This project focused on the continued design, fabrication, and testing of a new aircraft-mounted cloud collector, suitable for collecting samples with micromolar concentrations of relevant chemical species. It also supported participation in the DYCOMS-II field project as a test of the collector's performance. Additional support for this work was provided by the NCAR Advanced Studies Program through a graduate fellowship to PhD student Derek Straub. We focus attention here on the design and function of the new cloud sampler and briefly summarize DYCOMS-II cloud composition results.

1.1 Cloud collector design

An axial-flow cyclone cloud collector (Figure 1) capable of taking multiple samples per flight was designed to fit in a Particle Measurement Systems (PMS) wing pod canister on the NSF/NCAR C-130 aircraft. PMS optical probes are used on a variety of research aircraft; therefore, many have standard mounts and electrical interfaces to accommodate the PMS canister. Since the axial-flow cyclone cloud collector fits into one of these canisters with modifications only to the canister end caps, it could, in theory, be mounted on a variety of research aircraft without substantial modification. Changes in collector performance, however, are expected if flight speeds differ greatly between a newly selected aircraft and the C-130.

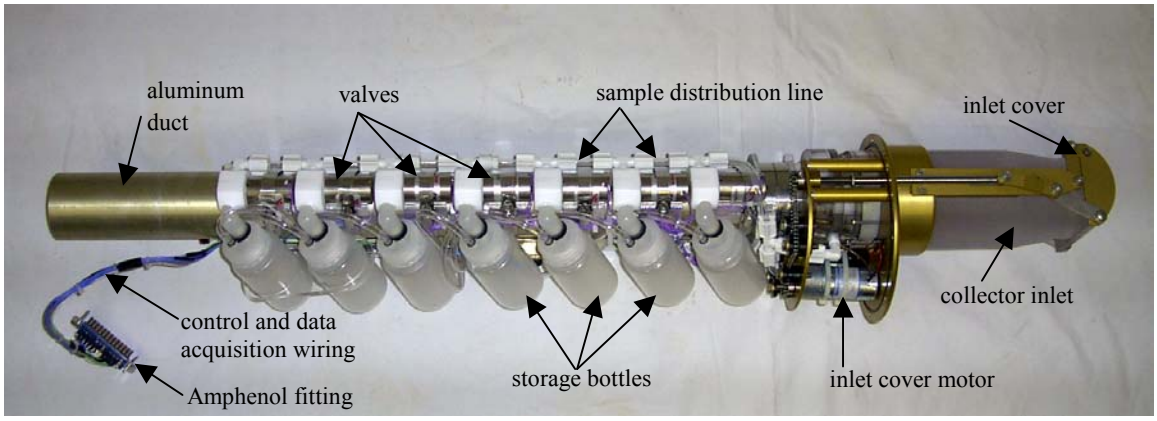

Figure 1. Picture of the final axial-flow cloud water collector shown outside of the modified PMS canister.

Like most cloud collectors, the axial-flow cyclone collector takes advantage of cloud drops' inertia for their collection. As air enters the inlet, stationary vanes in the middle of the air stream introduce a rotational flow around the centerline of the collector duct. Droplets with sufficient inertia are thrown to the interior walls of the instrument by centrifugal force, where they deposit and are later collected. The axial-flow design was selected because it was expected to be less sensitive to changes in aircraft orientation and speed, which can be problematic in more traditional inertial impaction systems (Huebert and Baumgardner, 1985). It was also expected to minimize droplet shatter and re-entrainment.

Throughout the design phase of this project, a computational fluid dynamics (CFD) software package, FLUENT v5.0, was used as a design aid. Model simulations were used to examine temperature and pressure fields within the collector under sampling conditions, to determine the effectiveness of the stationary vanes in producing a rotational flow field, and to determine the efficiency of cloud drop collection as a function of drop size. Changes to the collector design were made in response to model output and then simulations were rerun. In all, 12 iterations of collector geometry were examined before a final design was accepted.

The interior collection surfaces of the final axial-flow collector design are shown schematically in Figure 2. During sampling, the inlet cover (not shown in the figure) is moved out of place using a small motor. Ram pressure is used to drive droplet laden air into the collector duct, negating the need for a high volume pump. The air stream encounters the stationary vanes, which introduce a rotational flow around the duct centerline. Small drops follow the air streamlines and are transported through the collector. Somewhat larger drops are forced to turn by the flow, but cannot follow the streamlines exactly and are collected by impaction on the interior walls of the collector. Figure 3 contrasts the passage of 1 µm diameter drops through the collector while 10 µm drops are collected on the interior walls of the collection cylinder. The 30 µm drops impact on the curved vanes, eventually being shed and collected on the cylinder walls. Shear flow from the air passing through the collector drives the collected water downstream along the walls until it encounters an extraction slot. Only drops impacted upstream of the slot are transported into the slot and are then available for collection. Two low-pressure ports draw sample out of the slot and into a sample storage system. A series of solenoid valves then directs the liquid into one of seven storage containers.

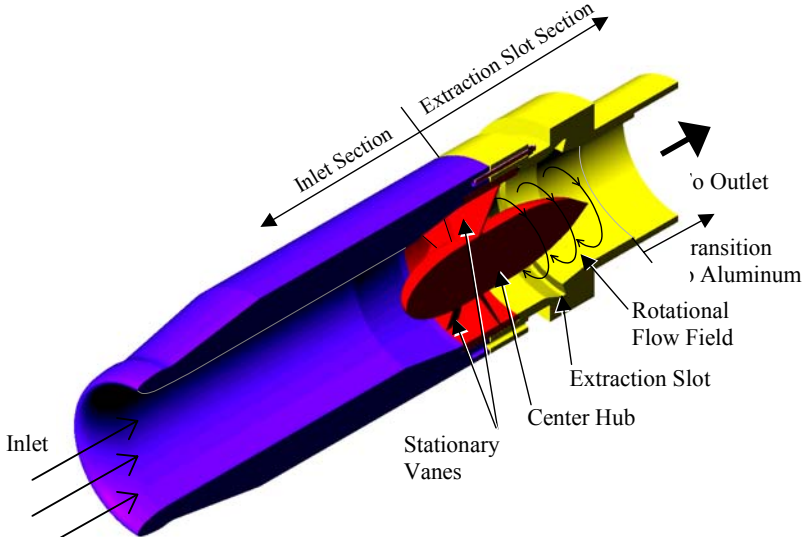

Figure 2. Interior schematic of the cloud collector.

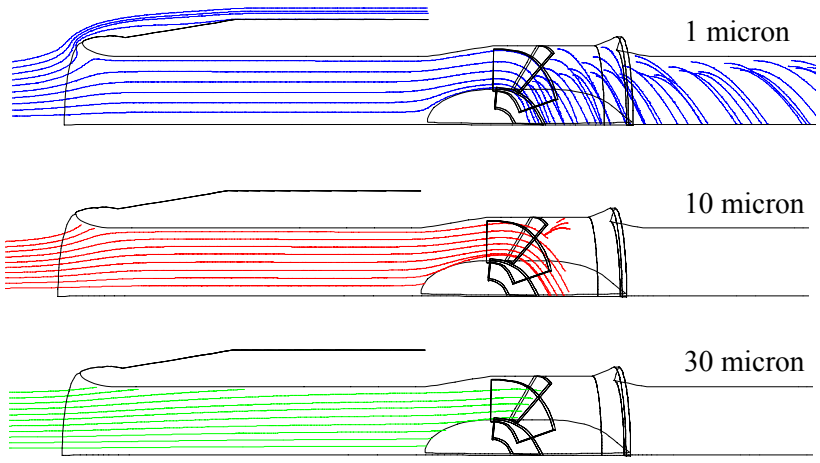

Figure 3. CFD trajectories for a) 1 µm, b) 10 µm, and c) 30 µm diameter drops.

Collection efficiencies for the axial-flow cyclone collector were predicted for a variety of drop sizes by the FLUENT model. The 50% size cut diameter, the diameter at which half the drops are collected, was predicted to be 8 µm. This size was chosen to collect most cloud drops but exclude most smaller, unactivated aerosol particles.

Because the collector is located in a pod on the wing of the aircraft, it is inaccessible during flight and collector functions must therefore be controlled and monitored remotely. A data logging and control program was written using National Instruments' Labview v5.1 software. The program was integrated with the C-130 data stream, so that aircraft and other online parameters could be automatically plotted and/or saved along with the axial flow collector data from the aircraft cabin. Virtual switches on the program main panel are used by an on-board operator to open and close the inlet cover and to direct the collected sample to one of the seven collection containers.

1.2 DYCOMS-II Field Testing

The Dynamics and Chemistry of Marine Stratocumulus, Phase II (DYCOMS-II) field project took place in July 2001. Flights were based out of Coronado Island, near San Diego, CA, and flew southwest over the Pacific Ocean. The main focus of the project was the study of the entrainment of free tropospheric air into the marine boundary layer and the production of drizzle in marine stratocumulus cloud layers (Stevens et al., 2002). Although chemistry was not the primary focus of the campaign, the significant in-cloud flight time provided a good opportunity to evaluate the field performance of the axial-flow cyclone cloud collector. A total of 9 flights of approximately 10 hours each were flown.

In order to determine the appropriate collection time for each of the seven possible samples per flight, the water collection rate was predicted using output from the onboard Particle Volume Monitor (PVM). The Labview program combined the PVM output with the previously predicted collection efficiencies for droplets sized 6 to 26 μm and output the predicted collection rate in real-time on the program screen. Predicted collection rates varied between 2 and 13 g min^{-1}.

Throughout the project the collector systems, both hardware and software, functioned as expected and samples for chemical analyses were successfully collected. It became obvious however, that the amount of cloud water collected in the sample bottles was much lower than predicted. Post-flight examination of the collected sample volume showed it to be well below expected amounts. To investigate the possibility that the sample collection rate prediction scheme was misrepresenting real conditions, drop size distributions measured in-flight with an SPP-100 were used to more rigorously estimate the collection rate. Results from this exercise indicated that the mean collection efficiency of the axial-flow cyclone collector was approximately 9% of the expected values.

1.3 Laboratory Investigation

In order to investigate the apparent collection losses in the axial-flow cloud collector, a quantitative calibration was performed in the laboratory at CSU. Fluorescein-tagged monodisperse drops ranging in size from 4 to 20 μm diameter were created with a Vibrating Orifice Aerosol Generator (VOAG) and introduced into the collector. Various collection surfaces were then extracted and the internal collection efficiency of the cloud sampler determined as a function of drop size.

To create aircraft equivalent air speeds through the collector, three large blowers were plumbed in parallel behind the collector. Individual 4 to 5 hour calibration runs, along with several replicate runs, were performed for drops sized 4, 5, 6, 8, 10, and 20 μm diameter. After each run, the surfaces onto which the drops had deposited were soaked in a known volume of extract solution and the fluorescein concentration was measured with a spectrofluorophotometer. The mass of fluorescein deposition on each surface was then calculated, after correcting for deposition of doublet and triplet drops using the procedure of Straub and Collett (2002).

The total fraction of introduced fluorescein recovered from the internal surfaces of the axial-flow cyclone collector averaged 98.3%, indicating an efficient recovery of drops of all tested sizes within the collector. Internal collection efficiency curves for drops collected upstream of the extraction slot compared well with model-predicted collection efficiencies corresponding to lab conditions (Figure 4). This good agreement lends confidence in the ability of the modeling approach to provide a realistic picture of collector performance. The modeled and experimental deposition of droplets within the inlet and on the vanes (Figure 5) also showed good agreement, suggesting that the apparent under-sampling of cloud water during DYCOMS-II was caused by factors other than drops failing to reach the duct wall upstream of the extraction slot.

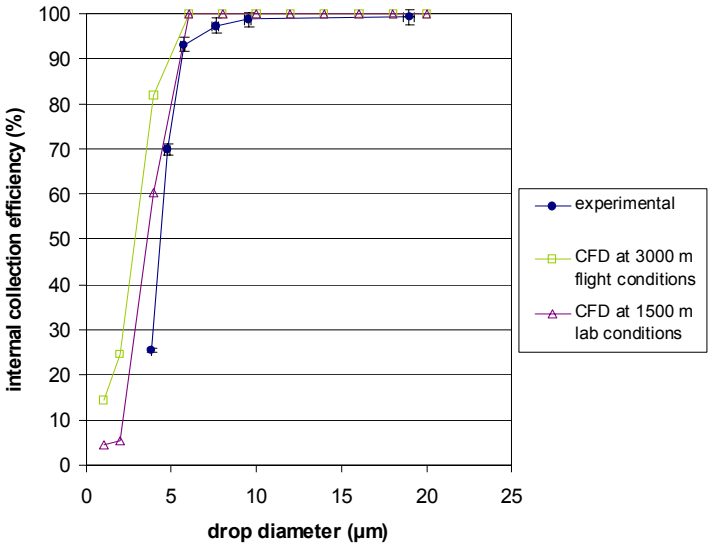

Figure 4. Experimental and modeled internal collection efficiency curves for the axial-flow cyclone cloud collector.

Visual observations of the water flow pattern inside the collector during lab testing indicated that the extraction slot was not efficiently removing the collected water. Drops injected into the inlet deposited on the collector walls upstream of the extraction slot and, migrated towards the slot. The water, however, was also observed flowing on the wall downstream of the slot, indicating the extraction slot design itself was inadequate for efficiently transferring collected water from the sampler walls to the sample storage system. Very little water was actually observed in the extraction slot and what was there tended to pool and circulate along the upstream edge without descending to the bottom of the slot where it could be removed by the sample ports. This occurred even with a vacuum applied to the sample ports to simulate in-flight conditions.

The results of the laboratory calibration suggest that while the CFD software was generally reliable and useful in predicting deposition patterns and flow fields within the collector, a new approach is required to accurately simulate or experimentally evaluate the interaction of the collected water and the complex geometry of the extraction slot. While the majority of the available droplets are impacted on the collector wall as expected, they are not removed by the extraction slot, apparently because they flow over the slot, rather than into it. This explains the low sample volume yield observed during DYCOMS-II. This conclusion suggests that the collector should provide a representative sample of collected cloud water for measurement of chemical composition; however, longer than expected sampling times are needed to ensure collection of adequate sample.

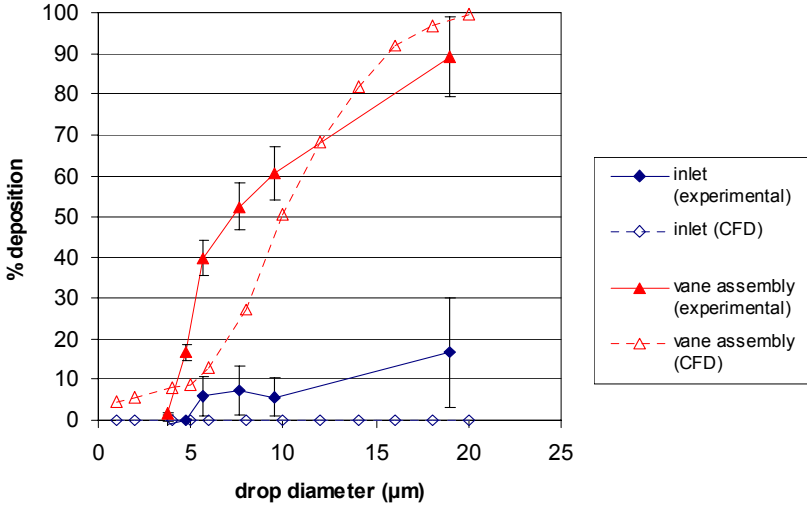

Figure 5. Experimental and modeled drop deposition in the axial-flow collector inlet and on the vane assembly.

1.4 DYCOMS-II Cloud Composition

In total, 50 individual cloud water samples suitable for chemical analyses were collected in marine stratocumulus clouds over the course of the DYCOMS-II campaign. Measured pH values ranged from 3.23 to 4.82 with a mean of 4.02. The ionic concentrations were dominated by sea salt as might be expected (Figure 6), however significant concentrations of non-sea salt Ca^{2+}, Mg^{2+}, and SO_4^{2-} as well as NO_3^- and NH_4^+ were measured in all sampled clouds. Fe and Mn were also present in measurable quantities in most cloud water samples. While several research flights had 5-day back trajectories that

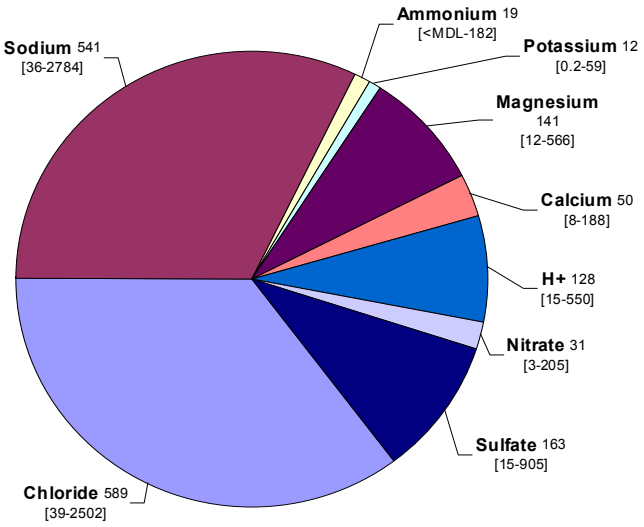

Figure 6. Mean ionic composition of DYCOMS-II cloud water in μN. Mean values follow the species name and the range is shown in brackets.

It was possible to estimate the aqueous oxidation of S(IV) in the sampled clouds using ammonium as a tracer. Using published rate laws, and measured species' concentrations, it was determined that oxidation by H_2O_2 should dominate aqueous sulfate production. Abundant hydrogen peroxide measured in this summer environment implies that the S(IV) oxidation rate is limited by the availability of SO_2.

While there are several reports of cloud water pH from airborne measurements, previous data on the chemical composition of remote marine clouds are (surprisingly) absent from the literature. Other reports of remote marine cloud chemistry are almost exclusively limited to coastal sites with ground-based sampling. In this regard, the DYCOMS-II data represent a unique and much needed addition to our first hand knowledge of remote marine cloud chemistry.

passed over North America, most did not. Transport of continental pollution, emission from ships, marine biological activity, and possibly Asian dust are all potential sources of non-sea salt material to the remote marine study environment.

Within samples from the same flight, solute concentrations tended to vary with the size of cloud drops, suggesting that the drops were diluted by condensational growth. Drop number was strongly correlated to the precursor aerosol number concentrations. Between flights, concentrations varied widely, but were also related to drop size. Flights with smaller drops, tended to have higher solute concentrations and flights with larger drops had lower solute concentrations (Figure 7) .

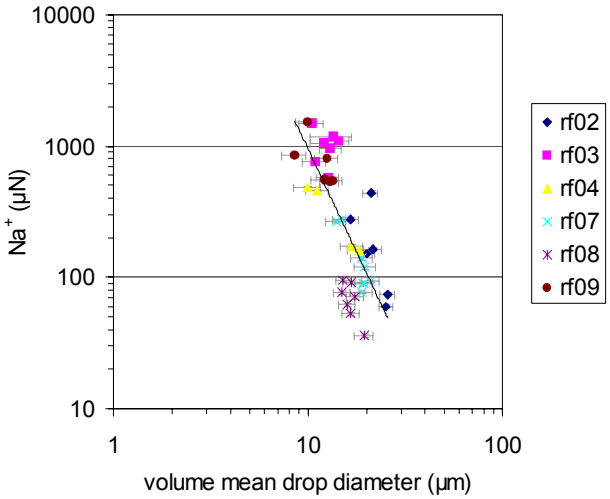

Figure 7. Na^+ is used as an example to show the cloud water concentration trend between flights.