United States
Environmental Protection
Agency

Environmental Sciences Research
Laboratory
Research Triangle Park NC 27711

EPA-600/2-80-053
March 1980

Research and Development

Design and Performance of an Aerosol Mass Distribution Monitor

RESEARCH REPORTING SERIES

Research reports of the Office of Research and Development, U.S. Environmental Protection Agency, have been grouped into nine series. These nine broad categories were established to facilitate further development and application of environmental technology. Elimination of traditional grouping was consciously planned to foster technology transfer and a maximum interface in related fields. The nine series are:

1. Environmental Health Effects Research
2. Environmental Protection Technology
3. Ecological Research
4. Environmental Monitoring
5. Socioeconomic Environmental Studies
6. Scientific and Technical Assessment Reports (STAR)
7. Interagency Energy-Environment Research and Development
8. "Special" Reports
9. Miscellaneous Reports

This report has been assigned to the ENVIRONMENTAL PROTECTION TECHNOLOGY series. This series describes research performed to develop and demonstrate instrumentation, equipment, and methodology to repair or prevent environmental degradation from point and non-point sources of pollution. This work provides the new or improved technology required for the control and treatment of pollution sources to meet environmental quality standards.

This document is available to the public through the National Technical Information Service, Springfield, Virginia 22161.

EPA-600/2-80-053
March 1980

DESIGN AND PERFORMANCE OF AN AEROSOL MASS DISTRIBUTION MONITOR

by

W. Stöber, F. J. Mönig, H. Flachsbart and N. Schwarzer
Fraunhofer Institute of Toxicology and Aerosol Research
5949 Grafschaft-Sauerland
and
4400 Münster-Roxel
Federal Republic of Germany

Research Grant No. 803592

Project Officer

Jack Wagman
Director, Emissions Measurement and Characterization Division
Environmental Sciences Research Laboratory
Research Triangle Park, North Carolina 27711

ENVIRONMENTAL SCIENCES RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U. S. ENVIRONMENTAL PROTECTION AGENCY
RESEARCH TRIANGLE PARK, NORTH CAROLINA 27711

DISCLAIMER

This report has been reviewed by the Environmental Sciences Research Laboratory, U. S. Environmental Protection Agency, and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the U. S. Environmental Protection Agency, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

ABSTRACT

The aerosol mass monitor has been built to measure the masses of non-volatile aerosols in the range of 0,05 to 5 μm aerodynamic particle diameter. The instrument consists of a newly designed spiral duct aerosol centrifuge equipped with highly sensitive quartz sensors for in situ weighing of the deposited aerosol masses. The instrument further includes a clean air device for maintaining constant aerosol flow conditions, and electronic parts for the operation of the quartz sensors. The mass of aerosol deposited on the quartz crystals is automatically measured by an electronic counter while the rotor of the centrifuge continues spinning. The data are handled by a microprocessor.

This report was submitted in fulfillment of Research Grant No. 803592 by the Fraunhofer Institute of Toxicology and Aerosol Research under the sponsorship of the U. S. Environmental Protection Agency. This report covers the period from January 19, 1977, to April 1, 1979, and work was completed as of November 30, 1979.

CONTENTS

Abstract iii
1. Introduction 1
2. Summary. 3
3. Recommendations 4
4. General Description of Instrument 5
4.1 Aerosol Centrifuge 5
4.1.1 Centrifuge Rotor 5
4.1.2 Driving Unit 10
4.1.3 Clean Air Unit. 10
4.1.4 Method of Operation 15
4.2 Mass Measurement System 20
4.2.1 Quartz Sensors 20
4.2.2 Rotor Electronics 20
4.2.3 Inductive Power Supply 26
4.2.4 Control Electronics 26
4.2.5 Frequency Measurement 29
4.2.6 Principle of Piezoelectric Microbalance . 30
4.2.7 Aerosol Concentration Measurement 31
4.3 Microprocessor 31
5. Handling of Quartz Inserts 34
6. Calibration 35
7. Instrument Operation 38
8. Instrument Performance 40

References 45

1. INTRODUCTION

In most applied research projects and in many routine investigations of airborne material like ambient particulate air pollutants, emission sources, mine dusts or other particulate matter suspended in gases, the particle size distribution in the airborne state is the most important information to be acquired. In the analysis and abatement of air pollution, the particle size distribution determines the degree of penetration of aerosol filters by the airborne material, influences visibility through polluted air and affects the aerodynamic stability and life time of aerosols, smog and other particulate clouds. With regard to health hazards, the size distribution rules the respirability of particulate air pollutants and, thus, governs the potential toxic effects of airborne material.

Since most aspects of air pollution are related to the respective masses of the pollutants, experimental data directly giving the corresponding particle mass distribution of the suspended matter would be most desirable. Thus, the most preferable measurement is by gravimetric procedures. A number of instrument types, most notably cascade impactors, comply with this requirement and yield gravimetric information of size fractions of precipitated airborne material. However, a number of practical drawbacks like low sensitivity, wall losses, load limitation, and, most significantly, batchwise operation requiring tedious sample weighing limit the applicability of this device. Clearly, an instrument operated as a continuous monitor of mass distributions of airborne particulate matter would be highly desirable. No such instrument, however, is available as of now.

Therefore, the principal objective of this project was to design,

build and test an instrument which continuously samples airborne particulate matter, precipitates the particles under size separation and monitors the particle mass distribution of the deposit without interrupting the sampling process.

A subordinate objective of this project is to facilitate the collection of size-selective deposits of particulate matter of very low airborne concentration for subsequent physical and chemical analysis by X-ray fluorescence, microprobe, electron microscopy, neutron activation, wet chemical processing, etc. In this case, the monitoring system will be used to ensure adequate sampling times by observing the build-up of the required amounts of deposited mass.

2. SUMMARY

Under the terms of the grant, a monitoring device for the gravimetric determination of aerosol mass distribution was built. The instrument combines the size-separating capability of a spiral duct aerosol centrifuge with the mass sensor function of a thin piezo-electric quartz crystal. The arrangement is to permit telemetric read-outs during sampling and has a total of 13 size-selective sensors and one reference crystal.

3. RECOMMENDATIONS

Till now the mass monitor only measures surface concentrations, referred to equal length intervals of the channel, given by the crystal diameter. In order to get the more interesting aerosol mass concentrations and an aerosol mass spectrum referred to equal particle diameter intervals further calculations are necessary. This can be done by another program for the microprocessor and by connecting a plotter to it.

4. GENERAL DESCRIPTION OF INSTRUMENT

The design concept of the projected particulate mass distribution monitor is a combination of two recently developed technologies in aerosol instrumentation:

1) aerodynamic aerosol size spectrometry in the spiral duct of the rotor of a centrifuge (1,2,3,4),

2) electronic gravimetry of mass deposits by means of oscillating quartz crystal detectors (6,7,8,9).

An overall view of the instrument is shown in Figure 1.

4.1 AEROSOL CENTRIFUGE

4.1.1 Centrifuge Rotor

The rotor consists of the spiral duct plate and the lid with the inlet system. The rotor is mounted on the motor shaft and held in place by one left-handed nut (Figure 2). The rotor contains the spiral duct (length 151 cm, depth 4 cm), the laminator, the aerosol inlet, and 2 filters; one at the entrance of the laminator, and one at the end of the spiral duct, as shown in Figure 3. In the outer wall of the duct, there are the 14 quartz mounting inserts. All these parts can be easily removed. The oscillators and some additional electronic circuits are mounted on a print board at the bottom of the rotor. At this end the rotor is closed by a disc of print board material (Figure 4). In the center of this disc one half of the core transformer for the inductive power supply is mounted. The ring electrode in the middle of the disc is part of a condensor, which couples the high frequency signals of the quartz oscillators to the counter.

The spiral duct is closed by a lid which contains the inlet

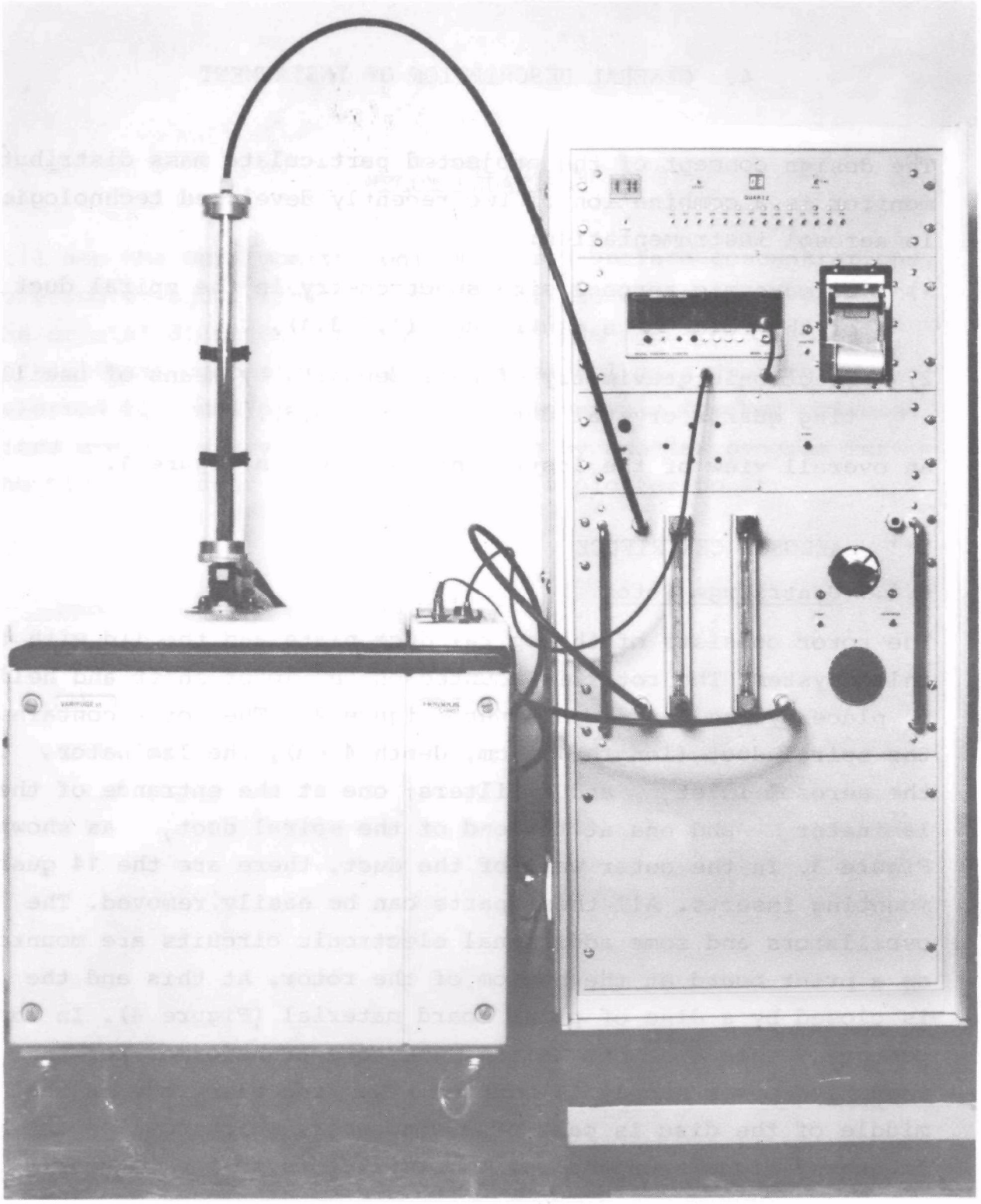

Figure 1 Complete mass monitor

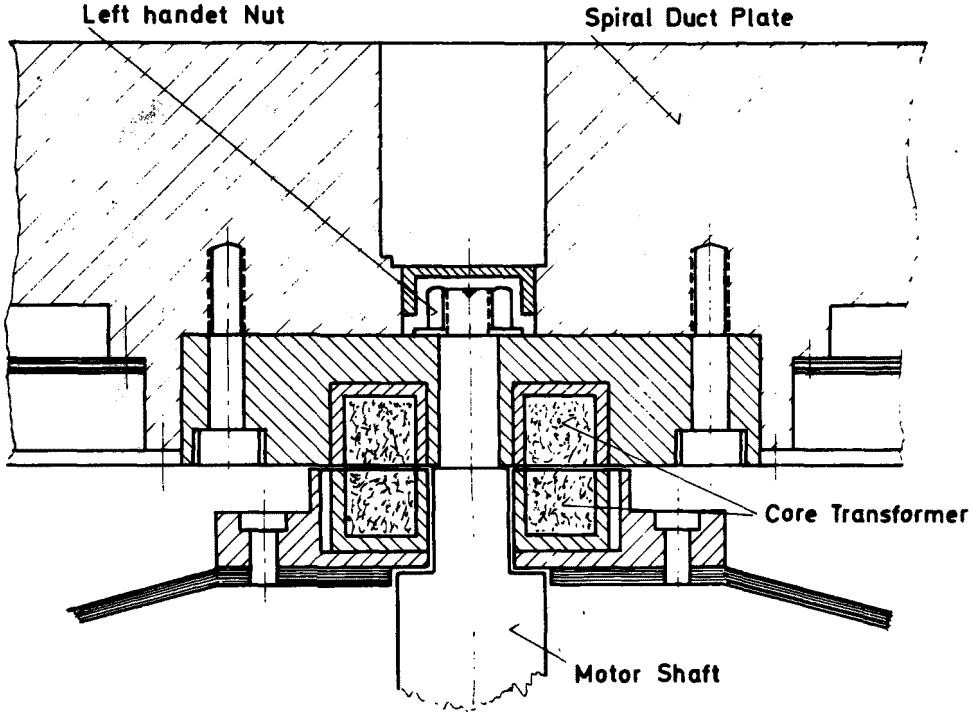

Figure 2 Vertical section of mounted centrifuge rotor

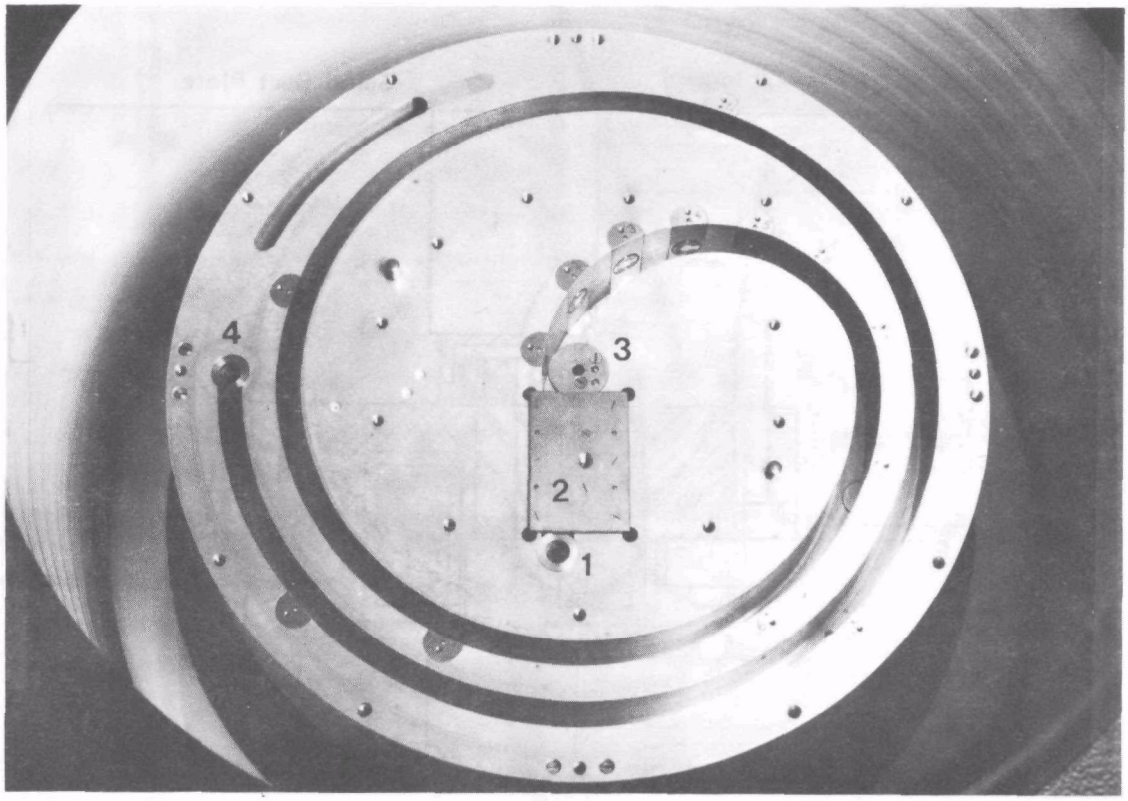

Figure 3 Centrifuge rotor with spiral duct and quartz inserts, filters, (1) and (4), Laminator, (2) and aerosol inlet, (3)

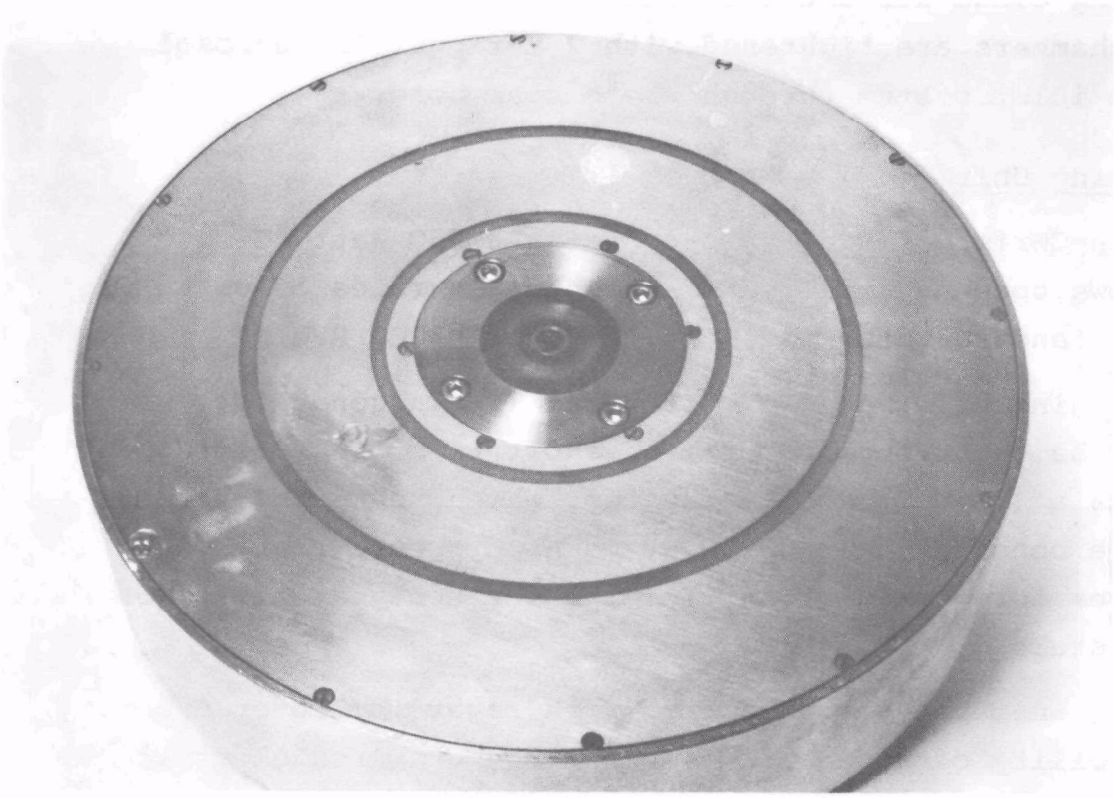

Figure 4 Rotor lower side

system (Figure 5). A vertical section of the inlet system is shown in Figure 6. With regard to rotation it consists of two parts: one part is rotating within the bearing, the other one is mounted on the outer ring of the bearing and does not rotate. In the center of the unit there are two chambers; the upper one for the incoming clean air and the lower one for the total flow. Both of these chambers are tightened with 2 V-rings. The aerosol enters the inlet system through the center bore.

4.1.2 Driving Unit

The Heraeus-Christ driving unit contains a 650 Watt DC-Motor which allows speed stabilized rotor spinning rates of up to 6000 RPM. Standard rotor speed is, however, 3000 RPM.

After switching off the motor, it works like a generator, which can brake the centrifuge rotor in a short time. The driving unit further has a 150 Watt cooling system, which keeps the rotor-temperature constant near 20 ^{o}C by means of convection. Constant rotor temperature is important for the frequency stability of the quartz crystals.

The driving unit has been modified with regard to rotor mounting, higher stability of speed and temperature, and less electric noise.

4.1.3 Clean Air Unit

For operating the quartz centrifuge, a device producing and controlling clean air or carrier gas is necessary (Figure 7). The gas flow of this unit is shown schematically in Figure 8. The closed circuit gas flow is achieved by a magnetic piston pump, C, which is controlled by a regulating transformer. The carrier gas is drawn by the pump from the end of the spiral duct, F, passing a 4-way-valve in position "2" and two silicagel columns, 2, then passing a glassfiber filter,1, into the origin of the spiral duct,G,via the centrifuge inlet system,H.

Figure 5 View of closed rotor with inlet system and driving unit

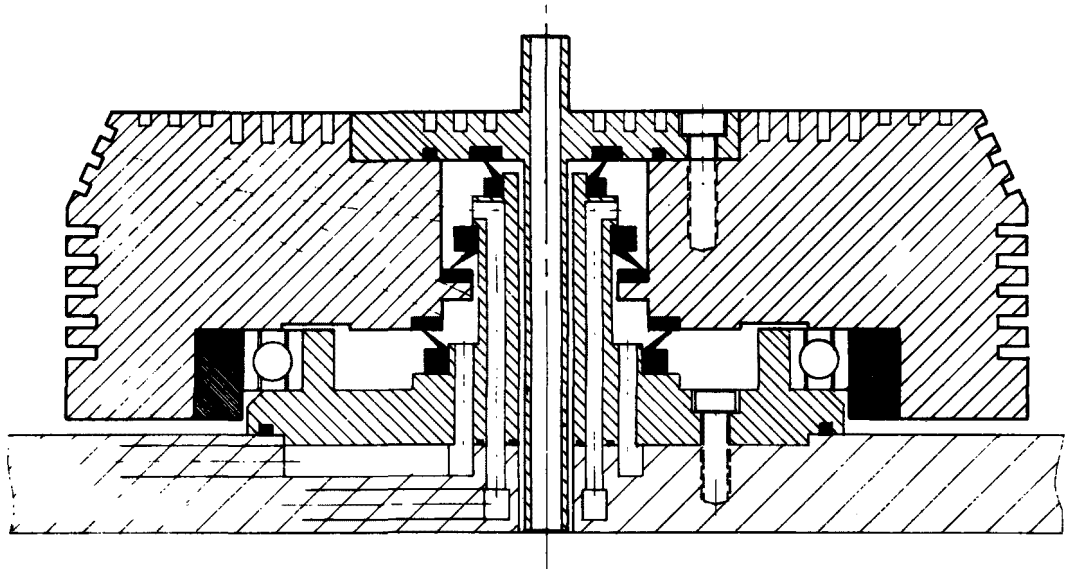

Figure 6 Vertical section of inlet system

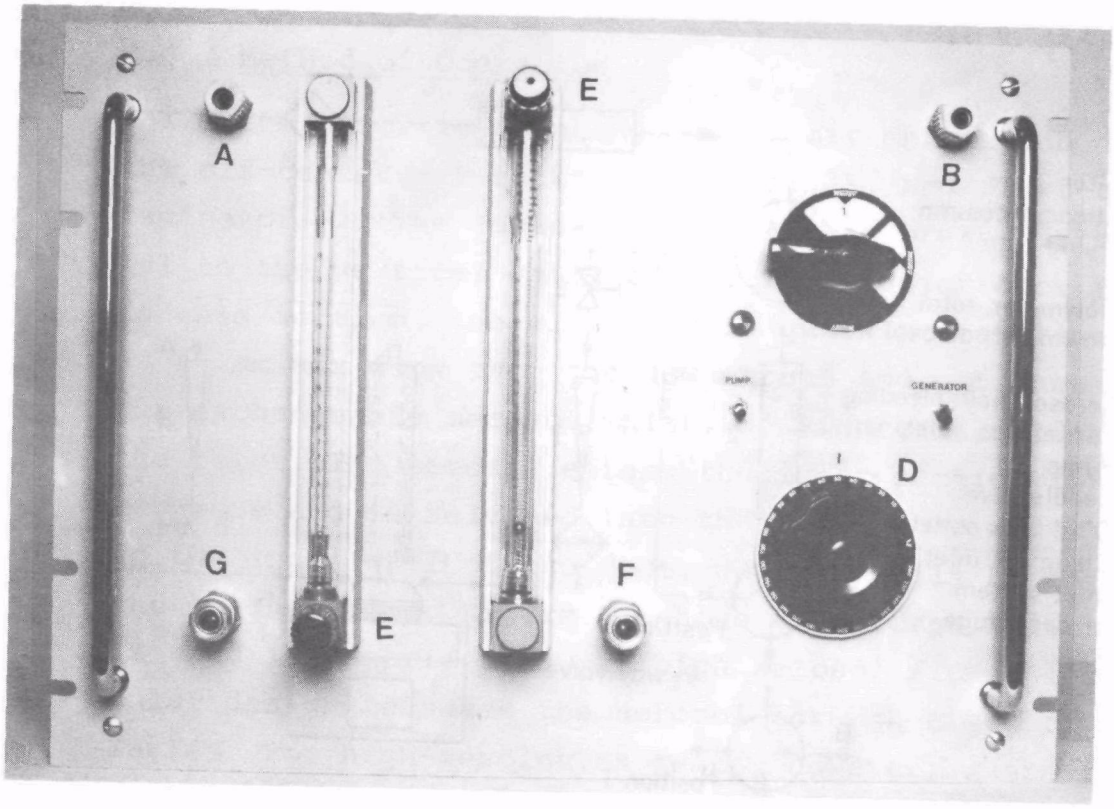

Figure 7 Front panel of clean air device

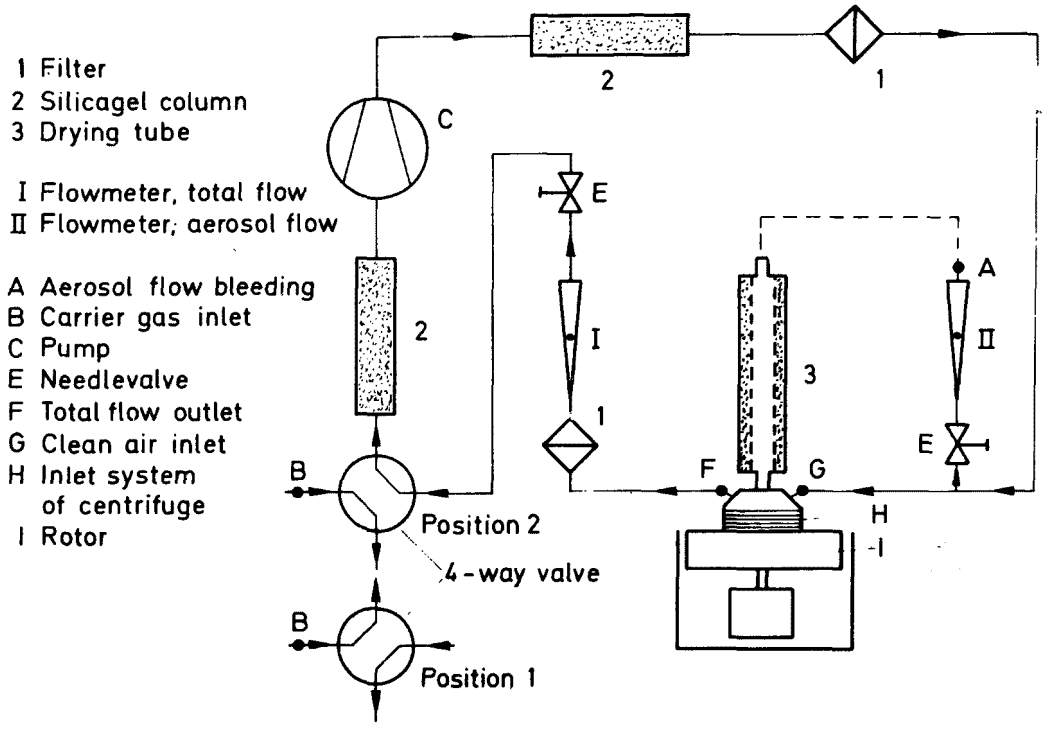

Figure 8 Flow schematic of clean air device

By regulating the needle valve at flow meter II, carrier gas is bleeded from the closed circuit gas flow and replaced by aerosol containing gas thus establishing the desired sampling rate.

When aerosols with high humidity are to be sampled the drying tube, 3 (Figures 8 and 9), should be mounted to the aerosol inlet of the centrifuge.

4.1.4 Method of Operation

While the rotor is spinning, clean air enters the duct through the off-center air inlet, 1 (Figure 3), and flows to the inserted laminator, 2. The laminator has 11 thin brass foils mounted parallel to the vertical walls of the rectangular duct (Figure 10). In this section, the clean air is quickly stabilized and emerges as a laminar flow from the downstream end. It then approaches the exchangeable aerosol inlet, 3 (Figure 3), at the center of the rotor. The aerosol enters the inlet section through the center bore and is released into the duct as an air sheath parallel to the inner wall of the duct, where it will be entrained into the laminar flow. Figure 11 shows an aerosol inlet section which is used in experiments with high aerosol flow rates. This particular design releases the aerosol through a set of short capillaries. For high-resolution tests, however, where reduced aerosol flow rates are required, another design is employed which has a narrow slit adjacent to the inner wall. This aerosol inlet is shown in Figure 12.

When leaving the center of rotation, the aerosol particles are subjected to the centrifugal forces of the spinning rotor and start moving in a radial direction across the laminar air stream. Their trajectories are determined by the operating conditions of the centrifuge and the aerodynamic size of the particles. Thus, while the air is flowing down the duct to the outlet, 4 (Figure 3), the particles are settling, according to their size, in different locations on the outer wall with the quartz inserts.

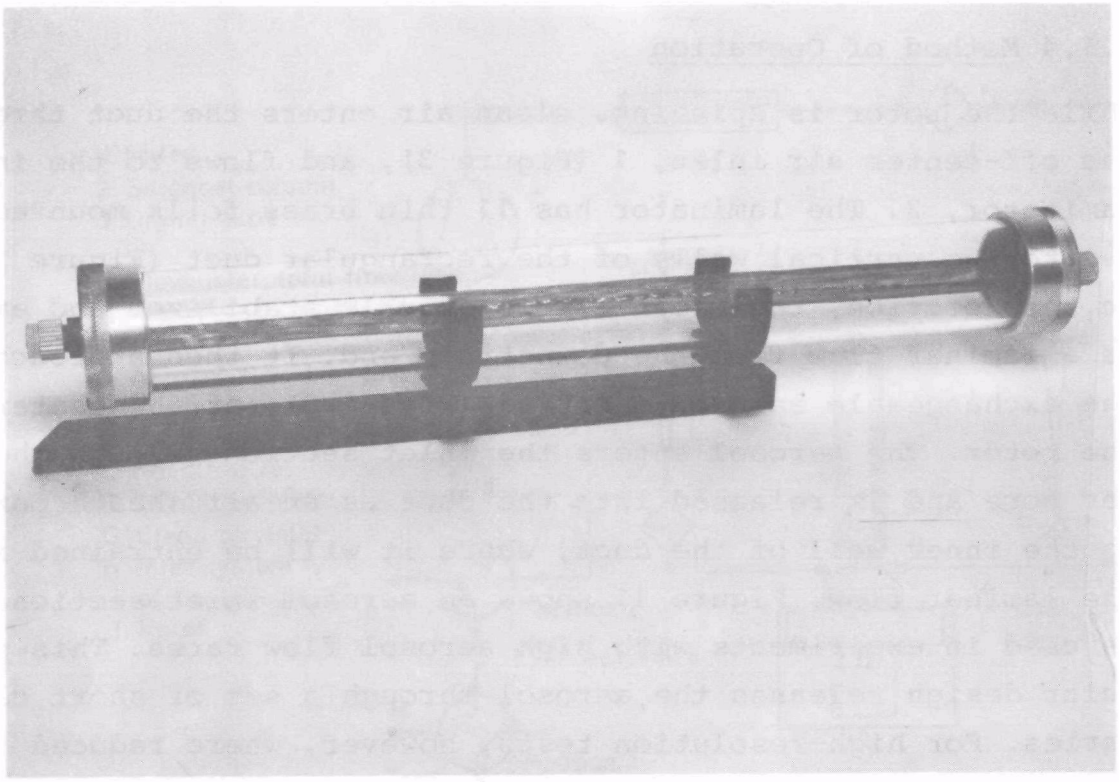

Figure 9 Drying tube

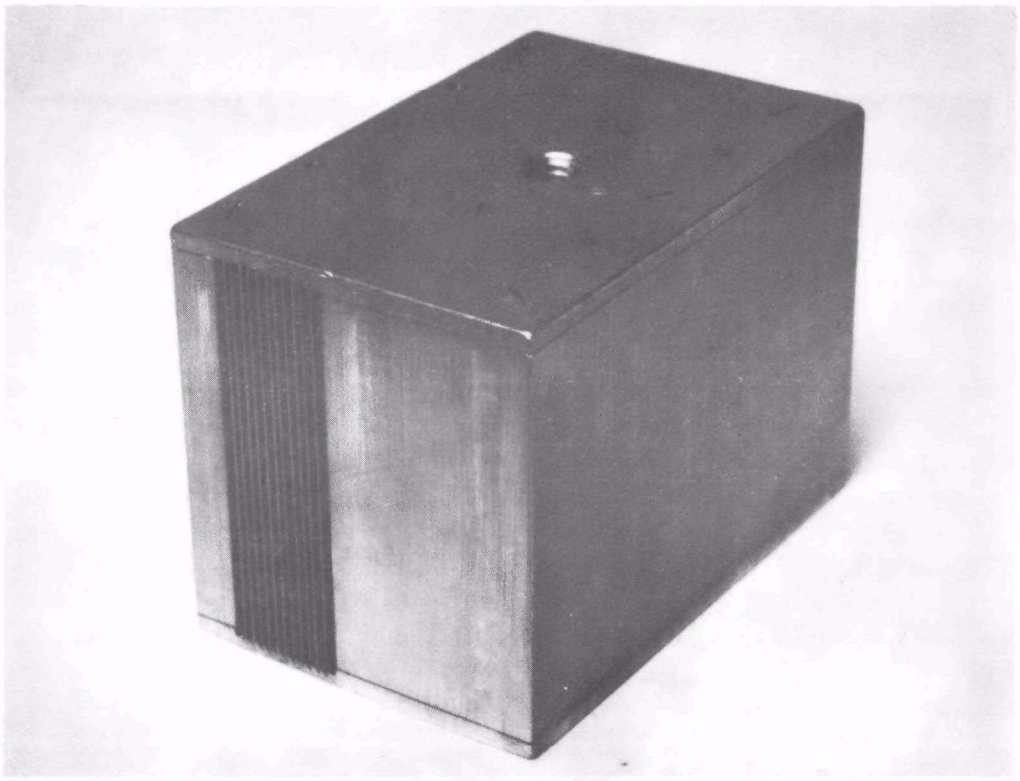

Figure 10 Laminator

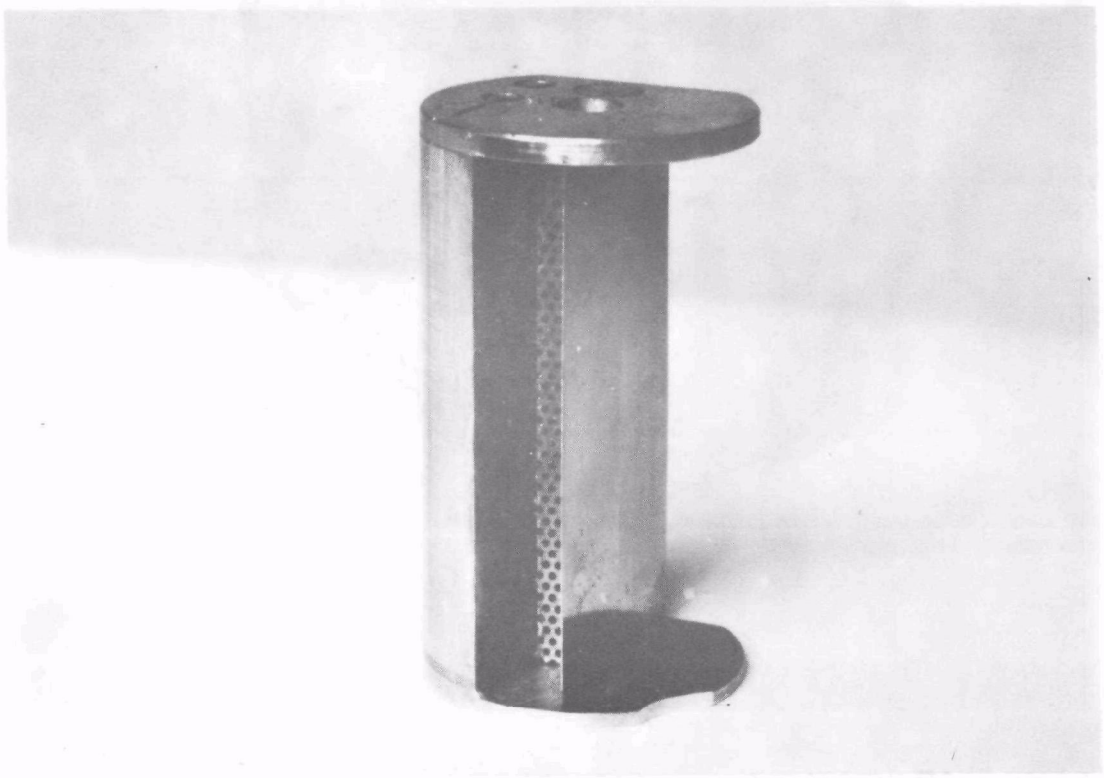

Figure 11 Aerosol inlet (low resolution)

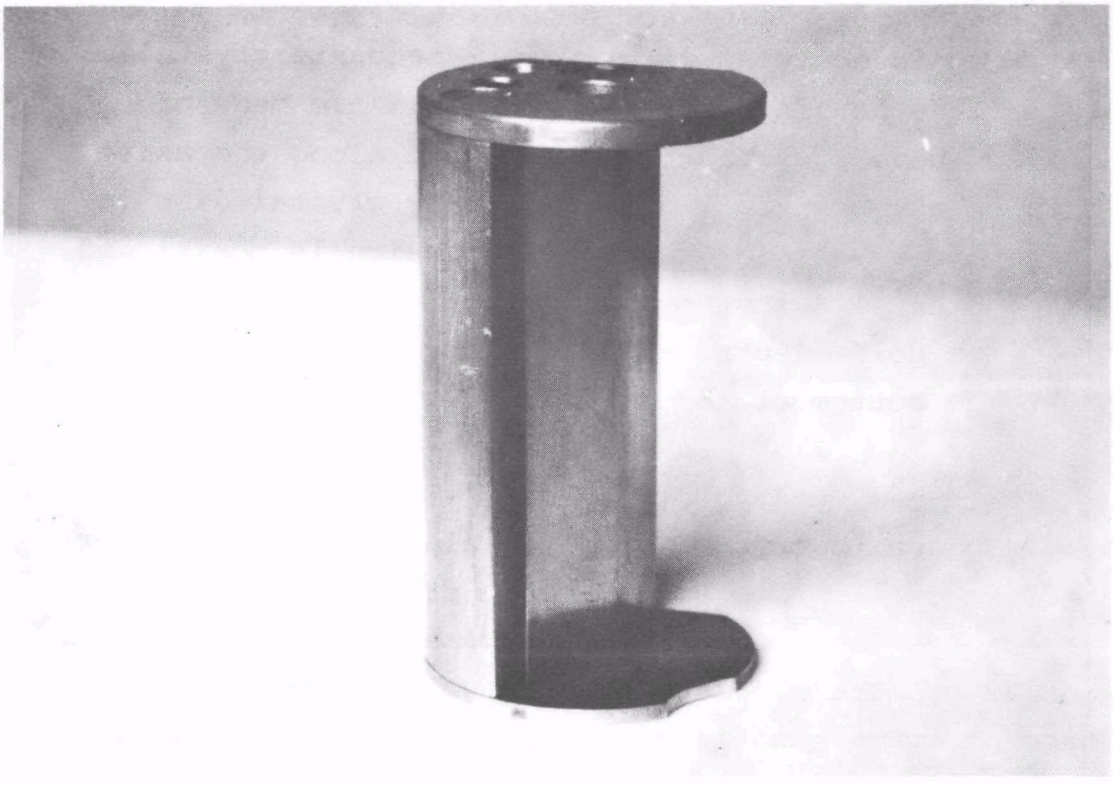

Figure 12 Aerosol inlet (high resolution)

4.2 MASS MEASUREMENT SYSTEM

4.2.1 Quartz Sensors

The centrifuge rotor is equipped with 14 10-MHz-quartz crystals, 12 mm in diameter. The housings of these crystals are made of stainless steel and have a cylindrical shape cut along the axis by the spiral duct, as shown in Figure 13. Every crystal disc is mounted on a high quality 4 pin quartz holder which is positioned flush in a hole in the stainless steel insert. Two of the pins are connected to the gold electrodes of the crystal and the 2 pin connector at the bottom of the insert.

4.2.2 Rotor Electronics

For operation every quartz crystal needs an electronic oscillator circuit. The quartz oscillators are of the Pierce-Colpitts type (Figure 14). They have 3 stages: oscillator, separator and output. A photograph of a complete oscillator is shown in Figure 15. The oscillator stage of every oscillator unit is permanently activated and all crystals are vibrating. Read-out selection of the oscillators (5) is done one at a time by an electronic counter (Figure 16). This counter has 14 outputs by which the second and third stage of the chosen oscillator unit is switched on. The 10 MHz output signal is controlled by an electronic switch, which is part of every oscillator unit, and the HF output amplifier which is connected to the ring condenser at the bottom of the rotor. As the distance between the two ring electrodes is 4 mm only the 10 MHz signal can easily be coupled to the non rotating part of the ring condensor which is shown in Figure 17.

Clock and reset pulses for the oscillator selector are generated by two special band pass circuits which are activated via the

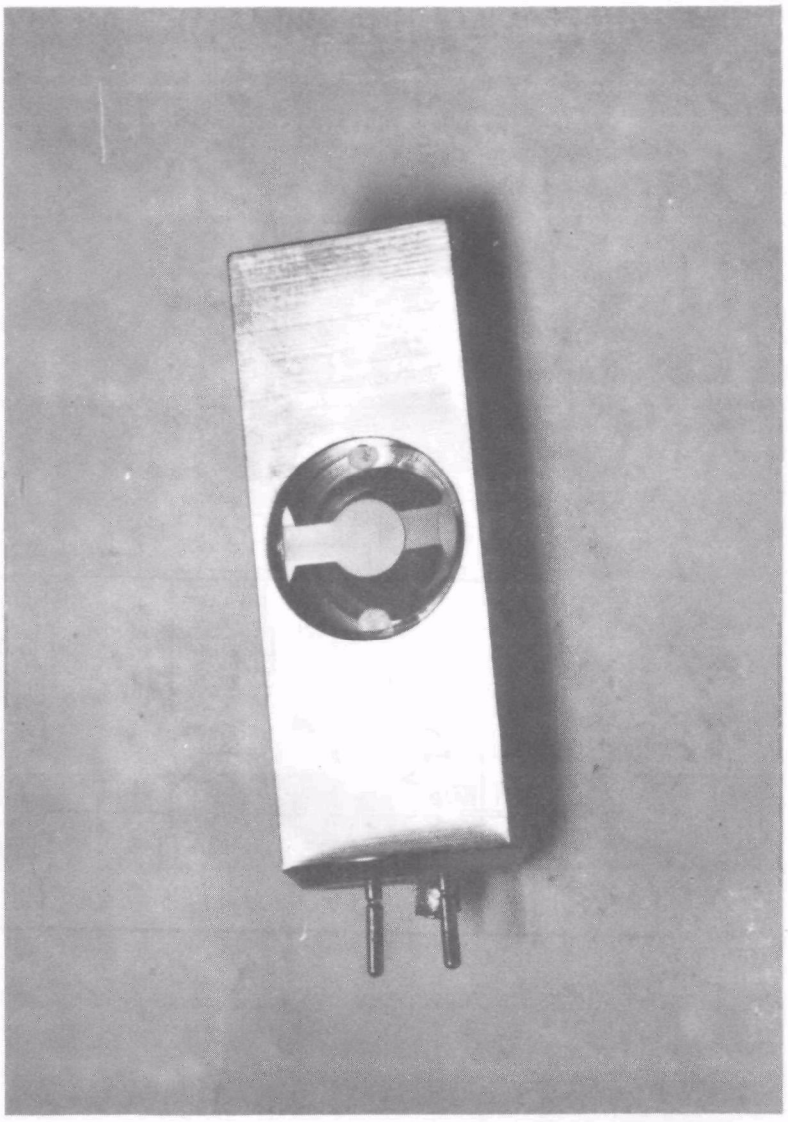

Figure 13 Insert with quartz crystal mounted on 4 pin holder

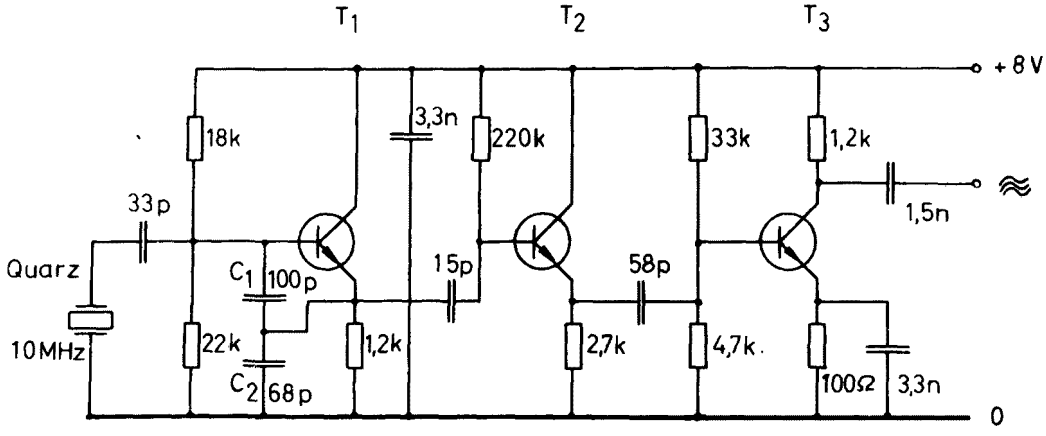

Figure 14 Pierce Colpitts oscillator

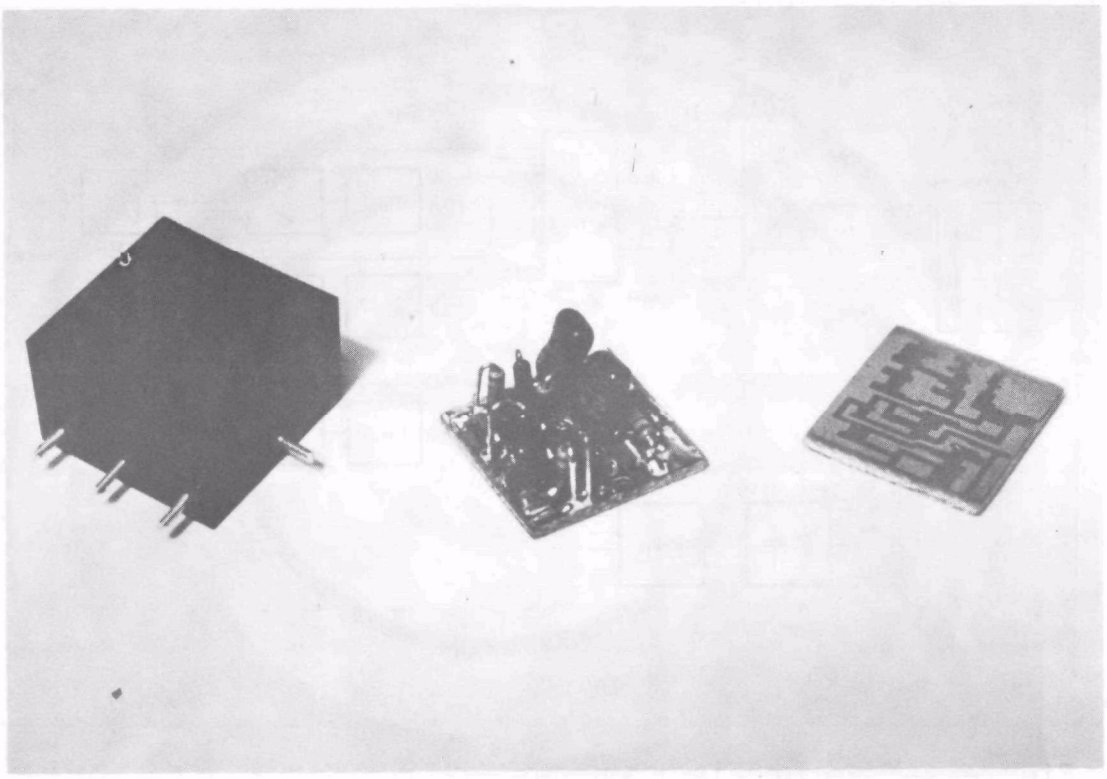

Figure 15 Oscillator construction

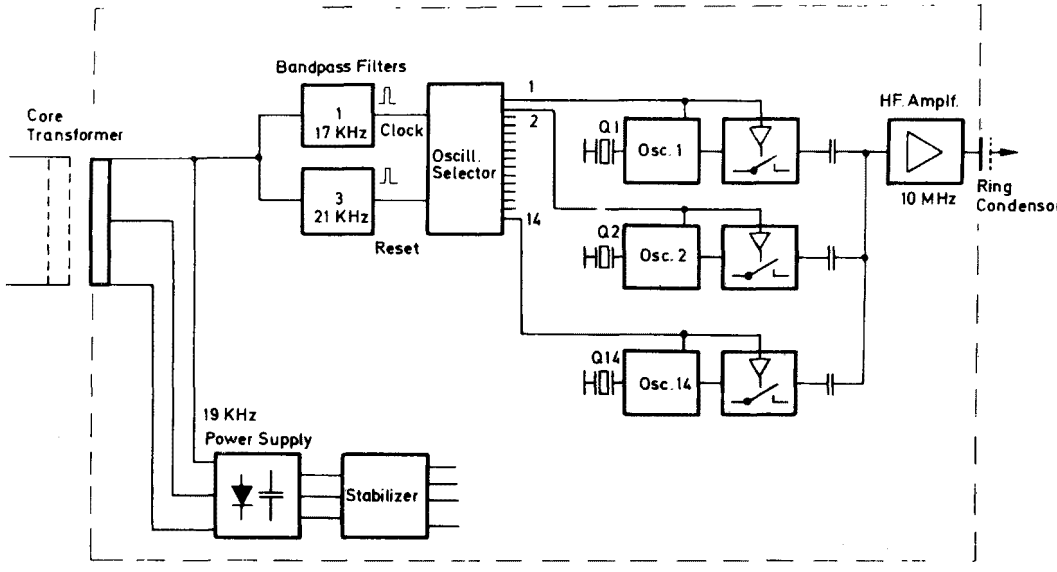

Figure 16 Rotor electronics

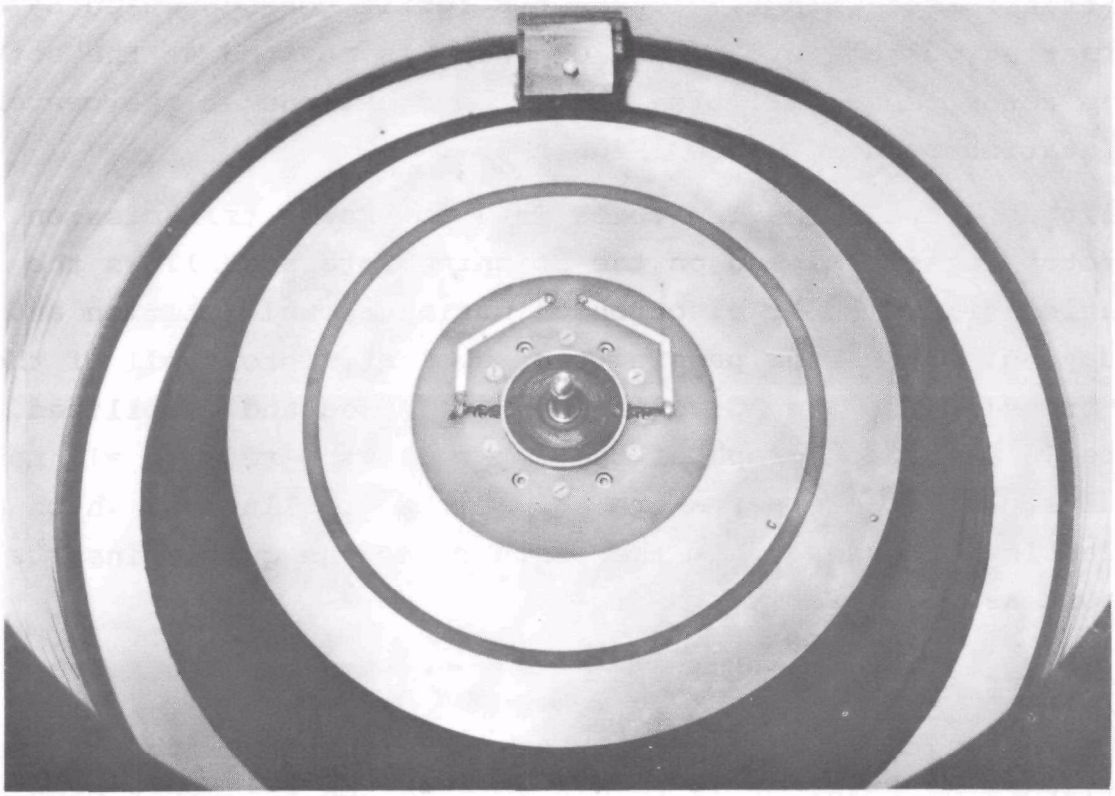

Figure 17 Disk with ring condensor half (second ring), core half and motor shaft

core transformer when the operation frequency of the rotor supply voltage is changed from 19 KHz to 17 KHz or 21 KHz, respectively, for some milliseconds.

4.2.3 Inductive Power Supply

The power consumption of all rotor circuits is 1.5 watts only. So it was possible to build an inductive power supply. It consists of a transformer with two cup-shaped ferrit cores powered by an alternating voltage of 19 KHz. One core is mounted at the bottom of the rotor center (Figure 4) and the other one at the center of a stationary disc below (Figure 17).

The airgap between the two cores is 1 mm. Power transmission to the rotor doesn't depend on the spinning rate and allows the operation of the rotor electronics during actual rotation and standby as well. After passing the secondary rotor coil of the core transformer, the AC-voltage is rectified and stabilized. Figure 18 is a photograph of the rotor printboard with all rotor circuits. The square parts are the quartz oscillators, which are mounted in such a way that the distance to the quartz inserts is as short as possible.

4.2.4 Control Electronics

This part has two selective counters: one for choosing the quartz oscillators by means of 14 toggle switches on the front panel of the control unit, and the other one for choosing the measuring time (Figure 19). This is the time in which the 10 MHz signal of the chosen oscillator can pass from the oscillator to the frequency counter. The measuring time can be set between 0.5 and 99 minutes.

The rotor-mounted oscillator selector is synchronized to the selective quartz counter in the control electronics. This is effected by a reset pulse (manually, or automatically after reading quartz 14) which brings both circuits into start position.

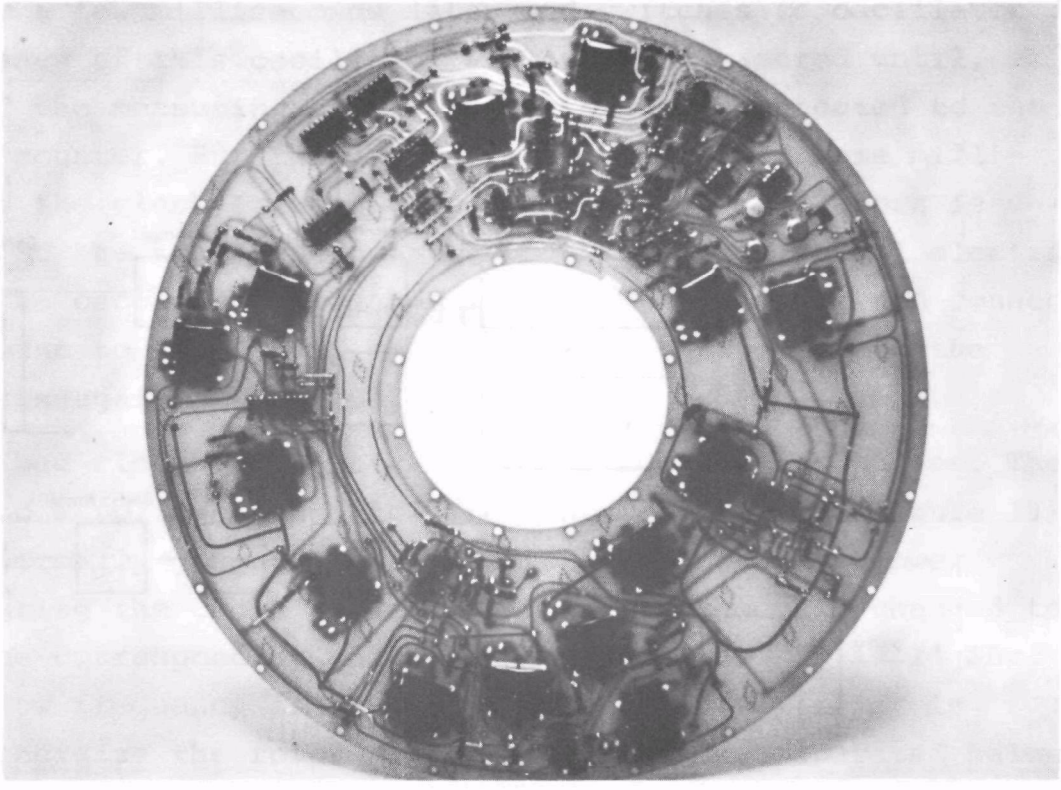

Figure 18 Rotor print board

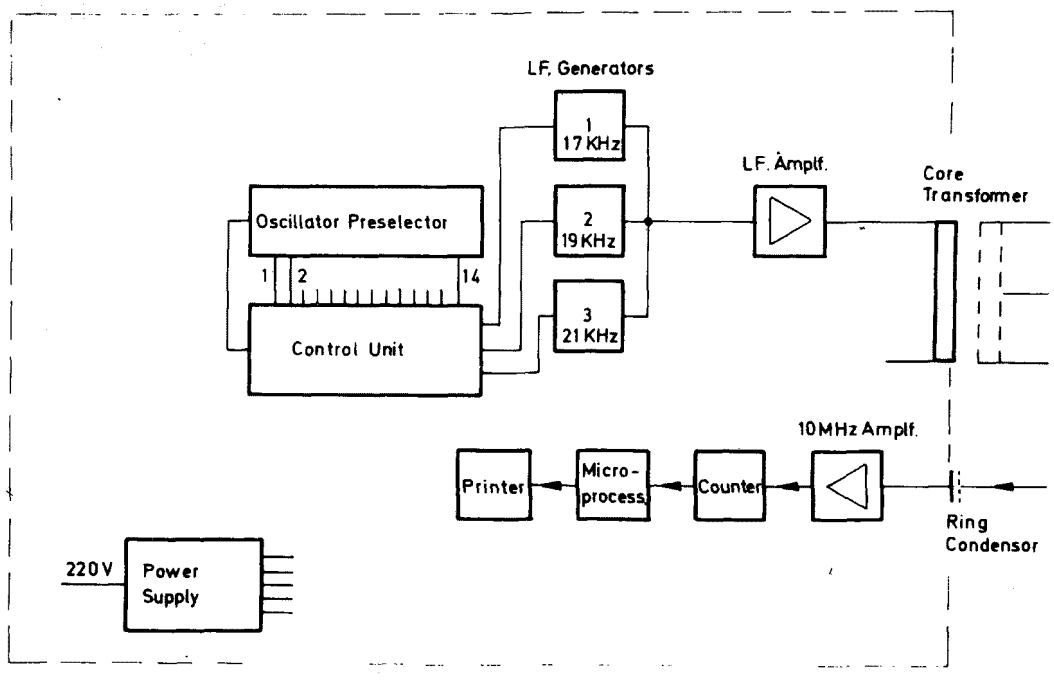

Figure 19 Principle of control electronics

If, for instance, oscillators 1 and 3 are selected (by means of toggle switches 1 and 3 at the front panel), the first clock pulse connects oscillator 1 to the frequency counter. At the end to the preset measuring time, the second clock pulse is generated and, in the same way, effects oscillator 2. But, since this oscillator has not been selected the next clock pulse is generated only a few milliseconds later and switches to oscillator 3. The frequency of this oscillator will now be measured until, at the end of the measuring time, oscillator 4 is connected to the frequency counter. But this again is only done for some milliseconds as the clock frequency for non-chosen oscillators is $\sim$ 300 Hz. So the selective quartz-counter in the control electronics and the oscillator selector in the rotor quickly and consecutively count to 14. Then a reset pulse is generated and the cycle starts again.

The described clock and reset pulses are rectangular pulses. They are sent to a voltage tuned low frequency oscillator (Figure 19) which is normally generating 19 KHz for the inductive power supply. During the clock pulse time this frequency is changed to 17 KHz; the corresponding value for the reset pulses is 21 KHz. Then the low frequency is amplified in a power amplifier in order to energize the rotor circuits via the two separated halves and the airgap of the core transformer.

4.2.5 Frequency Measurement

After passing the ring condenser at the bottom of the rotor, the signal of the selected quartz oscillator passes to a selective 10 MHz amplifier. This amplifier is necessary to get a good signal amplitude for the frequency counter and to suppress the noise of the driving unit. Frequency measurements are made by means of a Ballentine frequency counter. It has a high stability time base with temperature stability of better than $2 \cdot 10^{-9} \cdot {}^{o}C^{-1}$. Frequency measurements with a resolution of 0.1 Hz can be made within 10 seconds.

However the shortest measuring time for a single quartz oscillator depends upon the control electronics and is at least 0.5 minute. Since there is no synchronisation between counter time base and clock pulses of the control electronics, at least 2 complete frequency measurements of one oscillator are made within 0.5 minute. The last of these frequency values is stored in the microprocessor. The shortest measuring time for a complete run of 13 deposit sensors and 1 reference crystal is 7 minutes.

4.2.6 Principle of Piezoelectric Microbalance

A quartz crystal disc, ground along appropriate crystallographic planes (AT-quartz) and used as a component of an electronic oscillator circuit, will vibrate with tangential amplitudes along the face of the disc. The mechanical frequency of these oscillations is determined by the material properties and the dimensions of the crystal. In particular, the thickness of the disc determines the individual resonance frequency of the crystal.

For surface layers on the crystal, a theoretical relationship

$$\Delta f = -\frac{N}{d^2 \cdot \rho} \cdot \frac{\Delta m}{F} \quad \text{where } f = \frac{N}{d}$$

predicts a frequency shift Δf of the resonance frequency f as an effect of a layer of total mass Δm on the active surface area F of the crystal of thickness d. As material constants, the frequency constant $N = 167\ \text{kHz} \cdot \text{cm}$ and the density $\rho = 2{,}65\ \text{g} \cdot \text{cm}^{-3}$ of quartz also enter the equation. Thus, the surface concentration

$$\varphi = \frac{\Delta m}{F} \qquad (1)$$

is linearly related to the frequency shift Δf by a proportionality factor

$$\frac{1}{C_f} = -\frac{d^2 \cdot \rho}{N} \qquad (2)$$

which, for a given material, depends only upon the crystal thickness d, and the surface concentration is

$$\varphi = \frac{1}{C_f} \cdot \Delta f \qquad (3)$$

For a 10 MHz AT-quartz of 0,0167 cm thickness

$$\frac{1}{C_f} = 4.4 \cdot 10^{-9}\ g \cdot cm^{-2} \cdot Hz^{-1}$$

and a surface concentration

$$\varphi = 4.4\ ng \cdot cm^{-2}$$

will be caused by a frequency shift of $\Delta f = -1$ Hz.

The factor $\frac{1}{C_f}$ is the gravimetric constant. To measure the surface concentration, only the shift Δf of the oscillator frequency has to be known. This frequency shift is the difference of two absolute frequency values which have been measured by the electronic counter in a given time interval.

4.2.7 Aerosol Concentration Measurement

In order to obtain aerosol mass concentrations and a mass spectrum instead of surface concentrations, further calculations are necessary. Surface concentrations refer to equal length intervals of the spiral duct, while size-related mass concentrations refer to equal intervals of particle diameter. Therefore, a faktor K_F for every crystal position must be calculated from the slope of the (centrifuge) calibration curve. These values are shown in table 1.

Thus the size-related mass concentration K belonging to one crystal is

$$K = \varphi \cdot F \cdot K_F \cdot \frac{1}{V_{ae}} \qquad (4)$$

where F is an area represented by quartz diameter and channel depth and V_{ae} is the volume of the sampled aerosol.

4.3. MICROPROCESSOR

All data, that are important for the calculations, are received by the microprocessor MP from the control electronics and the

TABLE 1. K_F VALUES FOR CALCULATION OF AEROSOL CONCENTRATION

Quartz Oscillator	Factor K_F for equal particle diameter intervals of $1 \mu m$ (i.e., $\frac{\Delta \text{ length}}{\Delta \text{ diameter}}$)
02	0.6
03	1.4
04	2.9
05	5.2
06	7.8
07	12.8
08	25.5
09	49.2
10	76.9
11	133.0
12	293.6
13	402.6
14	521.9

TABLE 2. MP PROGRAM COMBINATIONS

Mode	Program
0	DFI + QL
1	DFI + DF
2	DFI
3	DFI + Pause
4	QL
5	QL + Pause
6	DF
7	DF + Pause
8	DFI + QL + DF
9	DFI + QL + DF + Pause

frequency counter. These data are: quartz number (2 digits), quartz frequency (9 digits), measuring time (4 digits), and mode of operation (1 digit). Data transfer is made just before the measuring time of an oscillator ends.

When the microprocessor is started, it first stores the absolute resonance frequency of a chosen crystal and the printer prints the crystal number followed by the frequency value in Hz, and measuring time in minutes (referring to program start). This is repeated until the resonance frequencies of all chosen crystals are printed. Then the MP "looks" at the position of the mode switch to decide which type of program is to be run. Table 2 shows the possible types of programs and program combinations.

Program "DFI": The MP compares the actual frequency value of a crystal with the first stored value and calculates the difference Δf. This difference is an integral or cummulative value and, therefore, gets the MP name "DFI". The unit of DFI is Hz. When particles settle down on the crystal surface, the resonance frequency drops. In this case, DFI is counted as a positive value. If however the frequency rises, which can be caused by decreasing humidity of the winnowing air, the DFI value decreases and the printed value gets a minus sign by the MP, showing that this is a false value.

Program "QL": In this program the surface concentration φ according to Eq. 3 is calculated. The result has the units $10^{-9} g \cdot cm^{-2}$, but only "NGR" is printed because of the limited character number of one printer line. QL means "Quartz Layer" and is an MP name for φ. If the DFI value was negative, the mass value gets an interrogation mark for a negative mass value.

Program "DF": In contrast to DFI, DF is a differential value, refering to the last frequency value.

Program "Pause": If the number of printed data becomes too large, a special pause program can be chosen in mode positions 3, 5, 7 or 9. The pause program automatically starts when the absolute frequencies have been printed.

The cycle time of the pause program is calculated by multiplying the measuring time in minutes with the constant 56 stored in the MP. If for example the shortest measuring time of 0.5 minutes is chosen the shortest cycle time of the pause program is 28 minutes. In the first quarter of this time all the 14 values are printed, then calculating and printing is stopped for 21 minutes, and after this pause the cycle starts again. The pause program works well when 1, 2, 4, 7, 8, or 14 crystals are switched on; for other numbers the pause cycle time of 2 corresponding crystals no longer is a constant.

5. HANDLING OF QUARTZ INSERTS

Several measurements showed that, for particle diameters above 1 μm, a coating of the crystals is necessary in order to get a good mechanical coupling of the deposited particles to the crystal surface. Coating is done by applying some microliters of an adhesive dissolved in cyclohexane to the cleaned crystal surface and removing the excess with filter paper. After drying the crystals in an oven at 80 ^{o}C for 30 minutes, the inserts can be put into the grooves of the spiral duct.

The frequency shift caused by coating the crystals should be 2000 to 3000 Hz.

As the size tolerances of the inserts are very small, the walls of the grooves should get a thin film of a vacuum grease every now and then.

The useful range of frequency shift is 2000 Hz for uncoated crystals and about 5000 Hz for coated ones. In order to obtain

long operating times, coating of all crystals is possible.

When approaching the end of the useful range of frequency shift, the inserts must be cleaned in an ultrasonic bath (20 to 40 KHz) of cyclohexane for 1 to 3 minutes. After this they are carefully rinsed with pure cyclohexane and ethanol and then blown dry with compressed air.

If the crystals are very dirty, they can be cleaned by gently wiping them with a cyclohexane soaked cotton swab.

However, care should be taken that the gold electrodes of the crystals will not be damaged.

6. CALIBRATIONS

For calibration, a thin brass foil was inserted along the outer channel wall, and the aerosol centrifuge was operated at the standard spinning rate of 3000 RPM, while the total flow was 11.5 $L \cdot min^{-1}$ and the aerosol flow 1 $L \cdot min^{-1}$. Fluorescein served as a test aerosol. At the locations of the crystals, small samples were cut out of the foil, and the particle diameters were measured by means of a scanning electron microscope. The result is shown in table 3. The diagram, Figure 20, shows the calibration curve with aerodynamic particle diameters for quartz positions Q2 to Q14 plotted vs channel length. The accuracy of the diameter values is estimated to be better than 5 percent.

A calibration of the quartz crystals is not necessary because the surface concentration depends only on the gravimetric constant $\frac{1}{C_f}$ and the shift in frequency Δf, as given in Eq. 3. This implies that the particles are evenly distributed over the crystal surface and well coupled, which was check by some quantitative tests with low (80 $\mu g \cdot m^{-3}$) and high (1 $mg \cdot m^{-3}$) airborne concentrations of fluorescein aerosols. In these measurements, the frequency shifts of the quartz sensors were found to be proportional to the deposited masses on the crystals. Photometrically determined mass values were within less than 15 percent of the mass values

TABLE 3. CALIBRATION VALUES OF MASS MONITOR

Quartz Crystal	Deposition Length in cm	Aerodynamic Diameter in μm
01	2.0	-
02	5.7	7.5
03	8.5	4.480
04	11.3	2.913
05	14.1	2.102
06	17.0	1.656
07	21.0	1.250
08	27.0	0.922
09	35.0	0.691
10	48.0	0.505
11	66.0	0.359
12	89.0	0.213
13	117.0	0.125
14	141.0	0.065

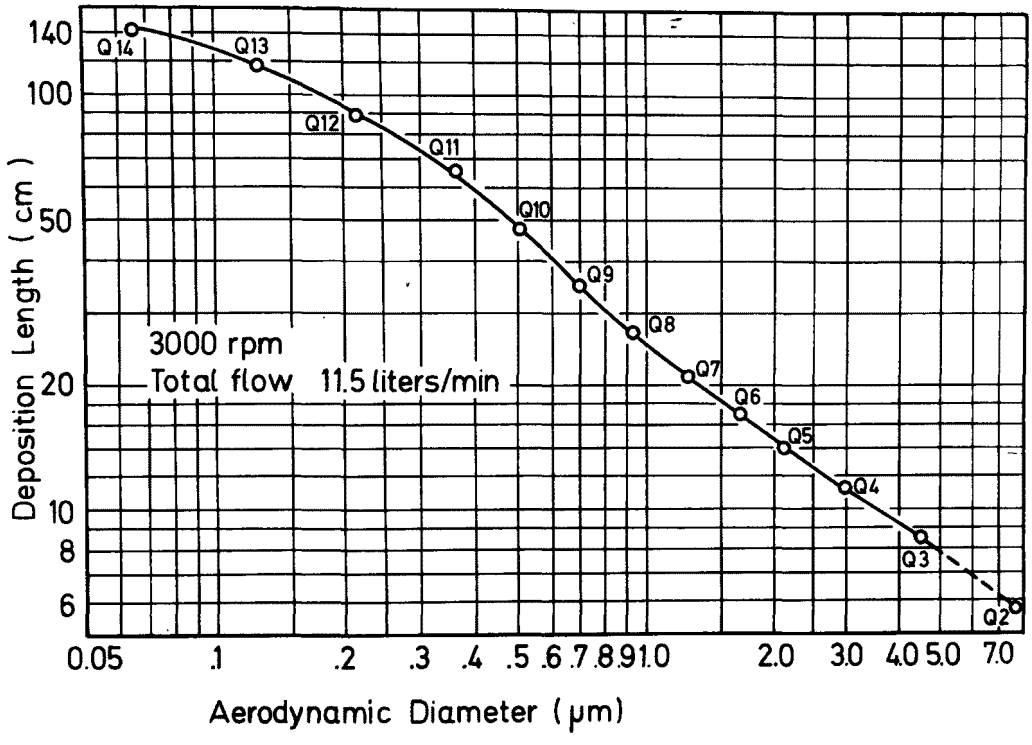

Figure 20 Centrifuge calibration curve

derived from the frequency shifts of the crystals according to Eq. 3.

7. INSTRUMENT OPERATION

1. The cleaned quartz crystals are inserted into the proper grooves of the rotor duct. Then laminator and aerosol inlet are inserted and the rotor is closed by the lid which contains the inlet system with the air inlet (blue), the outlet (red), and the aerosol entrance (Figure 5). Inlet and outlet are coupled to corresponding connectors on the lid of the driving unit.

2. Then, a connection to the clean air device (Figure 7) is made. The aerosol entrance of the centrifuge is connected to junction A, which allows the operation of the centrifuge with particle-free dry air. At the beginning of a measurement, the centrifuge should be run in this way for some time in order to get constant quartz frequencies.

3. The cooling system and the electronics of the driving unit is switched on by pressing button 1 (Figure 5). The switch may remain in this standby position if the room temperature is below the fixed value of 20 ^{o}C of the rotor. If this is not the case, the cooling machine should only be switched on when the lid of the driving unit is closed in order to avoid condensation of water at the walls of the rotor chamber.

4. The centrifuge motor can only be started if the lid of the driving unit is closed and the cooling machine is switched on. Rotor starting is done by rotating switch 3 (Figure 5) into position "infinite". In the other positions, the switch works like an alarm clock which switches off the centrifuge motor when the chosen time is over. Rotor speed is corrected by dial 4. Adjustment of this dial is only necessary in case of a new calibration of the rotor speed. The standard

speed is 3000 RPM which is read by the instrument shown on top of the right side of Figure 5. The Rotor speed is well stabilized and should be constant within 0.1 percent after a warm-up time of 30 minutes.

5. Control electronics, frequency counter, printer and inductive power supply are to be switched on.

6. Before the pump is turned on, the 4-way-valve (Figures 7 and 8) should be in standard position "2" which allows a circulation of the particle free and dry air through the spiral duct of the rotor. (Position "1" of the 4-way-valve is only used for flushing the rotor with carrier gas if a gas other than air is chosen.) Total flow (flowmeter I) should be near 11 $L \cdot min^{-1}$, aerosol flow (flowmeter II) near 1 $L \cdot min^{-1}$. After a warm-up period of 30 minutes the total flow is adjusted to the standard value of 11.5 $L \cdot min^{-1}$ by changing the pump supply voltage through a regulating transformer and dial D (Figure 7) while the needle valve E (flowmeter I) is fully opened. The aerosol flow is adjusted to the standard value of 1.0 $L \cdot min^{-1}$ by means of needle valve E of flowmeter II.

7. The quartz crystals can be chosen by means of the toggle switches at the control unit; measuring time and mode of printing by means of the thumb-wheel switches. For a precise frequency measurement the resolution must be switched to 0.1 Hz by pushing the left black and the adjacent white button of the frequency counter. The sensitivity of the counter must be switched to maximum by means of the sensitivity knob.

8. In order to get printed values, the microprocessor must be switched on. Program start is effected by pressing the MP start button. By means of the mode switch, different programs of the MP can be run as shown in table 2.

9. After the initial frequencies of the selected crystals have been printed and a repetition shows that they become constant, sampling can be started. This is done by disconnecting the aerosol entrance from junction A (Figure 7), and connecting the tube to the aerosol source. Sampling will start immediately with the sampling rate set at flow meter II. If the humidity of the aerosol is very high the drying tube (Figure 9) should be used.

10. In order to stop aerosol sampling, the aerosol entrance must be disconnected from the source and connected to junction A of the clean air device. Then, the centrifuge motor is switched off by a rotation of dial 3 (Figure 5) into position 0. If dial 5 is in 0-position the centrifuge is not braked, in the other positions braking takes place. Braking becomes stronger the more clockwise the dial is rotated.

11. In standby operation, the pump, the frequency counter and the inductive power supply should remain switched on in order to get low frequency drifts.

    The lid of the driving unit can only be opened when the centrifuge rotor has stopped. Then, the lid opens by pushing button 2 (Figure 5).

8. INSTRUMENT PERFORMANCE

Standard rotor speed of the centrifuge is 3000 RPM. A good resolution of deposited material is obtained for particles of aerodynamic diameters from 7.5 μm to 0.065 μm at a total flow of 11.5 $L \cdot min^{-1}$ and an aerosol flow of 1 $L \cdot min^{-1}$.

The sensitivity of the mass monitor is given by the gravimetric constant of the 10 MHz quartz crystals:

$$\frac{1}{C_f} = 4.4 \cdot 10^{-9}\ g \cdot cm^{-2} \cdot Hz^{-1}.$$

The weighing accuracy depends on the quality of the particle coupling to the crystal surface and on the frequency drifts caused by changes in temperature and humidity. With regard to the mass measurement of large solid particles there is a boundary above particle diameters of 5 μm. Particles of 7.5 μm in diameter are too large, to be well coupled to the surface of quartz Q2 although it is coated. However there are no problems with 7.5 μm droplets, even with an uncoated quartz Q2.

Since the coupling problem is solved by coating the crystals (for particle diameters of 1 μm to 5 μm) and the rotor temperature is kept constant near 20 ^{o}C, the only significant disturbance of the frequency is caused by changes in humidity. A humidity increase of 1 percent causes a frequency drop of about 1 Hz. Changes of humidity can be measured by means of quartz Q1 which is mounted at the beginning of the spiral duct where no particles can be deposited. In order to get good results, humidity should be kept as constant as possible. This is done by drying the carrier gas in the clean air device.

As the aerosol flow is nearly $\frac{1}{11}$ of the total flow, humidity changes of the aerosol flow cause total humidity changes of $\frac{1}{11}$ only. For aerosols of high humidity, a drying tube should be attached to the centrifuge aerosol inlet as shown in Figure 1.

Carrier gas drying is accomplished by two drying tubes, which may serve for an operating time of some 120 hours. But during this time, depending upon the humidity of the sampled aerosol the effective humidity is slowly increasing and a corresponding frequency drift is caused. Measurements showed that, after a warm-up time of 1 hour, these frequency drifts (of newly inserted and cleaned crystals) are <1 Hz per hour. But there are differences in the drift values of different crystals which could be caused by small differences in the mounting of the crystals to the 4 pins.

Assuming unwanted frequency drifts of 1 Hz per hour, frequency shifts caused by particle deposition should be at least 2 or 3

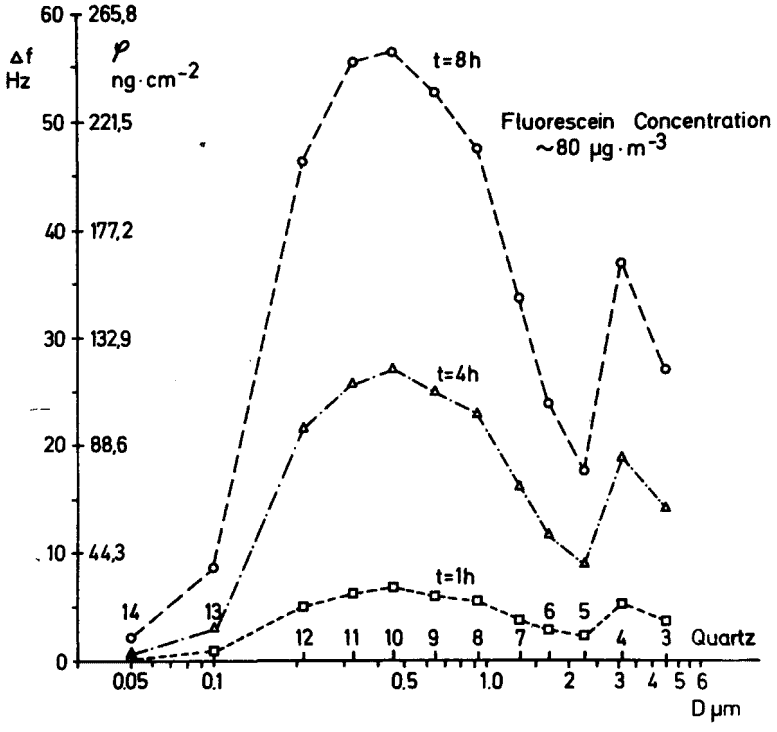

Figure 21 Deposited masses of fluorescein

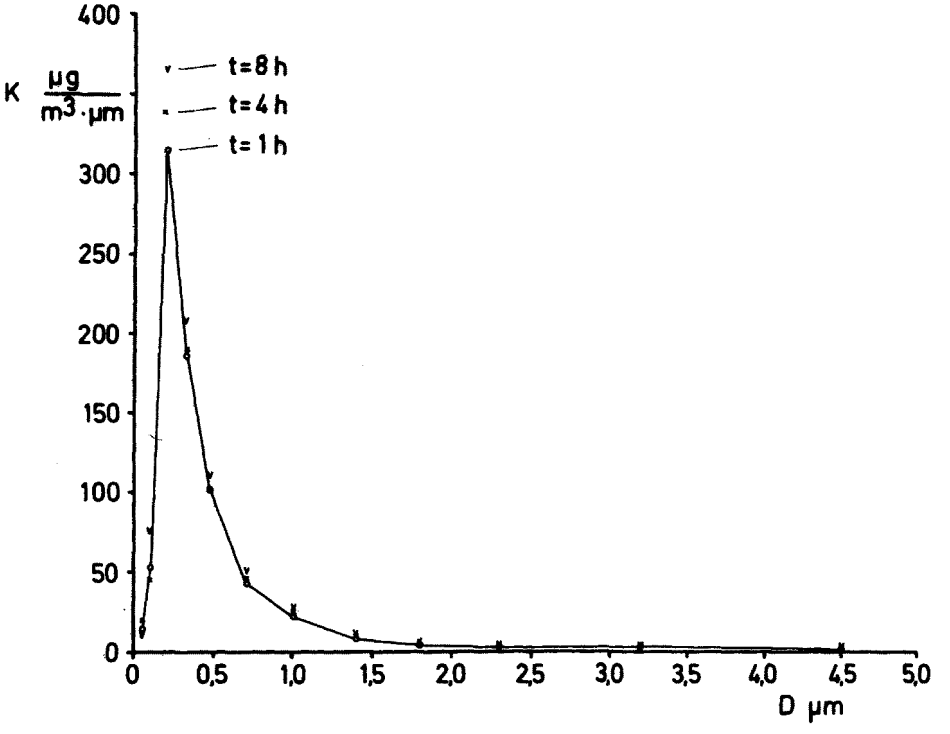

Figure 22 Spectrum of fluorescein aerosol

times higher. Thus, the measurement of an aerosol spectrum with a concentration near 50 $\mu g \cdot m^{-3}$ is possible.

The high sensitivity of the mass monitor is shown in the example of sampling a fluorescein test aerosol with a concentration of 80 $\mu g \cdot m^{-3}$ over a time period of 8 hours. During this time the centrifuge was operated under standard conditions. Figure 21 shows the result: frequency shift Δf, and corresponding surface concentration φ, calculated by the instrument microprocessor is plotted vs the particle diameter D. Already after one hour of operation a spectrum of surface concentrations can be seen, the shape of which does not change very much after sampling times of 4 and 8 hours.

(These values have been measured by means of the German version of the mass monitor, which has a smaller channel depth than the EPA version, causing small differencies in the calibration curve).

Figure 22 shows the airborne mass vs. particle size distribution of the same fluorescein test aerosol as calculated by Eq. 5 and the K_F values given in table 1. The second maximum at the Q4 quartz crystal near particle diameters of 3 µm is nearly lost in Figure 22 because of the relatively small K_F values for larger particles.

REFERENCES

1. Stöber, W. and H. Flachsbart, Environmental Science and Technology, 3 1280 (1969).

2. Stöber, W. and H. Flachsbart, Journal of Aerosol Science, 2 103 (1971).

3. Kops, J., Hermans, L., and Vate, J.F. van de, Journal of Aerosol Science, 6 329 (1975).

4. Stöber, W. in B.Y.H. Liu (Editor), Fine Particles, Academic Press 1976, p. 351.

5. Meinke, H. und F.W. Gundlach, Taschenbuch der Hochfrequenztechnik, Springer-Verlag 1962, p. 142, 1112, 1163.

6. Sauerbrey, G.Z., Zeitschrift für Physik, 155 206 (1959).

7. Chuan, R.L., Journal of Aerosol Science, 1 111 (1970).

8. Olin, J.G. and G.J. Sem, Atmospheric Environment, 5 653 (1971).

9. Carpenter, T.E. and D.L. Brenchley, American Industrial Hygiene Association Journal, 33 503 (1972).

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA-600/2-80-053	2.	3. RECIPIENT'S ACCESSION NO.
4. TITLE AND SUBTITLE DESIGN AND PERFORMANCE OF AN AEROSOL MASS DISTRIBUTION MONITOR		5. REPORT DATE March 1980
		6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) W. Stöber, F. J. Mönig, H. Flachsbart and N. Schwarzer		8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Fraunhofer Institute of Toxicology and Aerosol Research Federal Replublic of Germany		10. PROGRAM ELEMENT NO. 1NE833D EB010 (FY-79)
		11. CONTRACT/GRANT NO. R803592
12. SPONSORING AGENCY NAME AND ADDRESS Environmental Sciences Research Laboratory - RTP, NC Office of Research and Development U. S. Environmental Protection Agency Research Triangle Park, N.C. 27711		13. TYPE OF REPORT AND PERIOD COVERED Final 1977-1979
		14. SPONSORING AGENCY CODE EPA/600/09
15. SUPPLEMENTARY NOTES		

16. ABSTRACT

An aerosol mass monitor has been built to measure the masses of non-volatile aerosols in the range of 0.05 to 5 μm aerodynamic particle diameter. The instrument consists of a newly designed spiral duct aerosol centrifuge equipped with highly sensitive quartz sensors for in situ weighing of the deposited aerosol masses. The instrument further includes a clean air device for maintaining constant aerosol flow conditions, and electronic parts for the operation of the quartz sensors. The mass of aerosol deposited on the quartz crystals is automatically measured by an electronic counter while the rotor of the centrifuge continues spinning. The data are handled by a microprocessor.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Air pollution		13B
Aerosols		07D
Particle size		14B
Centrifuges		09A
Crystal detectors		08G
Quartz		14B
Microbalances		

18. DISTRIBUTION STATEMENT RELEASE TO PUBLIC	19. SECURITY CLASS *(This Report)* UNCLASSIFIED	21. NO. OF PAGES 52
	20. SECURITY CLASS *(This page)* UNCLASSIFIED	22. PRICE

EPA Form 2220-1 (9-73)