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PRACTICAL ASPECTS OF AEROSOL CHARACTERIZATION IN AN ENVIRONMENT OF CONTROLLED TEMPERATURE AND RELATIVE HUMIDITY

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ABSTRACT

In the fields of environmental and pharmaceutical aerosols it is of importance to create atmospheres of controlled temperature and relative humidity for the investigation of airborne particulate behavior. Close control of humidity, especially where relative humidity sensing is used, also requires close temperature control because of the dependence of relative humidity on temperature. Unique designs of apparatus have been employed, in many cases, without considering the implications of the level of control required to obtain statistically acceptable data. This paper describes a novel apparatus, contrasts it with those in the literature and states specifically the level of control which can be achieved practically and how this compares with theoretical expectations.

INTRODUCTION

A controlled environment apparatus should only be designed after thorough consideration of the requirements. Failure to recognize all the basic elements of the problem may often lead to unreasonable expectations in respect to some factors, while ignoring others which may equally influence the final performance. Precision in control requires careful attention to the response characteristics of the conditioning equipment, the apparatus itself, as well as to those of the control devices. Though the performance is influenced by each component it is the interaction of one with the other that determines the ultimate efficiency of control (1).

Interest in condensation growth has arisen largely as a result of environmental aerosols (2-5). Condensation growth of particulate and gaseous aerosols is responsible for meteorological cloud formation. Aerosol materials may act as nuclei at relative humidities approaching saturation vapour pressure in the atmosphere and similarly in the high relative humidity environment of the respiratory tract. The atmosphere in the lung approaches saturation since it is in equilibrium with the tissue fluids at 37°C. The relative humidity may be calculated by freezing point depression of blood (6) to be 99.5%.

The relative humidity, RH, at equilibrium may be expressed as a fraction of the actual vapour pressure, P divided by the saturation vapour pressure, P_o, which is the same as the water activity, a_w:

$$RH = P/P_o = a_w \qquad \ldots\ldots\ldots\ldots\ldots\ldots \quad 1$$

The concentration in a droplet which has equilibrated with the environment is not the same as the iso-osmotic solute concentration at which the test solution and isotonic saline have the same a_w at 37°C. The reason for this is that the droplet vapour pressure, P', is greater than P in the previous equation due to the Kelvin effect, which raises the vapour pressure due to the curvature of the surface. Thus:

$$\ln P' = \ln P + 4 M_w \gamma / KT \rho_1 D_1 N_a \qquad \ldots\ldots\ldots \quad 2$$

Where M_w is the molecular weight of water, γ is the surface tension of the solution, K is the Boltzman constant, T is the absolute temperature, ρ_1 is the density of the liquid, D_1 is the equilibrium droplet diameter and N_a is Avogadros number.

METHODS

Aerosol Particle Size Measurement

Cascade impaction was performed using a Delron DCI-6 device (Delron, Powell, OH) according to the method of Groom and Gonda (7) and data fitted by a computer analytical technique (8).

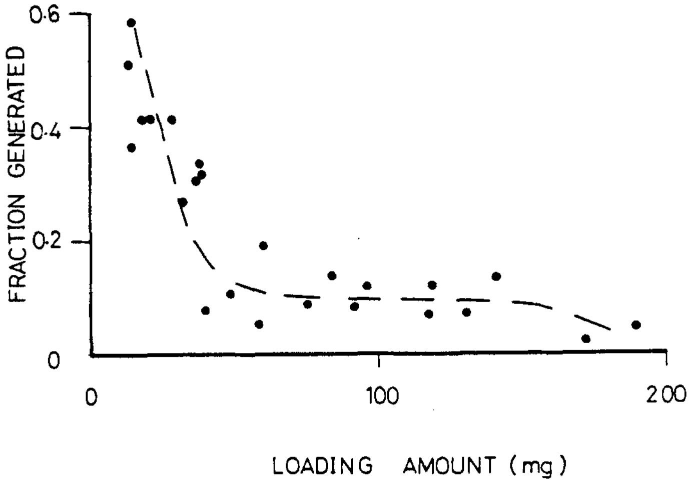
Spectrofluorimetry

Disodium fluorescein (AR Grade, Koch Light, Colnbrook, U.K.) was generated as the experimental aerosol. The samples, once collected, were measured at an emission wavelength of 516 nm and an excitation wavelength of 486 nm.

Aerosol Generator

A Nebulet^R vertical spinning disc aerosol generator (9) was employed to generate the dry powder aerosols. The device was operated without a throughput of air generating mechanically by the action of the spinning disc.

Figure 1 shows a graph of the ratio of the fractional output of the aerosol generator (Total output/loaded amount) plotted against the amount loaded. From this figure it may be assumed that at a loading value less than 10 mg the amount generated will fall to zero, at zero loading. It has been shown that the output of this device using 100 µg samples and a throughput of air was very erratic (10). The amount generated (multiplying the loading value by the fraction of the load generated, in Figure 1) at



Figurel
Ratio of the output of the vertical spinning disc nebulizer plotted against the loading amount (mg).

between 10 and 150 mg, indicated by the dotted line, was equivalent to 8.5 ± 1.5 mg every 2 minutes. The fall in the fraction generated at loads from 150 mg to approximately 200 mg was a consequence of overloading the device. The effect of overloading was that the powder bed absorbed the energy imparted by the disc both through direct contact and particle-particle interaction.

There are many powder generating devices available such as spinning disc/top (11) and fluidized bed (12) aerosol generators. In these experiments the disadvantages of these devices are twofold. Firstly in order that constant output may be achieved these apparatus' need to be in operation for a considerable time, which requires a large reservoir of the aerosol powder. Secondly the generation of the aerosol relies upon an air supply. This becomes a complication when the control of the temperature and relative humidity is being considered. Air which is used to generate the aerosol may not be humidified because of the effects on powder flow (13), although it may be pretreated to control temperature. Assuming that the control of temperature is accurate the effects of this incoming air on the controlled environment may be calculated. The practical complications of this technique together with the disadvantage of the amount of aerosol powder required to achieve constant output with these devices makes them inappropriate for use in these studies. The aerosol generator employed was appropriate both in terms of the amount of aerosol generated and the operating conditions under which it was considered practical to control the environment into which the powder was generated. Finally this device was easily manipulated and adapted for use in these studies.

Controlled Temperature and Relative Humidity Apparatus

Figure 2 shows the two water systems employed in these experiments. The first, uppermost, water bath was used to heat and humidify the air and water to 37°C and was pumped from this bath to the jacketed aerosol inlet port. Water from this bath was also pumped to the jacketed equilibrium chamber.

The second water bath supplied water for the cascade impactor jacket situated beneath the equilibrium chamber, also maintained at 37°C.

Both of these systems were maintained under controlled conditions by means of proportional temperature controllers (YSI, Yellow Springs, OH).

Figure 3 shows the method by which air was preheated by passing it through a jacket around the aerosol inlet port. It was then pumped through an air flow meter and into the preheating and humidifying system. The air was preheated by passing it through a glass coil submerged in the uppermost water bath and into bubbling flasks containing salt solution. Finally the air entered the equilibrium chamber. A fraction of this air was

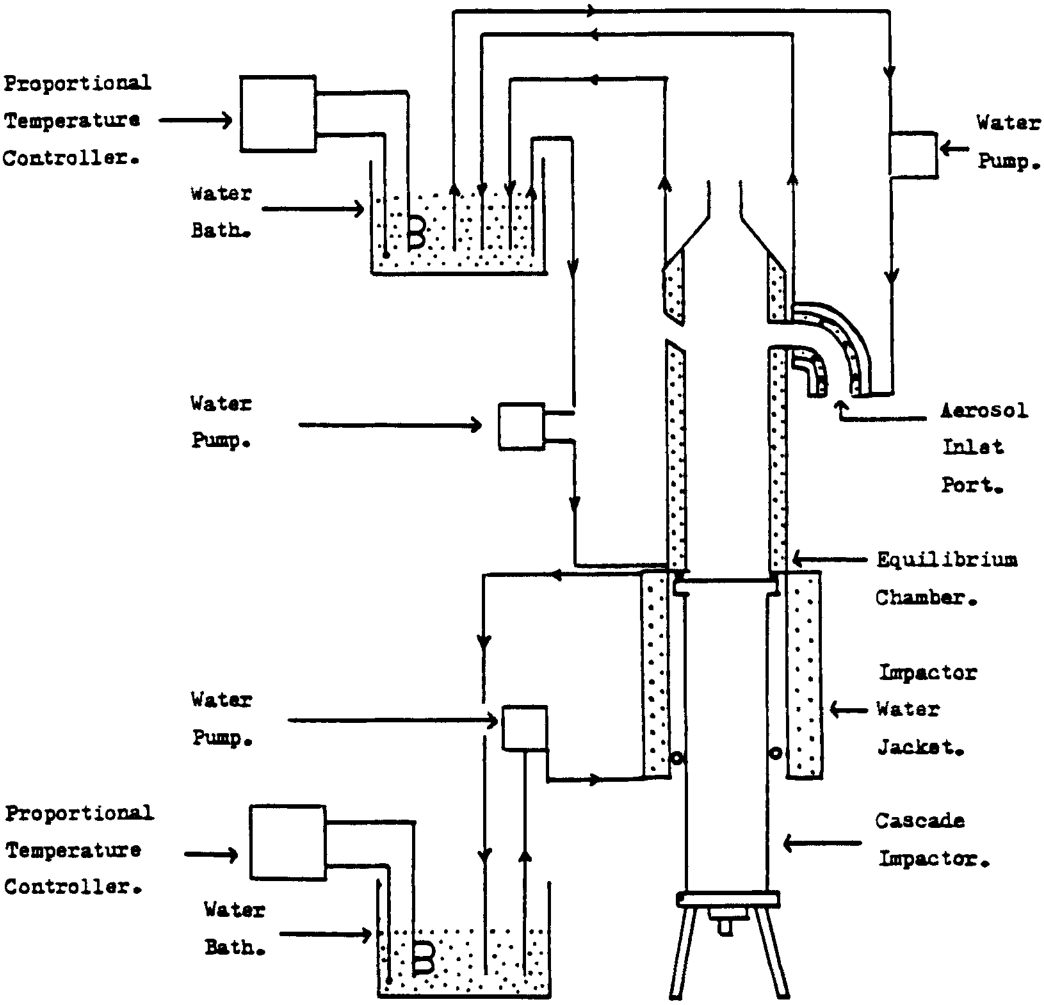
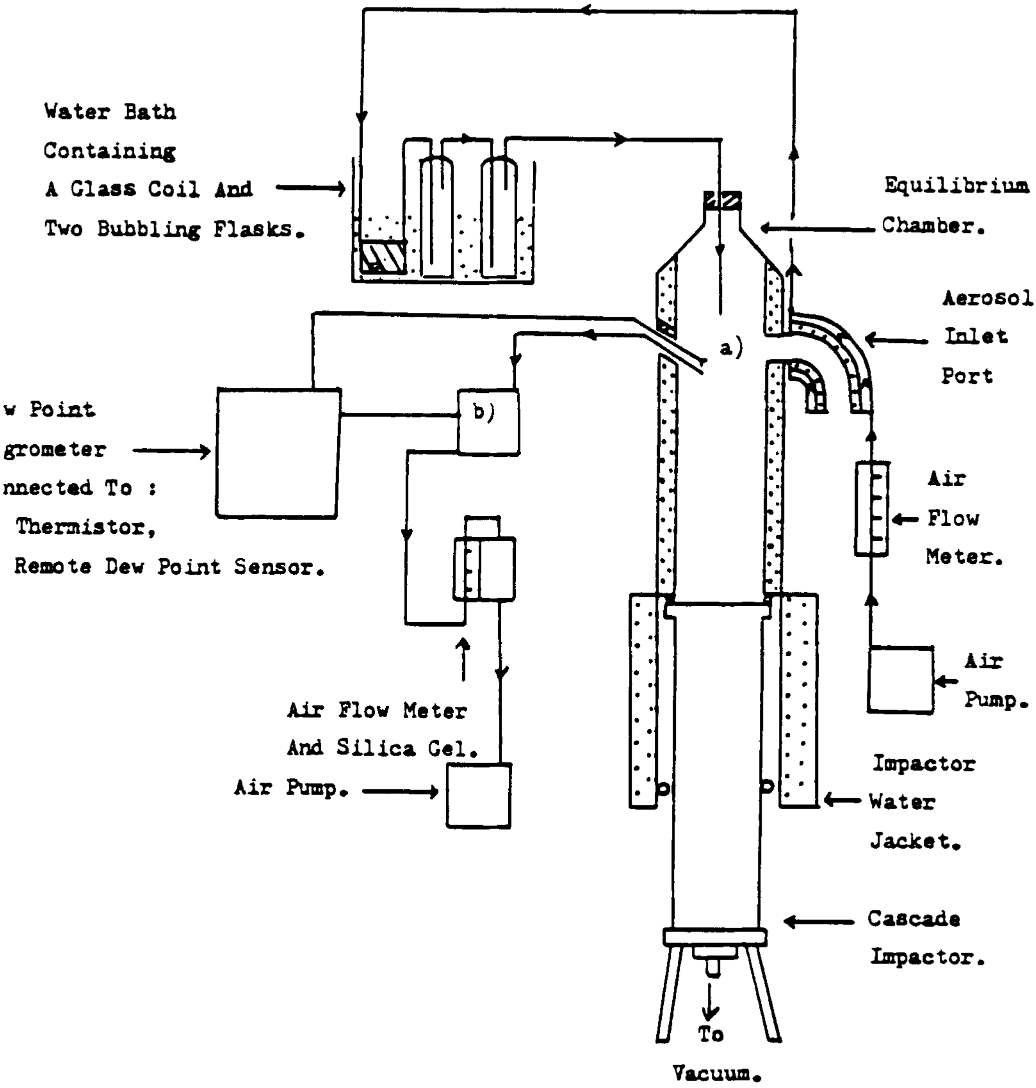


Figure2
The water system of the controlled temperature and relative humidity apparatus.

removed for analysis by a remote sensor which measured the Dew Point of the air (EG+G Instruments, Waltham, MA). By placing a thermistor in the equilibrium chamber, and connecting this to the remote Dew Point sensor, a digital visual display of the temperature and relative humidity was obtained.



<u>Figure3</u>
The air system of the controlled temperature and relative humidity apparatus.

The apparatus was encased in a modular fashion in an expanded polystyrene housing 1/4" in thickness. Thus, disturbing one section of the apparatus did not influence the equilibrium in the whole system.

Operating Procedure

The operation of the controlled temperature and relative humidity apparatus combined the control of the temperature of a water circulating system with the temperature and relative humidity control of the air supply.

Air was pumped into this system at 14.5 L/min. Two litres per minute were drawn from the equilibrium chamber for analysis. The remaining 12.5 L/min were drawn through a cascade impactor. By placing a water manometer at the aerosol inlet port the inflow and outflow were finely balanced. The 7.85L equilibrium chamber was constructed of pyrex and water jacketed. At the air flow above the residence time was approximately 40 seconds.

The salt solution employed throughout these studies to provide an atmosphere of high relative humidity was potassium dichromate. This theoretically gave a saturated vapour of 98% RH (14) at 37°C.

The aerosol generator port was situated at right angles to the equilibrium chamber of the controlled temperature and relative humidity apparatus. The inlet port was designed to reduce the distance travelled by the aerosol. This was achieved by shaping the tube to the inlet to form the tangential tube from the aerosol generator. This tube was also water jacketed.

The aerosol generation and collection was carried out once the system had equilibrated. From a cold start equilibrium was established within 3 to 5 hours by monitoring the temperature and relative humidity in the chamber.

Prior to the generation of the aerosol and following the equilibration of the impactor, the vacuum pump was set in operation. This allowed final equilibration of the impactor and eliminated the likelihood of surface condensation. Immediately upon the operation of the pump a rubber seal between the impactor and its water jacket was positioned. Up to this point the air was vented through the gap between the impactor and its housing. From this point the impactor approached dynamic equilibrium.

The generation of the micronized disodium fluorescein powder aerosol was then carried out for a 2 minute period after which the airflow through the impactor was continued for a further 40 seconds. The impactor was removed ensuring that the vacuum pump was left on until the instrument had been totally removed from the humidified air source. Failure to do this would leave the humidified air motionless within the rapidly cooling impactor thus producing condensation. After dismantling the impactor, removing the slides and the filter, it was replaced in the apparatus with clean slides and filter in preparation for the next experiment. A reduced period of equilibration, for the entire apparatus, of between 30 and 60 minutes was all that was required at this stage. It was important that while the impactor was out of the apparatus a blanking pad was placed over the opening to the impactor water jacket, not completely blocking the orifice as the air was still being pumped through the system. This precaution reduced the likelihood of the incursion of cooling, destabilizing air from the atmosphere into the equilibrium chamber.

RESULTS AND DISCUSSION

Accuracy of Control of Temperature and Relative Humidity

Theoretical: It has been demonstrated that little or no growth occurs for disodium fluorescein (DF) at 37°C up to 50% RH (8). At high RH the control of this factor and temperature is very important in the study of hygroscopic growth. DF, for example, exhibits significant differences in hygroscopic growth, and thus in particle size, over small intervals of RH at high RH. In these studies the system was maintained at 37 ± 0.1 °C. The RH was controlled at 96.6 ± 1.2 and at 20 ± 5 %, which constitutes high and ambient conditions respectively.

Assuming negligible Kelvin Effect and that the equilibrium droplet was below the solubility limit of DF then from equation 1:

$$R = P'/P = 1 \tag{3}$$

The equation for the increase in the equivalent volume diameter then becomes:

$$R' = (P_s/P_1\ (1+M_w/M_s)\ (H(i+j)-j)/(1-H)))^{1/3} \tag{4}$$

Where P_s and P_1 are the density of the solid particle and droplet respectively, M_s is the molecular weight of fluorescein, i is the number of ions into which a dissolved molecule is dissociated in water and j is the number of crystal molecules per salt molecules. From this equation (8) it may be derived that at high RH the error in R depends on errors in H as:

$$dR'/R = 1/3(dH/(1-H)) \tag{5}$$

This will also apply to the aerodynamic growth ratio since both the numerator and the denominator of equation 5 are multiplied by the same factor.

The a_w of dilute electrolyte solutions are relatively insensitive to temperature changes (15). Similarly the partial vapour pressure P in equation 3 is not subject to large errors when temperature fluctuates, a temperature of 1°C causes only a 0.2% change in P at 37°C. However, the saturation vapour pressure of pure water rises by 5% per degree C between 36.8 and 37.2°C (7).

The previous equation can be written:

$$dR'/R = 0.017\ (dT/((1/H) - 1)) \tag{6}$$

TABLE 1

Particle Size (μm) and Distribution Data for Micronized Disodium Fluorescein Aerosol Powder Generated at 20% and 97% Relative Humidity and 37°C.

Relative Humidity	Replicate *MMAD(μm)	Mean MMAD(SD)	Replicate ** σ_g	Mean σ_g(SD)
20	3.6, 4.1, 4.0, 3.7	3.8 (0.24)	1.4, 1.5 1.7, 1.6	1.5 (0.13)
97	5.2, 5.2 5.2, 6.4	5.5 (0.6)	1.3, 1.3 1.5, 1.5	1.4 (0.12)

* Mass Median Aerodynamic Diameter

** Geometric Standard Deviation

The limit of the recorded sensitivity of the thermistor used to measure the temperature in these studies was 0.1°C. The extremes of temperature giving a recorded range of 37 ± 0.1°C were therefore 37 ± 0.15°C. From this observation and the mean and standard deviation of the relative humidity, the limit of accuracy in the measurement of the hygroscopic growth ratio may be calculated. This limit is based on the combination of temperature and RH which gives the greatest error. Thus inserting values for H of 0.978 ((96.6 + 1.2)/100) and for dT of 0.15 gives a value for dR'/R of 11.4%.

Experimental: The data shown in Table 1 is an example of the behavior of a sample micronized disodium fluorescein powder generated four times at both high and low relative humidity. An increase in particle size was observed at high RH in comparison with that at low RH. The mean growth ratio and deviation from has been calculated by dividing each individual replicate at high RH by those at low RH. For the data shown above these values were 1.43 ± 0.16. Thus the error in the growth ratio is 11.18% ((0.16/1.43) x 100). The data from a total of 12 of the studies (96 Aerosol generation experiments) have been pooled to calculate a mean error in growth ratio of 7.27 ± 2.68% with the highest single error of 11.32%. The particle size distributions were log-normal, as indicated in Table 1 by geometric standard deviation estimates and exhibited little or no change as a function of RH in any of these studies.

All of the observed mass median aerodynamic diameter data obtained in these experiments fell within the limit of theoretical error which can be attributed to the growth ratio as a function of RH and temperature.

Comparison of this Apparatus with Those in the Literature

Several in vitro controlled temperature and relative humidity devices have been described in the literature. Hiller (16,17), Halbert (18), and Bell (19) employed climatic cabinets which were heated directly and humidified either using salt solutions or by passing known volumes of air, at a constant temperature, through water. Tang (20) used a device similar to that in the present study. His apparatus consisted of a 850 ml pyrex jacketed growth chamber in which the residence time for aerosol particles was 20 seconds. A boiler condenser unit was used to humidify the air, with cooling water thermostat regulated at 25°C in this particular account. The efficiency of this piece of equipment cannot be contrasted with the one used in these experiments as the temperatures at which each was undertaken were significantly different.

Bell (19) humidified the air and passed it through a water bath at constant temperature and finally into a heated cabinet. The system was maintained at 37°C and 98% RH. Assumptions concerning the RH in different parts of the apparatus were made in order to estimate the experimental conditions and it must be concluded that no error margins could be given.

Hiller (16,17) used a 200L insulated plexiglass chamber with heat strips and a digital thermometer to control environmental conditions. A variable speed fan inside the chamber provided thermal and aerosol mixing. The chamber was humidified by bubbling air through a heated water bath at a flow rate necessary to obtain the desired humidity. For studies at ambient humidity, the temperature was maintained at 36.8 ± 0.4°C and the relative humidity at 23.6 ± 1.4%. For studies at airway humidity, the temperature was 37.1 ± 0.4°C and the relative humidity at 96.4 ± 0.2%. Otanyi and Wang (21) employed a system in which the RH was 99.5% at 36.5°C. The errors in these values were not noted.

CONCLUSIONS

In order to overcome problems in preliminary design (22) the apparatus described in this communication was arranged such that the two heat sources did not interact. The volume to be maintained at constant temperature was reduced by modular insulation thus minimizing the influence of radiation of heat. All electrical devices were situated outside the insulation and with maximum exposure to the atmosphere for the dissipation of heat generated by their action. The use of an aerosol generator which requires no throughput of air overcame the problem of preheating and humidifying the air used to generate aerosols which might arise with other devices. The added variable of introducing unequilibrated air into the system has thus been avoided.

The control of temperature using this device was 37 ± 0.1°C and at high RH, RH control was 96.6 ± 1.2%. This compares favorably with Hiller's values.

In conclusion the device has been used successfully to investigate hygroscopic growth of aerosols showing that measurements were within the theoretical limits of control of the apparatus.

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