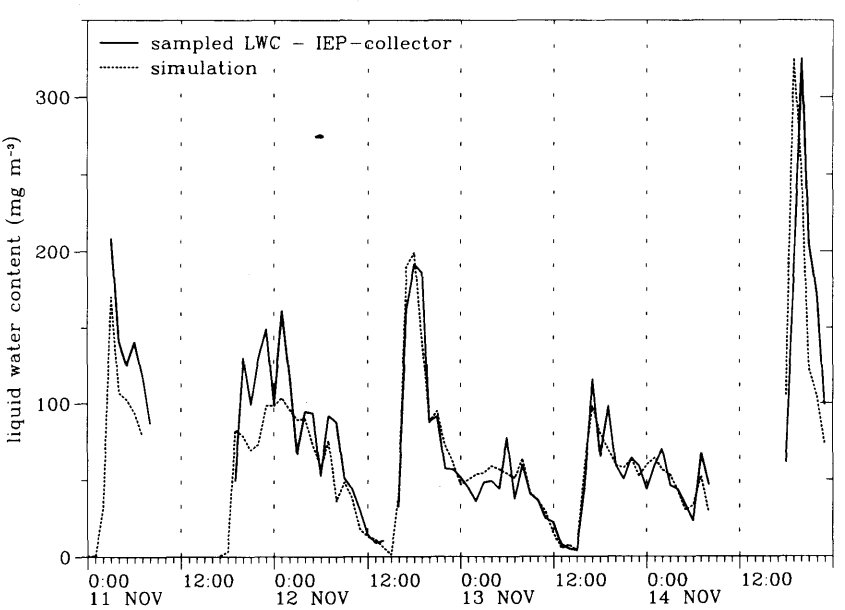

Fig. 13. Comparison between measured and simulated liquid water content for the IEP-collector.

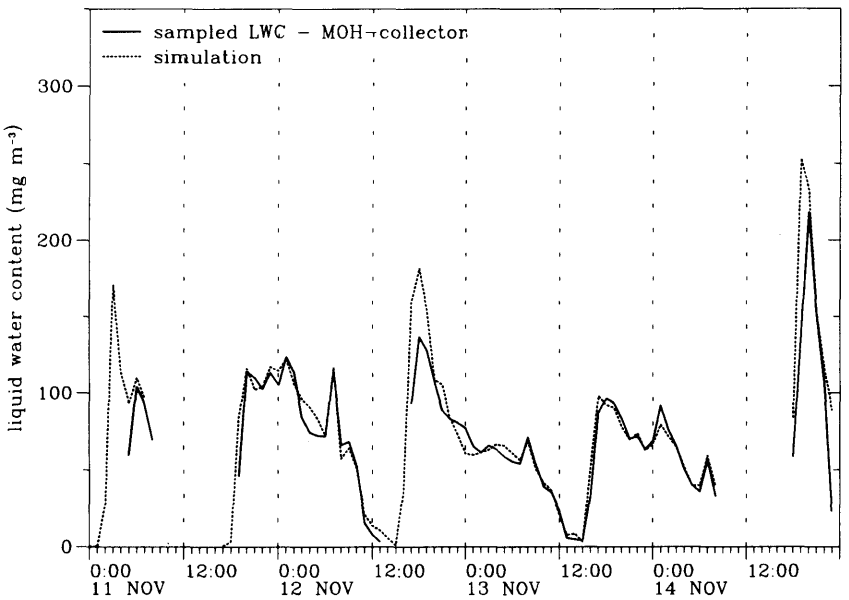

Fig. 14. Comparison between measured and simulated liquid water content for the MOH-collector.

(Flossmann et al., 1985; Pandis et al., 1990; Seidl, 1989; Noone et al., 1988; Ogren et al., 1989; Munger et al., 1989; Schell and Georgii, 1989). Also during the Po Valley Fog Experiment 1989 the size dependence of cloudwater composition was studied.

Fig. 15 shows the results of measurements of the size dependence of the concentration of non-volatile material in fog droplets, which are discussed in detail by Ogren et al. (1992). The data were obtained with a counterflow virtual impactor (CVI) (Noone et al., 1988; Ogren et al., 1989) and an eight-stage aerosol impactor operated at ambient humidities (Berner, 1989) in combination with a FSSP 100 (Fuzzi et al., 1992; Arends et al., 1992). Hereby the mass concentration of non-volatile material in the droplets was determined by the sum of the concentrations of the major ions $SO_4^=$, NO_3^-, and NH_4^+ deposited on the impactor stages covering the size ranges 1–2, 2–4, 4–8, and 8–16 μm, divided by LWC in the corresponding size ranges. The data points (triangles) in Fig. 15 represent the solute concentrations averaged over all observations during the field experiment, plotted against the mean diameter of the corresponding size ranges. CVI measurements to obtain the size dependence were performed by varying the cut-off diameter of the instrument systematically between 5 and 20 μm. Hence the CVI results in Fig. 15 (points) represent the solute mass concentrations of a broad size range plotted against the diameter of mean mass of the sampled droplets (Ogren et al., 1992). These results can be summarized by the following approximative equation:

$$C(d) = 4.77 \times 10^4 \times \exp[-0.65 \times d] - 0.95 \times d + 50. \quad (5)$$

Using this size dependence of droplet concentration and the total collection efficiencies of the fogwater samplers, the mean concentrations in the samples can be estimated. A comparison of the differences in the chemical analyses measured and simulated is shown in Fig. 16. The single points represent the measured differences in the sulfate concentrations, the solid line shows the output from the calculations. The simulation, as well as the experimental data show generally higher concentrations in the samples of the IEP-collector, but also periods of higher concentrations in the samples of the MOH-collector. Although the simulation up to now does not consider all possible effects ruling the chemical concentrations in the fogwater samples, it indicates that the assumptions

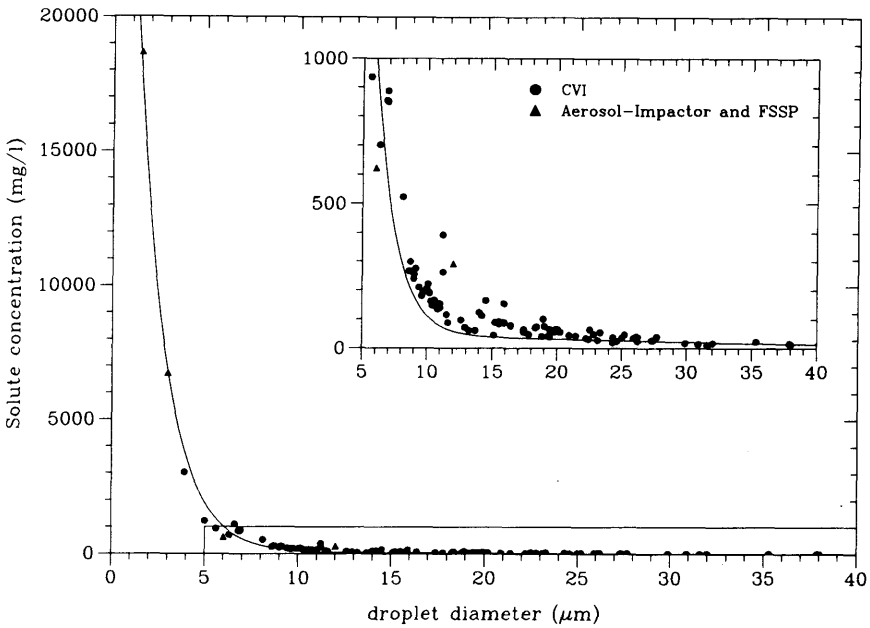

Fig. 15. Measured size dependence of droplet solute concentrations.

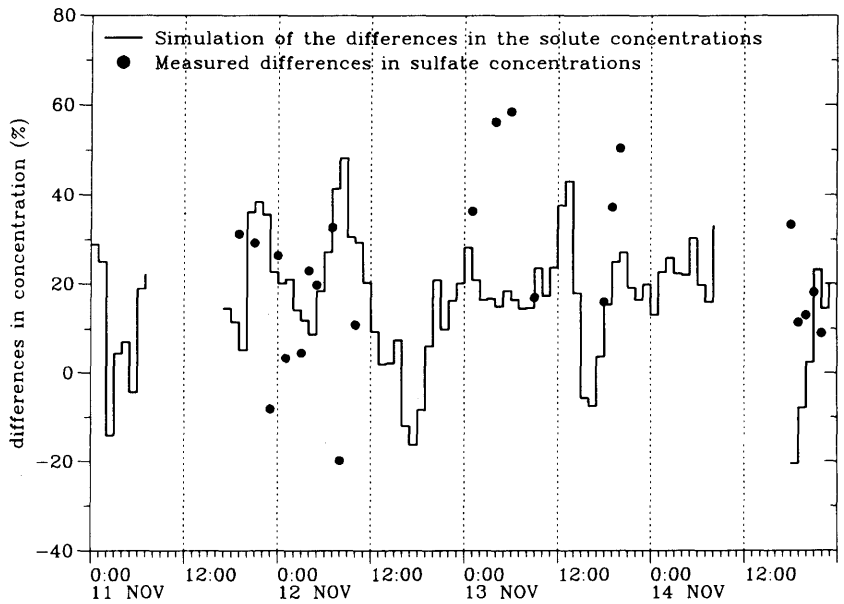

Fig. 16. Comparison between measured and simulated differences in the solute concentrations of both fogwater collectors.

made concerning the shape of the total collection efficiencies of both fogwater collectors are right. It becomes obvious, that the smaller cut-off diameter, calculated and measured for the MOH-collector, must not lead to higher concentrations in the samples if the size dependence of fogwater solute concentrations described above is assumed. The higher collection efficiencies for larger droplets of this sampler lead to a dilution and therefore to generally lower concentrations in the samples.

5.4. *Consequences for tower measurements*

Regarding tower measurements to obtain vertical gradients of fogwater solute concentrations, it is essential to know, to what extent observed differences between different levels derive from the sampling characteristics of the used collector or from microphysical/chemical processes. It was shown, that the collection characteristics of the IEP-collector are strongly dependent on the windspeed, whereas the MOH-collector is not influenced to such an extent by changing atmospheric conditions. Figs. 17 and 18 show the simulated fogwater concentrations for event 2 from 11 November to 14 at the ground and in 25 m for both collectors, which are based only on the measured size dependence of the solute concentrations, droplet spectra, windspeeds and the fog collector characteristics. To calculate the concentrations in 25 m it was supposed to have the same droplet size distribution and LWC as at the ground, so that the calculated differences derive only from the measured higher windspeeds in the 25 m level (Wobrock et al., 1992). It can be stated, that the concentrations in the samples of the MOH-collector are not supposed to vary very much due to higher windspeeds and that they will be equal or even about 10% lower in 25 m. On the contrary, the samples of the IEP-collector will be much higher concentrated in 25 m compared to the ground level (Fig. 18) if, as already stated, no vertical changes in droplet size distribution or LWC are assumed.

To compare the temporal variation of the solute concentrations simulated with the concentrations that were measured by the two collectors at the ground level, in Figs. 17 and 18 the sums of the major ions in the samples of the collectors are also shown. The simulations are based only on the results of the analysis of the size dependence (eq. (5)) and the temporal variations of the droplet size distributions. The very good agreement of the simulations with the measured values illustrates,

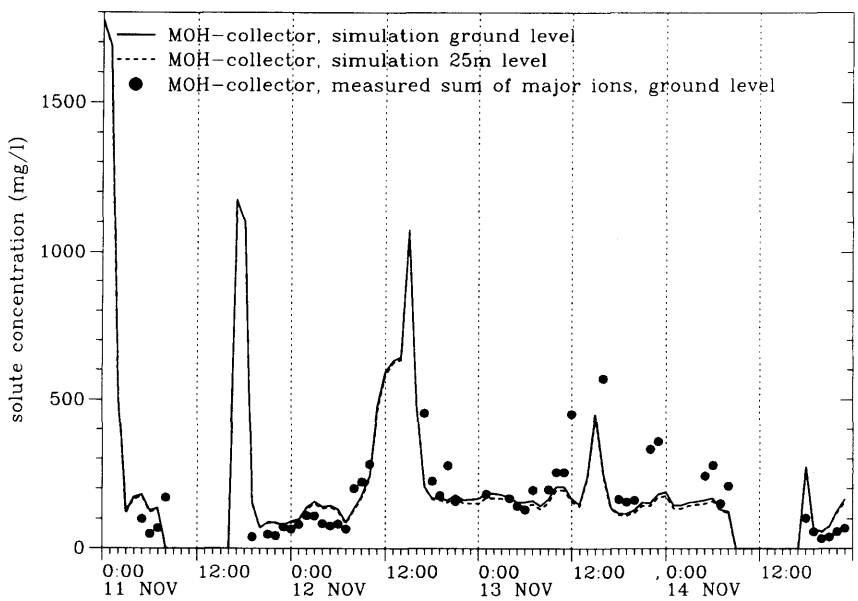

Fig. 17. Comparison between the simulated solute concentrations at the ground and in 25 m for the MOH-collector. The points represent the sum of major ions from the chemical analyses of the samples (ground level).

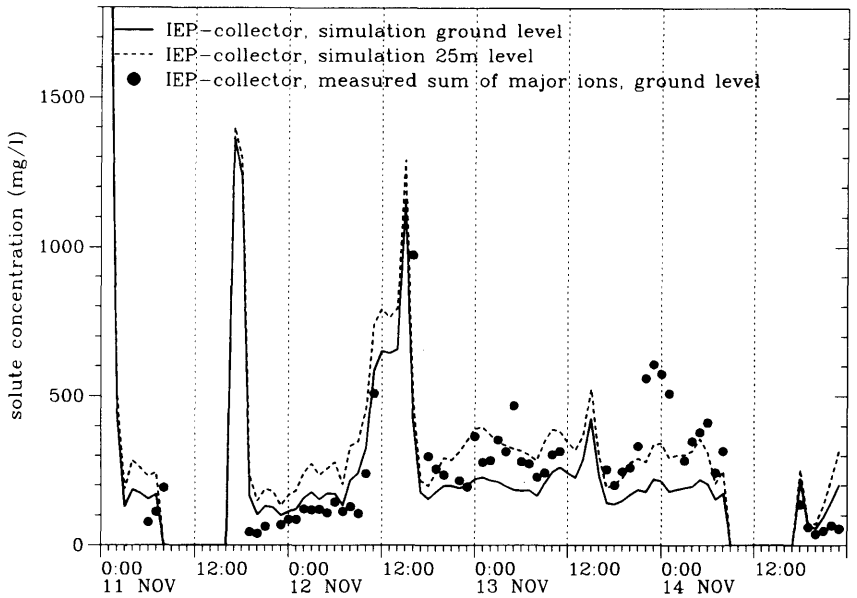

Fig. 18. Comparison between the simulated solute concentrations at the ground and in 25 m for the IEP-collector. The points represent the sum of major ions from the chemical analyses of the samples (ground level).

that changes in fogwater composition can be explained to a large extent by variations in liquid water content and droplet size distribution and that they must not necessarily derive from changing chemical conditions in the fog layer.

6. Conclusions

The intercomparison tests in CHIEF show, that condensation or evaporation processes seem not to be a problem for both collectors. The differences in the amount of sampled water are considerable and are correlated to the droplet size distribution and the wind velocity. Since only few direct intercomparisons were performed in CHIEF between the IEP-collector and the MOH-old, used during the campaign in Bologna, experimental data obtained in the field and model calculations additionally were used for the characterisation of the samplers.

The differences in collection efficiencies, as well as the differences in the chemical data between both collectors were shown to be specific responses of each collector due to the microphysical and meteorological conditions in the fog. Since the inlets of the IEP-collector are oriented horizontally at an angle of always 90° with respect to the wind, its collection efficiency is limited for bigger droplets and has the tendency to decrease for increasing windspeeds. Droplets with diameters >28 μm are not collected.

The MOH-collector does not respond to changing environmental conditions to such an extent if its inlets are always directed into the wind. The total collection efficiency of this sampler is in the range of 50–60% for all droplet diameters >8 μm and therefore somewhat lower in the range from 9 to 23 μm compared to the IEP-collector.

Considering the results of this investigation it can be stated, that both collectors are suitable instruments for the investigation of fog chemistry and microphysics if the expected windspeeds remain in the range of 1 m s^{-1}. For the interpretation of the results, the total collection efficiency curves of both collectors must be taken into account, because the chemical composition of the fogwater samples is strongly dependent on the amount of water which was collected in each droplet size interval.

Some modifications however could improve the collection characteristics of both collectors:

providing the MOH-collector with a wind vane could avoid losses due to possible temporal misalignments; providing both collectors with an appropriate arrangement to fit the inlet velocities to the actual windspeed to achieve isokinesis; improvements in the organisation of the water flow into the vials of both collectors would prevent from possible handling errors.

7. Acknowledgements

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REFERENCES

Arends, B. G., Kos, G. P. A., Wobrock, W., Schell, D., Noone, K. J., Fuzzi, S. and Pahl, S. 1992. Comparison of techniques for measurements of fog liquid water content. *Tellus 44B*, 604–611.

Baumgardner, D. 1983. An analysis and comparison of five water droplet measuring instruments. *J. Climate Appl. Meteor. 22*, 891–910.

Berner, A. 1988. The collection of fog droplets by a jet impaction stage. *Sci. Total Environ. 73*, 217–228.

Berner, A. 1989. Haze and its relation to atmospheric accumulation aerosol. *Sci. Total Environ. 86*, 251–263.

Durham, M. D. and Lundgren, D. A. 1980. Evaluation of aerosol aspiration efficiency as a function of Stokes Number, velocity ratio and nozzle angle. *J. Aerosol Sci. 11*, 179–188.

Enderle, K.-H. and Jaeschke, W. 1988. An intercomparison study dealing with chemistry and physics of fogwater collection. In: *Chemistry and physics of fogwater collection, Gesellschaft für Strahlen- und Umweltforschung, München 6* (eds. W. Jaeschke and K.-H. Enderle), 7–87.

Flossmann, A. I., Hall, W. D. and Pruppacher, H. R. 1985. A theoretical study of the wet removal of atmospheric pollutants. Part I: The redistribution of aerosol particles captured through nucleation and impaction scavenging by growing cloud drops. *J. Atmos. Sci. 42*, 583–606.

Fuchs, N. A. 1975. Sampling of aerosols. *Atmos. Environ. 9*, 697–707.

Fuzzi, S., Facchini, M. C., Orsi, G., Lind, J. A., Wobrock, W., Kessel, M., Maser, R., Jaeschke, W., Enderle, K.-H., Arends, B. G., Berner, A., Solly, I., Kruisz, C., Reischl, G., Pahl, S., Kaminski, U., Winkler, P., Ogren, J. A., Noone, K. J., Hallberg, A., Fierlinger-Oberlinninger, H., Puxbaum, H., Marzorati, A., Hansson, H.-C., Wiedensohler, A., Svenningsson, I. B., Martinsson, B. G., Schell, D. and Georgii, H.-W. 1992. The Po Valley Fog Experiment 1989: an overview. *Tellus 44B*, 448–468.

Gerber, H. 1991. Direct measurements of suspended particulate volume concentration and far infrared extinction coefficient with a laser diffraction instrument. *Appl. Optics 33*, 4824–4831.

Hinds, W. C. 1982. *Aerosol Technology*, John Wiley & Sons, 424 p.

Knollenberg, R. G. 1972. Comparative liquid water content measurements of conventional instruments with an optical array spectrometer. *J. Appl. Meteor. 11*, 501–508.

Mallant, R. K. A. M. 1988. A fog chamber and wind tunnel for calibration of cloud water collectors. In: *Acid deposition at high elevation sites* (eds. M. H. Unsworth and D. Fowler), Dordrecht, Kluwer Acad. Publ., 479–490.

Marple, V. A. and Willeke, K. 1976. Impactor design. *Atmos. Environ. 10*, 891–896.

Munger, J. W., Collett Jr., J., Daube Jr., B. and Hoffmann, M. R. 1989. Chemical composition of coastal stratus clouds: dependence on droplet size and distance from the coast. *Atmos. Environ. 23*, 2305–2320.

Noone, K. J., Charlson, R. J., Covert, D. S., Ogren, J. A. and Heintzenberg, J. 1988. Cloud droplets: Solute concentration is size dependent. *J. Geophys. Res. 93*, 9477–9482.

Ogren, J. A., Heintzenberg, J., Zuber, A., Noone, K. J. and Charlson, R. J. 1989. Measurements of the size-dependence of solute concentrations in cloud droplets. *Tellus 41B*, 24–31.

Ogren, J. A., Noone, K. J., Hallberg, A., Heintzenberg, J., Schell, D., Berner, A., Solly, I., Kruisz, C., Reischl, G., Arends, B. G. and Wobrock, W. 1992. Measurements of the size dependence of the concentration of non-volatile material in fog droplets. *Tellus 44B*, 570–580.

Pandis, S. N., Seinfeld, J. H. and Pilinis, C. 1990. Chemical composition differences in fog and cloud droplets of different sizes. *Atmos. Environ. 24A*, 1957–1969.

Schell, D. and Georgii, H.-W. 1989. Design and operation of a two-stage fogwater collector. In: *Mechanisms aand effects of pollutant transfer into forests* (ed. H.-W. Georgii), Dordrecht/Holland: Kluwer Academic Publ., 221–229.

Seidl, W. 1989. Ionic concentrations and initial S(IV)-oxidation rates in droplets during the condensational stage of cloud. *Tellus 41B*, 32–50.

Vincent, J. H., Stevens, D. C., Mark, D., Marshall, M. and Smith, T. A. 1986. On the aspiration characteristics of large-diameter, thin-walled aerosol sampling probes at yaw orientations with respect to the wind. *J. Aerosol Sci. 17*, 211–224.

Winkler, P. 1986. Observation of fogwater composition in Hamburg. In: *Atmospheric pollutants in forest areas* (ed. H.-W. Georgii). Dordrecht/Holland: D. Reidel Publ. Comp., 143–151.

Wobrock, W., Schell, D., Maser, R., Kessel, M., Jaeschke, W., Fuzzi, S., Facchini, M. C., Orsi, G., Marzorati, A., Winkler, P., Arends, B. G. and Bendix, J. 1992. Meteorological characteristics of the Po Valley fog. *Tellus 44B*, 469–488.