Gas Dynamics, Characterization, and Calibration of Fast Flow Flight Cascade Impactor Quartz Crystal Microbalances (QCM) for Aerosol Measurements. J. R. Grant, A. N. Thorpe, C. James, A. Michael, M. Ware, F. Senftle, and S. Smith Center for the Study of Terrestrial and Extraterrestrial Atmospheres Howard University, Washington, D.C. 20059

During recent high altitude flights, we have tested the aerosol section of the fast flow flight cascade impactor quartz crystal microbalance (QCM) on loan to Howard University from NASA. The aerosol mass collected during these flights was disappointingly small. Increasing the flow through the QCM did not correct the problem. It was clear that the instrument was not being operated under proper conditions for aerosol collect ion primarily because the gas dynamics is not well understood. A laboratory study was therefore undertaken using two different fast flow QCM's in an attempt to establish the gas flow characteristics of the aerosol sections and its effect on particle collection, Some tests were made at low temperatures but most of the work reported here was carried out at room temperature.

The QCM is a cascade type impactor originally designed by May (1945) and later modified by Anderson (1966) and Mercer et al (1970) for chemical gas analysis. The QCM has been used extensively for collecting and sizing stratospheric aerosol particles (Chuan and Woods, 1984; Woods and Chuan, 1983; and Chuan et al, 1981). In this paper all flow rates are given or corrected and referred to in terms of air at STP. All of the flow meters were kept at STP. Although there have been several calibration and evaluation studies of moderate flow cascade impactors of less than or equal to 1L/rein. (Marple, Liu and Whitby, 1974), there is little experimental information on the gas flow characteristics for fast flow rates greater than 1 L/rein.

The Quartz Crystal Microbalance

To ensure the results were not merely artifacts of a particular instrument, these studies were carried out with the NASA 6-stage aerosol section QCM and also a 10-stage aerosol QCM designed by California Measurements Corp (CMC).

The NASA **QCM:** Each stage of this instrument is comprised of an inlet nozzle and a cylindrical housing for two quartz crystal oscillators and their associated electronics. The inlet nozzle is actually mounted in the bottom of the previous stage. When a stage is removed from the stack both its nozzle in the previous stage and housing are removed. The inlet gas and particles impinge on the upper quartz crystal, and any particulate matter in the gas may or may not adhere to the quartz surface and change its vibration frequency. The lower **crystal** oscillator is used for reference to produce a change in beat frequency and to compensate for frequency shifts due to temperature changes. As the upper crystal picks up particles, the beat **frequency** between the two crystals changes with the increase in mass.

The California Instruments **QCM:** This instrument is generally similar to the NASA QCM but has 10 stages and was calibrated by Hering (1987) for stratospheric sampling using flow rates of 1.2 L/rein. Each stage is 3 inches in diameter compared to a diameter of 1 3/4 inches for the NASA QCM. Unlike the NASA instrument, the inlet jet for each stage is located in the stage housing.

Experimental Method

The first set of experiments were performed with the NASA QCM. in order to simulate flight conditions the QCM was set up as shown in Figure 1A. Two GAST vacuum pumps were mounted in a large partially evacuated chamber which was used to simulate the low pressure condition in the stratosphere. Varian electronic pressure gauges (Model # WV100-2, designated G-1, G-2, and G-3) and a MKS flowmeter (Type 0558A-050L-SV)were arranged as shown in the sketch. To simulate operation in the stratosphere, the inlet pressure and the chamber pressure were kept the same. The pumps evacuated the QCM directly, and it was observed that the exit pressure of the QCM was always about 28% of the inlet pressure. At any particular pressure the flow rate did not change when the pressure difference across the QCM was further increased. Therefore, the system was always in the choke flow mode of operation. It is clear that under flight conditions the instrument did not operate properly, and why it failed to collect additional aerosol particles with increase of flow rate.

To study the gas dynamics, a much simpler arrangement was used (Figure 1 B). In order to control the exit pressure the chamber was eliminated. The inlet and exit pressure could be controlled by the two valves, the pressure difference across the QCM could be varied, and the gas flow could be controlled from about zero to choke flow.



Figure 1- A- Sketch of experimental configuration used to simulate flight operation of the QCM. B-Laboratory experimental configuration to study the gas dynamics of the QCM.



Figure 2- The gas flow rate as a function of the inlet pressure for the NASA 6-stage QCM under simulated flight conditions.

Figure 2 shows how the gas flow through the 6-stage NASA QCM varies with inlet pressure using the experimental configurate ion shown in Figure 1A. As this configuration simulates the flight conditions, it is obvious that during flight the flow rate was linearly related to the pressure difference across the QCM. As the QCM is operating in the choke flow mode, such a linear relationship is expected. The choke flow is due to the nozzle with the smallest jets i.e. with the smallest total area. Figure 3 shows how the flow rate changes with the total area of the individual nozzles in the QCM's. As expected the saturation or choke flow is linearly related to the total area of the nozzle irrespective of the number of the jets per nozzle of the QCM.



Figure 3- Saturation flow rate as a function of the total area of the individual nozzles.



Figure 4- Gas flow rate as a function of the change in 'pressure across the NASA-QCM for an inlet pressure of 71 mm of Hg.

To obviate choke flow, a series of tests were made using the configuration in Figure 1 B, the configuration which should be used in further flights for aerosol collection. At fixed inlet pressures of 71, 100, 300, 500, 650, and 700 mm of Hg the flow rate was measured as a function of the pressure difference across the 6-stage QCM. Figures 4 and 5 are typical of the flow as a function of the pressure difference across the QCM for inlet pressures of 71 and 100 mm of Hg, respectively. The data shows saturation or choke flow for large pressure differences. A similar study was made on the CMC-QCM (Fig. 6). Choke flow occurs at a lower flow as would be expected because the area of each nozzle is much less than the nozzle areas for the NASA QCM.



Figure 5- Gas flow rate as a function of change in pressure across the NASA-OCM for an inlet pressure of 100 mm of Hg.



Figure 6- Gas flow rate as a function of change in pressure across the CMC-QCM for an inlet pressure of 71 mm of Hg.

The critical dimensions and other characteristics of each stage in the NASA QCM are given in Table 1. The jet-to-quartz crystal distance, S, is approximately one half of the jet diameter, W, for each stage, whereas the ratio of the throat-length, T, to the jet diameter is different for each stage. Similar data for the CMC10-stage QCM are given in Table 2. The critical parameters for the QCM round jet impactors shown in the Tables 1, and 2, are calculated using the following relations.

(1) Effective cut-off aerodynamic diameter, ECAD (Mercer and Stafford 1969) for the aerosol particles, is

ECAD (
$$\mu$$
m) = 1.257 x 10³ [W³/F]^{1/2},

where W is the jet diameter in cm and F is the volumetric flow rate in cm^3/min . per jet. (2) The flow Reynolds number, N_{RE} , for a circul

$$N_{RE} = \rho W v/$$

η where p is the physical density of the air, η is the viscosity of air, and v is the air flow velocity expressed in terms of the volumetric flow rate and j et diameter:

$$v = 4F/\pi nW$$
,

where n is the number of jets per nozzle.

(3) The collection efficiency of the impaction stages were determined with respect to the dimensionless Stokes number (See Fairchild and Wheat, 1984), defined as:

$$S_{tk} = \rho v (ECAD)^2 / 9\eta W$$

where p = 1 for unit density particles. The calculations were computed using the constants $p = 1.293 \times 10^{-3}$ g/cm³ physical density of air (particles) $\eta = 1.82 \text{ x} 10^{-4}$ viscosity of air at 20 "C

Table 1

Characteristic parameters of the NASA 6-stage QCM at a volume flow rate of 7.5 L/rein (at standard temperature and pressure)

Stage	No. of	Jet Diameter	Throat	Jet to Crystal					
Number	Jets	W	Length T	Separation S	S/W	T/W	ECAD	N_{Re}	Stokes
		(mm)	(mm)	(mm)			(pm)		#
1	1	6.6	7.11	3.31	0.502	1.077	7.783	1715	0.2038
2	I	4.2	6.83	2.17	0.517	1.626	3.95	2692	0.2034
3	1	2.88	9.06	1.39	0.483	3.146	2.25	3813	0.2045
4	2	1.72	3.36	0.97	0.564	1.953	1.464	3286	0.2023
5	4	0.79	2.33	0.42	0.532	2.949	0.644	3577	0.203 I
6	4	0.60	1.89	0.30	0.500	3.15	0.426	4711	0.2029

Stage	No. of	Jet Diameter	Throat	Jet to Crystal					
Number	Jets	w	Length T	Separation S	S/ W	T/W	ECAD	N _{Re}	Stokes
		(mm)	(mm)	(mm)			(µm)		#
1	1	6	11.20	3.35	0.56	1.87	15.086	377	0.2035
2	1	4	12.30	2.25	0.56	3.08	8.2118	565	0.2034
3	1	3	13.00	1.50	0.50	4.33	5.334	746	0.2012
4	1	2	13.00	1.55	0.77	6.50	2.903	1130	0.2033
5	1	1.392	12.95	1.55	1.11	9.30	1.682	1624	0.2024
6	1	1.005	14.00	0.50	2.01	13.93	1.034	2250	0.2033
7	1	0.689	14.10	0.45	0.65	20.49	0.586	3280	0.2028
8	2	0.494	14.25	0.30	0.61	28.87	0.5049	2287	0.2031
9	4	0.331	14.20	0.20	0.60	42.87	0.391	1706	0.2031
10	7	0.249	14.40	0.15	0.60	57.88	0.335	1303	0.2029

Table 2 Characteristic parameters of the California Measurements Corp. 10-stage Q : M at a v lume flow rate of 1.5 L/rein (at standard temperature and pressure)

Study of Individual Stages

As the number of jets and jet diameters vary between different QCM stages, it is desirable to know how the flow varies with pressure difference for individual stages. This was accomplished by removing each stage consecutively starting with stage # 6 and its inlet nozzle; then stage #5, and so on until all stages were removed. The pressure drop across individual stages is calculated as follows. For example with the stage #6 and its inlet jet removed from the stack, the exit pressure of stage #5 is determined. This is the same as the inlet pressure to stage #6, if stage #6 were in the stack. The exit pressure for the entire stack has been previously determined, and therefore the pressure drop across stage #6 when it is in **the** complete stack is the difference **between** the exit pressure for the 5-stage stack and the exit pressure for the 6-stage stack. The pressure difference across each stage in the stack was determined by using the same method. The sum of the pressure drop across the individual stages was **equal** to the pressure drop across the complete stack. As the flow rate is established by the pressure difference across the complete stack for a given inlet pressure, it is to be noted that the inlet pressure to a stage is a function of the flow rate. This is true because there is a pressure drop proportional to the flow rate across the preceding stages. Fig 7a shows the inlet pressure to stage six (outlet pressure for stage 5) as a function of the flow rate for a stack inlet pressure 71 mm Hg with stage #6 removed. The pressure difference versus flow rate for stage #6 at 71 mm Hg is shown in Fig. 7b.



 $\frac{1}{10} = \frac{1}{10} = \frac{1}{10}$

Fig. 7a Flow rate versus Inlet pressure to stage #6 at 71 mm Hg

Fig. 7b Flow rate versus pressure difference for stage #6 at 71 mm Hg.

Smoke Experiments

Using both QCM's several smoke sampling experiments were conducted using the complete stacks. Smoke was introduced simultaneously to the CMC and the NASA six-stage QCM's at atmospheric pressure. No filter was used and the air flow was set at approximately 75 % of choke flow rate; that is 7.5 L/rein for the NASA stack and 1.5 L/rein for the CMC stack. These are the same flow rates used in the tables. Typical beat frequency response curves are shown for stage 5 (NASA-QCM) and stage 7 (CMC-QCM) in Fig. 8.





Figure 8a Beat frequency response of the crystals on stage 5 versus collection time for the NASA QCM.

Figure 8b - Beat frequency response of the crystals for stage 7 versus collection time for the CMC-QCM.

Note the sharp increase in beat frequency at the specific time at which the smoke was introduced. The sharp increase in beat frequency is due to the accumulation of smoke particles on the quartz crystals. The **total** mass of particles collected is reflected in the frequency difference before and after the introduction of the smoke. The frequency difference is shown as a **function** of the stage number in the respective stack in Figure 9. Stage #5 and stage #7 in the NASA and CMC stacks respectively accumulated the largest mass of aerosol particles. In Tables 1 and 2, these stages correspond to an effective cut-off aerodynamic diameter of 0.644 and 0.586 µm respectively. Thus, within experimental error those stages with the same ECAD in their respective QCM's are collecting approximately the same size particles. To a first approximation the relative sensitivity is related to the flow: i.e. the frequency change for the NASA QCM is approximately 5 times that of the CMC-QCM



Figure 9- Cumulative mass collection measured as a frequency difference for the complete stack versus the number of stages for the NASA(a) and CMC(b)QCM's.

In both figures 8a and 8b after the increase due to the smoke introduction, there is almost always a steady decrease in frequency for both QCM's. Further study of this phenomena showed that if the flow rate was kept constant, using

clean air after smoke introduction, the frequency would decrease almost to the original value before the introduction of smoke. This decrease occurs because the particles are being blown off the crystal. In the previous flight experiments the crystals were not grease coated and the pumps were left on after sampling with filtered air being pumped through the QCM until the aircraft landed and therefore much of the collected aerosol mass probably blew off the crystals. in recent laboratory experiments the crystals were coated with grease, but the grease also blew off in both QCM's at high mass flows, However, it appears that very little grease blows off at a QCM inlet pressure of 100 mm of Hg or less at these high flow rates.

Discussion

The QCM is an instrument that is used extensively in aerosol research. However, to use it at high mass flow rates one has to alter standard procedures. Recently Chuan (1993) has successfully used a four stage QCM for stratospheric aerosol measurements at a flow rate of 1.3 L/min. Our experiments indicate that a higher flow rate is possible if one designs the nozzles carefully (i.e. using the proper number of jets to maintain a low mass flow.) and keeps the flow within the correct limits at low pressures, or use an appropriate amount of grease coating so that part of the accumulated mass does not blow off the quartz crystals. Experiments are continuing on the low pressure flow rates and amount of grease coating to be used on the crystals for different experiments.

Acknowledgements

We gratefully acknowledge the financial support of Howard University and the Center for the Study of Terrestrial and Extraterrestrial Atmospheres **funded** by NASA grant NAGW-2950. We would like to thank R. Chuan, D. Woods, and W, Chiang for their support and many helpful discussions and I. Heard for helping with the data analysis.

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