

## A Collector for Fog Water and Interstitial Aerosol

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### ABSTRACT

An active heatable cloud water collector for ground sampling is presented. The collector can be operated unattended for approximately one week, even in harsh winter conditions. The collection strands are Teflon tubes. A preset cycle of 15-min sampling followed by 250 s of mild heating using wires inserted into the tubes is used. The lower cutoff diameter for fog droplets is 7.3  $\mu\text{m}$ , and its overall collection efficiency is 79% for the liquid water content of fogs at the experimental site in central Europe. It performed reliably during a 2-yr experiment. The collected fog water interacts exclusively with inert materials such as Teflon and Perspex so the collector is well suited for trace analyses of fog water. The collector can be upgraded with an interstitial aerosol collection unit, at the expense of unattended operation. The lower cutoff diameter of the fog water collection strands is 8.1  $\mu\text{m}$  when the interstitial aerosol module is installed. The module efficiently collects particles with diameters <3.5  $\mu\text{m}$ . For these particles, size-segregated samples in four size classes at diameters down to 0.06  $\mu\text{m}$  are collected with a Berner-type impactor. The collector was successfully employed in a mountainous region of central Europe. Over 400 samples were collected within 2 yr. With the collection unit for interstitial aerosol added, 31 samples were collected in a 2-month period.

### 1. Introduction

The collection of fog water has a long history (Grunow 1955; Baumgartner 1958). The scientific scopes of such studies include cloud microphysical processes, fog water chemistry, the role of fog in the hydrology of watershed catchments, and the contribution of fog water fluxes to biogeochemical cycles in ecosystems. As with other atmospheric parameters such as temperature, wind, composition of air masses, and precipitation amount and chemistry, the long-term observation of fog water is a prerequisite for various studies such as the climatology of fog physics and chemistry. Under optimal conditions, fog water is collected and preserved for analysis automatically.

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Several types of fog water collectors have been developed and successfully applied [see Wieprecht et al. (2005) for a recent intercomparison experiment]. However, only limited studies have recorded the chemistry of fog water over extended periods and on a continuous basis (Acker et al. 1998; Anderson et al. 1999). These routine experiments employed passive fog collectors in which wind is utilized to drive the fog droplets to the collector, where they are collected via impaction on strings. The cutoff diameter for such samplers depends on wind speed. Nevertheless, these collectors are well suited for use at windy sites such as mountaintops. At sites with limited wind speeds, however, active fog collectors are needed (Fuzzi et al. 1996). Wieprecht et al. (2005) recently compared fog water collectors employing various collection techniques such as impaction on strings, jet and slit impaction, and counterflow virtual impaction. In principle, all these fog collectors can be modified for continuous operation through a combination with an autosampler.

Problems arise when ambient temperatures are below the freezing point of water. Collected droplets freeze immediately and deicing of fog water from the collector normally needs to be done by hand. Collectors with heating options were described and applied by Demoz et al. (1996), Collett et al. (1993), Fuzzi et al.

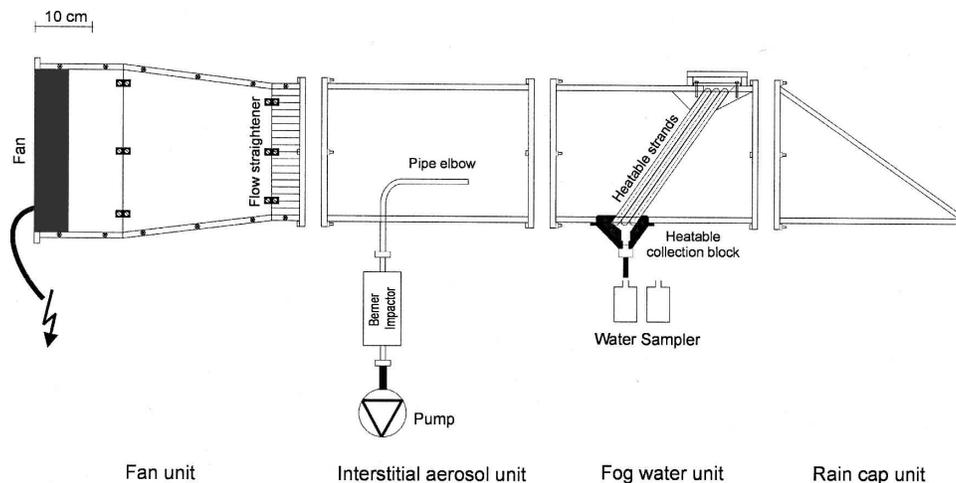


FIG. 1. Schematics of the four modules of the fog and interstitial aerosol collector. Foggy air moves from the right to the left. It enters the rain cap from below and travels through the strands where fog droplets are impacted. After exiting the fog water collection unit, it either enters the fan unit directly or travels through the interstitial aerosol unit, where subsamples are taken.

(1996), and Mertes et al. (2001). Presumably, these collectors have not been applied in routine applications under severe wintry conditions for the systematic collection of large volumes of foggy air. We present here a unique design of a fog collector that operates actively, continuously, and automatically. It has proven to be reliable in a field application over more than 13 months (Klemm and Wrzesinsky 2007), partly under heavy icing conditions.

Within a foggy air mass, a portion of the particles present is not as hygroscopic and is therefore present as relatively dry, interstitial aerosol particle material. The expected diameter of interstitial aerosol is usually smaller than  $2 \mu\text{m}$  (Seinfeld and Pandis 1998). However, there is often no sharp size cut between the interstitial aerosol and the fog droplets so that the transition range between these phases needs special consideration in a sampler design for interstitial aerosol. While the primary goal of our collector development was on routine sampling under various atmospheric conditions, we also constructed an optional module designed to sample interstitial aerosol particles from the foggy air. For this novel module, samples have to be taken manually, which limits its applicability in operational routines. It is intended for use in intensive campaigns, and its modular character makes it a valuable addition to the collector.

## 2. Fog collection modules

Our focus was the development of an active stand-alone system that is able to collect fog samples on an

event basis. Automated sample collection and reliable operation during harsh winter conditions were the most important design issues based on previously existing fog collectors. At temperatures below the freezing point of water, fog water freezes immediately after collection. Mild heating is the only operational method to retrieve fog water samples with minimum modification. Jet impactor, slit impactor (Collett et al. 1993, 1995; Schell et al. 1997; Straub and Collett 2001; Wieprecht et al. 2005), or counterflow virtual impactor (Schwarzenböck et al. 2000) designs appear inappropriate for modification to heatable versions. On impactor strings, the deposited and frozen droplets are distributed quite evenly and in a shallow layer, which is very advantageous for collector design.

The basic collector design and collection principle is similar to that of the original Caltech active strand cloud water collector (CASCC) and the heated version developed subsequently (Daube et al. 1987; Demoz et al. 1996). The basic collector (Fig. 1) consists of two main modules: the fan unit with the fan and the flow straightener and the fog water collection unit with the strands. The fan unit is square (maximum  $40 \text{ cm} \times 40 \text{ cm}$ ) and 56 cm long. The fan is a standard ebm W2E300-CP02-31, operating at 230 VAC and using a maximum current of 1.1 A. A honeycomb flow straightener is used to ensure a uniform and laminar airflow. The honeycombs are 6 mm in diameter, a total of 80 mm high, and are made of polycarbonate. For the fan unit, mainly Perspex material was used. The parts were glued together and bolted to each other with stainless steel.

TABLE 1. Specifications of the fog sampler without and with the interstitial aerosol collection unit. Subunits are defined in Fig. 1.

Parameter	Fog sampler (fan, fog water, and rain cap units)	Fog and interstitial aerosol sampler (all units)
Sampling area (cross section)	0.235 × 0.235 m <sup>2</sup>	
Screen inclination	35°	
No. of screens	6	
Diameter of strands	1.5 mm	
Strand spacing	5 mm	
Strand length (total)	81 m	
Airflow	0.446 m <sup>3</sup> s <sup>-1</sup>	0.362 m <sup>3</sup> s <sup>-1</sup>
Mean airspeed	8.1 m s <sup>-1</sup>	6.5 m s <sup>-1</sup>
Theoretical collection efficiency	88%	88%
50% cutoff diameter for fog droplets	7.3 μm	8.1 μm
Measured collection efficiency (for mean droplet spectrum at site)	79%	No data

The fog water collection unit is 25.5 cm × 25.5 cm and 40 cm long. Fog droplets traveling with the airstream are impacted on the collection strands. Once impacted, drops grow due to coalescence with other drops and eventually run down the slanted strands into the collection bottles, driven by the moving air and by gravity. Teflon tubing (1.5-mm outer diameter) was used as strand material to prevent contamination of the fog water. A 0.6-mm heating wire was inserted into the tubing. Because it was impossible to insert the wire into the tubing for more than 6 m in length, 4- and 5-m sections were manufactured. The ends were fed into the top element of the screen. Each screen consists of 45 windings of strands, adding up to 27 m of heatable tubing per screen. The top and bottom axes are made of stainless steel tubes and are inserted into Perspex tubing with grooves to hold the strands in place.

The heating wires are connected within the top element and are operated at 12 VAC. The heating wire is made of constantan (Cu55/Ni45) and has a specific resistance of 1.73 Ω m<sup>-1</sup>. The total heating power is 329 W. The screens are mounted in sliders within the collection unit at an angle of 35° to the vertical and can be removed for cleaning. Water that drips from the strands is collected in the Teflon collection block that drains it to the outlet. Heating elements are inserted into the block to avoid freezing. To optimize dispersion of heat within the block, brass rods were inserted in the holes, and the heating elements were screwed into these rods. The heating elements are standard soldering elements with a power of 50 W each. Although ten drilling holes were available, only three of them were used during the experiments.

Surface materials within the collection unit are Perspex and Teflon. For connecting the parts, Perspex glue and polyethylene screws were used. The cloud or fog water interacts only with Perspex and Teflon and is thus not contaminated by the collector with ions, metals,

carbon, and most other trace substances. To exclude raindrops from collection, a rain cap unit (made of Perspex) is fixed to the front end of the collector.

The mean airspeed within the collector was measured as an area-weighted average of 8.1 m s<sup>-1</sup> leading to an average airflow of 1604 m<sup>3</sup> h<sup>-1</sup> or 0.446 m<sup>3</sup> s<sup>-1</sup> (Table 1). The collection efficiency  $\varepsilon$  of the collector was calculated after Davidson and Friedlander (1978) and Demoz et al. (1996), taking into account the collection efficiency of one strand and the physical data of the collector (Table 1). The cut size diameter of a droplet collection efficiency of 50% is 7.3 μm. The theoretical maximum collection efficiency for  $d \rightarrow \infty$  is 88%.

During the experimental phase at the “Waldstein” site (Klemm and Wrzesinsky 2007), an FM-100 droplet spectrometer (Droplet Measurement Technologies, Boulder, Colorado) was employed to measure droplet size distribution in 40 size classes between 2 and 50 μm in diameter. For the mean fog droplet spectrum (shown as the cumulative curve in Fig. 2) the actual collection efficiency  $\varepsilon_{\text{real}}$  was calculated to be 79% using

$$\varepsilon_{\text{real}} = \frac{\sum_{\text{class}} \varepsilon(d_{\text{class}}) \text{LWC}(d_{\text{class}})}{\text{LWC}_{\text{total}}}, \quad (1)$$

where  $\varepsilon(d_{\text{class}})$  is the collection efficiency for the respective class diameter,  $\text{LWC}(d_{\text{class}})$  is the liquid water content measured for the class diameter, and  $\text{LWC}_{\text{total}}$  is the total liquid water content. Table 1 summarizes all measured and calculated parameters of the fog collector.

In comparison to earlier active strand collectors (Demoz et al. 1996), the 50% cut size diameter is rather high. This is due to the relatively large diameter of the heated collection strands. About 21% of the fog water passes the strands without being collected. Roughly half of the LWC that is not collected is represented by fog droplets with 7.3- or 8.1-μm (section 3) diameter

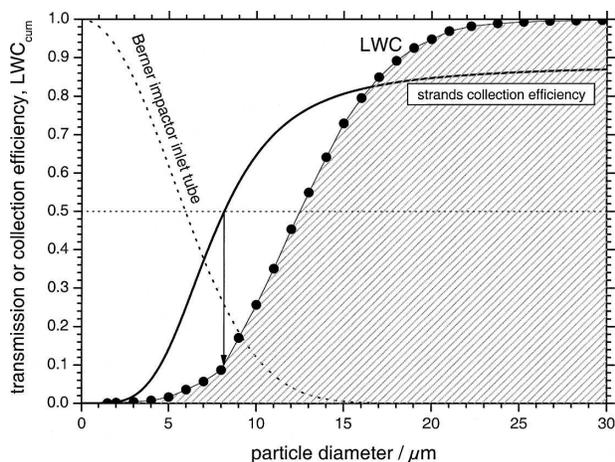


FIG. 2. Collection efficiency of the fog water strands (bold gray curve) with the version of the full collector with interstitial aerosol unit, transmission efficiency of the Bernier impactor inlet tube (dotted line), and the average cumulative LWC (dotted curve with associated shaded area) as a function of particle diameter at our collection site Waldstein. The vertical arrow indicates the 50% cutoff diameter of the fog water collection unit at  $8.1 \mu\text{m}$ .

and below. As the chemical composition of fog water droplets is a function of their sizes (Collett et al. 1993; Bator and Collett 1997; Hoag et al. 1999), the interpretation of chemical analyses must be performed cautiously.

The collector is operated with a programmable microcontroller following three guidelines. First, the visibility, as measured with a Vaisala “PWD11” present weather detector, served as main switch. At visibilities below 500 m, the fog collector was activated. Second, when the air temperature was below  $1^\circ\text{C}$ , the heating cycle of the strands was initiated. Third, when the collecting block temperature dropped below  $1^\circ\text{C}$ , the block was heated. For the collection of consecutive fog water samples on an event basis or during single events, a Teledyne ISCO 6712 automatic programmable water sampler was used.

At the given airflow through the fog collector, the heating power of the strands (329 W) could be used to heat the air from  $1.00^\circ$  to  $1.65^\circ\text{C}$ , if the heating was 100% efficient. The saturation vapor pressures for  $1^\circ$  and  $1.65^\circ\text{C}$  are 656.8 and 688.3 Pa, respectively, corresponding to the specific humidity of  $5.209 \times 10^{-3}$  and  $5.459 \times 10^{-3} \text{ kg m}^{-3}$ . This indicates that air can hold  $250 \text{ mg m}^{-3}$  more water vapor at  $1.65^\circ$  than at  $1^\circ\text{C}$ , which corresponds to the typical LWC of fog. Although total evaporation of  $250 \text{ mg m}^{-3}$  LWC cannot occur under these conditions because almost half of the available energy would have to be used for the vaporization process, our example calculation impressively shows that evaporation losses of fog water during collection

can be very large when the strands are heated during fog collection. Therefore, the collector was alternately operated with the fan on and the heating off, avoiding evaporation during collection, followed by mild heating and deicing of the strands with the fan inactive. We found that a cycle with 15-min collection and 250-s deicing was appropriate for our site.

From our experience we cannot derive a minimum temperature at which the collector still operates appropriately. Most fog events occurred at temperatures above  $-5^\circ\text{C}$ . It was less the temperature itself that challenged the collector and more the LWC during any temperature below the freezing point of water. For the conditions at about 850 m MSL in central Europe, the heat sample cycle as given above worked perfectly.

### 3. Interstitial aerosol sampling module

Between the fan module and the fog water collection module, an additional module can be implemented for sampling the interstitial aerosol (Fig. 1). As indicated in section 1, caution must be applied because a fraction of the larger fog droplets passes the fog water unit (Fig. 2). A perfect collection of particles above a given cutoff diameter on the order of  $1 \mu\text{m}$  is extremely difficult, especially for droplets that tend to break up at the required high flow velocities of several tens of meters per second (Schwarzenböck and Heintzenberg 2000). Therefore, we introduced a five-stage impactor of the Berner type (Berner 1984) to collect interstitial particles that pass the fog collection unit (Fig. 1). The lower cutoff diameters for the stages of this impactor are 0.06, 0.15, 0.37, 1.04, and  $3.5 \mu\text{m}$  for stages 1, 2, 3, 4, and 5, respectively (A. Berner 2006, personal communication). Because of possible contamination from larger droplets, stage 5 (lower cutoff  $3.5\text{-}\mu\text{m}$  diameter) is not used for sample collection and data analysis. As a result, particles with diameters between  $3.5$  and  $8.1 \mu\text{m}$  are excluded from collection with our system. This disadvantage of the exclusion of about 10% of the LWC from being collected is outweighed by minimum cross-contamination of the bulk fog water and the interstitial aerosol.

An induction elbow pipe is used as the inlet for the impactor. Particle segregation effects need to be considered at the subsampler inlet. Inertial and gravitational forces for larger particles and higher diffusion coefficients for smaller particles may selectively affect the transmission efficiency for particles of a given size. The most effective deposition to the sample line walls is caused by inertial deposition in the elbows bends.

Sample aspiration is established in an isokinetic and isoaxial fashion. Isoaxial conditions are guaranteed

through orientation of the subsample inlets directly into the flow field, downwind of the fog water collection strands. To establish isokinetic conditions at the pipe elbow inlet, a velocity profile was measured with high resolution at the cross sections of the sample line inlet. The inlet pipe elbow was positioned in a region with flow velocity similar to the intake flow velocity.

The pipe elbow itself is made of seamless stainless steel pipe. To minimize interference effects on the flow field, the elbow is designed according to specifications of Pitot tubes. The elliptical head shape of the inlet is designed to reduce aberration caused by angular deflection from the flow direction.

The flow regime of the sample line exhibits a Reynolds number of  $Re = 5300$ . The transmission efficiency was calculated taking turbulent inertial bend deposition (Pui et al. 1987), turbulent gravitational settling (Schwendiman et al. 1975), and turbulent diffusion (Friedlander 1977) into account and is also shown in Fig. 2. The 50% cut size diameter is  $5.9 \mu\text{m}$ . Interstitial aerosol particles in the diameter range of  $0.05\text{--}3.5 \mu\text{m}$  (Berner stages 1–4) efficiently pass the strands of the fog water unit ( $>89.5\%$  at  $3.5 \mu\text{m}$ ). Contamination from droplets on the impactor stage 4 is small because  $99.5\%$  of the LWC is present in droplets  $>3.5 \mu\text{m}$ . Therefore, interstitial aerosol particle material can be collected efficiently with this collection unit.

The implementation of the interstitial aerosol collection unit into the fog collector reduced the airflow to  $0.362 \text{ m}^3 \text{ s}^{-1}$ , and the mean airspeed to  $6.5 \text{ m s}^{-1}$ . This leads to a cutoff diameter of the strands of  $8.1 \mu\text{m}$  as opposed to  $7.3 \mu\text{m}$  with the fog collector alone (Table 1; Fig. 2).

#### 4. Field application and outlook

The fog collector was operated between February 2000 and February 2002 at the research site Waldstein in the Fichtelgebirge Mountains, northeast Bavaria (Matzner 2004; Klemm and Wrzesinsky 2007), at  $50^\circ 08' 32''$  latitude,  $11^\circ 52' 04''$  longitude, 775 MSL. During this long-term setup without the interstitial aerosol unit, a total of 449 samples, including 20 field blanks, were taken. With the collection unit for interstitial aerosol added, the collector was operated from October to December 2001. A total of 31 samples were collected. The fog collector works reliably and unattended even under cold winter conditions with heavy icing.

The orientation of the collector into the wind to guarantee isoaxial sampling conditions and minimizing wind shearing at the inlet with the risk of size-selective droplet collection is important.

The lower cutoff diameter of the fog sampler ( $7.3 \mu\text{m}$

without the interstitial aerosol unit and  $8.1 \mu\text{m}$  with it) seems not to be optimal for all sampling conditions. For the summer period, our modified version of the Caltech sampler CASCC (Daube et al. 1987) with  $0.51\text{-mm}$ -diameter collection strands exhibits a smaller cutoff diameter of  $4.3 \mu\text{m}$  and therefore better collection of LWC in the  $4.3\text{--}8.1\text{-}\mu\text{m}$  size range. Also counterflow virtual impactors and jet or slit impactors, as mentioned above, may have advantages such as sharper cutoff characteristics. However, during occasions of potential occurrence of freezing conditions, which are likely to occur eight months out of the year at our sampling site, the sampler presented here is superior for unattended operation. The scientific merit of uninterrupted fog water chemistry datasets, throughout all seasons, is evident.

For the separation between fog water and interstitial aerosol particle mass, the relatively large cutoff diameter of the active strands is not disadvantageous. To separate well between the droplets and the interstitial aerosol, a relatively large size range must be excluded from analysis. In our application, particles with diameters between  $3.5$  and  $8.1 \mu\text{m}$  (50% cutoff diameters) were excluded. For an active strand collector with a lower cutoff (e.g.,  $4 \mu\text{m}$ ), another size range would have to be excluded. In combination with the Berner impactor, that would have applied to the size range  $1.04\text{--}4 \mu\text{m}$ . The potential of such strategies in various environments and types of aerosol compositions will have to be studied in further experiments.

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