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Introduction

The following report entitled **Carbon Aerosol Morphology** is the result of research done through the University of Missouri - Rolla Graduate School program entitled Opportunities for Undergraduate Research Experience (OURE). My appointment was for the Summer - 1990 with the Cloud and Aerosol Sciences Laboratory, Department of Physics, located in Norwood Hall. The goal of this appointment was to study the size and morphology of combustion aerosols whose characteristics are dependent on the history of the particle, e.g. exposure to chemicals, scavenging effects, and other time dependent factors. Computer image processing software is available in the department for this purpose.

This research experience included several facets with the main portion of time spent on the development and perfection of a combustion chamber, specifically the burner assembly itself. This combustion chamber is used in the Cloud Physics laboratory to supply combustion aerosols of liquid fuels, primarily aviation fuel JP-4 supplied by McDonnell Douglas Corporation. These aerosols are used in the study of the effects of jet engine emissions in the atmosphere and thus upon global environment.

The appointment began with exposure to the operation of the equipment and instruments used within the Cloud Physics laboratory. This equipment being used to isolate and characterize aerosols by size, mobility, and critical supersaturation (SSc), the latter being a measurement of the amount of water vapor present in air to cause the particle to grow by the accumulation of water vapor. The understanding of these methods is important as they are used routinely to classify and characterize aerosols produced by nebulizers and by combustion processes.

As my project goal was to study aerosols produced by combustion processes, it was first necessary to perfect the operation of the combustion equipment which operated inadequately for the collection of these aerosols. The torch assembly used for combustion produced a flame which was unsteady in nature and not easily controlled. This problem had to be eliminated as the oscillations of the flame produced particles of inconsistent nature as seen by production of varying amounts of soot. The torch was analyzed and its problems were found. Based on a great deal of research time, the burner assembly was modified to produce a flame which was very stable and easily controlled. This required at least a month of the summer to accomplish.

Upon the perfection of the torch, it was then possible to produce a consistent flame pattern from which carbon aerosols were collected and analyzed by scanning electron microscopy. Due to the amount of time required to perfect the torch assembly it was not possible during the summer to complete the goal of the project. As such, time was taken during the Spring 1991 semester to continue to collect and analyze particles. It is from this work that significant data and conclusions have been possible. This information is in the following report.

In addition to the improvements of the combustion process, time was used to improve the operation of a wire-egression droplet generator which is currently being used by another department for the production of single droplets. Also, a computer software system (JAVA) for image processing was studied for its application to the characterization of the carbon aerosols collected.

Additional plans related to this research appointment include two presentations at regional science meetings. The first of these is the Missouri Academy of Sciences Meeting (MAS) on April 19, 1991 in Fulton, Missouri for a presentation entitled **Combustion Aerosol Morphology.** The second is at the Midwest Association for Cloud and Aerosol Physics (MACAP) at the University of Wisconsin, Madison, on May 17, 1991 for a presentation entitled **A laboratory liquid fuel burner and the properties of the associated combustion aerosols.**

Pollution

One of the greatest concerns facing the world today is the need to regulate and reduce environmental pollutants. Of particular interest are anthropogenic, that is man-made, air pollutants which arise from combustion processes as in power plants and factories. These atmospheric pollutants subsequently affect global surface conditions as they are removed from the atmosphere by natural precipitation processes like rainfall. Air pollutants pose immediate health hazards to man such as lung disease due to particulate matter. These pollutants also cause long-term global effects associated with chemical reactions and the Greenhouse Effect. As these effects are critical to the environment and its stability the need for pollution control becomes increasingly important.

Pollutants fall into five major categories which are particulate matter, nitrogen oxides, sulfur oxides, hydrocarbons and carbon monoxide. Each of these categories is associated with a damaging process to the environment such as acid rain production. This research is associated with the production and effects of particulate matter to the atmosphere, particularly that which is produced by organic fuel combustion.

Airborne particulate matter can arise from different sources. These sources can be of mineral origin such as milling and demolition operations or as organic origin such as forest fires and fossil fuel combustion. Organic particulate matter includes soot and flyash which may be as large as 100 microns or larger in diameter and combustion nuclei which may be as small as 0.01 micron in diameter. Figure 1 provides information relating particle sizes with associated features for reference.

Aerosols

An aerosol is a suspension of solid or liquid particles dispersed in a gas medium which is air in this case. The

combustion of fossil fuels produces carbon aerosols whose sizes range from those of nuclei to those of soot. This study is concerned with the particles produced by aviation fuels which have a size range from 0.01 micron to 0.10 micron in diameter. This aerosol size range is important to study as they act as initiation sites for the phase change of water vapor within the atmosphere. Particles which act as sites for the accumulation of liquid water are known as cloud condensation nuclei (CCN). If the particles acts as sites for ice crystal deposition they are called ice nuclei (IN). Scavenging theory, that is the rate at which particles are removed from the atmosphere, was first studied by Greenfield (1957) who studied scavenging processes by Brownian diffusion, turbulent shear diffusion and inertial capture. He found that small particles (less than 0.1 micron radius) and larger particles (greater than 1.0 micron radius) are affected by Brownian diffusion and inertial capture resulting in greater scavenging rates for those particles. This produces a minimum scavenging rate for particles between those minimum and maximum radii which is referred to as the Greenfield Gap. This relationship between scavenging rate and particle size was also demonstrated by Slinn and Hales (1971) with a minimum scavenging rate of particles with a radius of 0.01-0.15 micron (0.02-0.30 micron diameter). This is pertinent to the particles which we are studying whose sizes fall within this minimum scavenging range suggesting that these particles will be long-lived in the atmosphere.

Cloud condensation nuclei occur by natural phenomenon such as salt particles generated by bursting bubbles over oceans. Other sources for such nuclei occur by man-influenced chemical reactions within the atmosphere such as ammonium sulfate formation or particles produced by combustion processes. As already mentioned these nuclei act as sites where water vapor can change phase to a liquid or solid depending on temperature. As such, their presence reduces the amount of supersaturation, that is the amount of water vapor present, required for a phase change to occur. Much higher supersaturation levels are necessary in the absence of nuclei to initiate the phase change which will occur at lower supersaturation levels in the presence of nuclei. Once the phase change is initiated, the water droplet continues to grow by vapor deposition until its size and interaction with other particles plays a greater role in its growth than does vapor deposition. The particle as a droplet may begin to fall and impact with other droplets at that point. Thus, the presence of carbon aerosols results in their action as condensation nuclei which influence the behavior of water vapor within clouds. The combustion aerosols produced by jet engines are studied for their actions and influences as condensation nuclei.

Production of Aerosols

To study the size and morphology of the carbon aerosols produced from jet fuel, a laboratory combustion and aerosol collection

system is used. This system consists of a combustion chamber in which liquid fuel and air are mixed for rapid combustion and a commercial instrument which removes the aerosols produced. Other Cloud and Aerosol Laboratory equipment is available for further characterization of the aerosol produced. Description of these systems follow and are represented visually by the attached Figure 2.

In an effort to study the production of combustion particles by a jet engine a chamber, made of a cylindrical stainless steel bell jar (44 cm diameter x 61 cm height) equipped with sealed viewing ports is used to house a torch-like liquid fuel burner. Within this sealed chamber is fixed a burner assembly in which pressurized liquid fuel and filtered air enter, preheat and premix before combustion occurs. This burner allows the control of temperature, fuel flow and air flow to any desired parameter. It is entirely accessible from outside the chamber to ensure a controlled combustion environment within the chamber. The burner head itself is constructed of stainless steel with interchangeable cores by which the flame diameter may be altered. The head also uses an internal jet passage which ensures the atomization and mixing of the fuel and air. Combustion occurs with a flame stabilized at the head by a fine stainless steel mesh. Figure 3 illustrates a lean-burn flame produced by the combustion of JP-4 aviation fuel in the combustion chamber.

The ability to control the flame parameters is critical to this study as it predicts the type of particle produced. Flame stability is necessary for the stability of the resulting combustion aerosol. Aerosol stability, in turn, is needed to accommodate the various aerosol characterization facilities available in the Cloud Physics aerosol laboratory. The actual combustion process is very complex in which the chemical reactions take place. This process amounts to reaction zones in which the fuel and oxidizer are heated to reaction temperatures. Reaction zones occur in which rapid oxidation of the jet fuel produces heat, flame, water vapor, carbon dioxide, aerosols, and other by-products. Control and stability of the flame is essential to study a particular flame type without which combustion aerosols ranging from nuclei to soot are produced. This combustion system is perfected to allow continuous laminar flow flame conditions ranging from a high temperature, high velocity, blue flame to a much lower temperature, low velocity, yellow flame. These differences are due to the amounts of fuel and oxygen present during the combustion process. These different flame conditions are found to produce measurably different combustion aerosols.

Another parameter of this system is that it can be used to produce the major flame types, either premixed or diffusion type. The premixed flame is the one usually employed in which the reactants are homogeneous before the point of combustion. With modification of the burner head a diffusion type flame can be obtained in which the fuel and oxidizer meet and react within a reaction zone limited by the diffusion of fuel on one side to the oxidizer on the other. Flame types of mixed nature between

premixed and diffusion can also be obtained if necessary. The versatility of the combustion chamber also extends to the ability to burn different liquid fuels for the study of other combustion products as well.

Collection of Aerosols

The flow of combustion products exits the top of the combustion chamber and enters an ice-cooled condensation container. This container serves to cool the gases present and to remove water vapor which is generated by the combustion process. The "dried" products are then prepared for entry into a commercial electrostatic aerosol sampler. This preparation is done by passing the combustion products through a commercially available bipolar charger. This unit uses a radioactive source to produce a known charge distribution on the particles as they exit the unit. This is needed as many combustion products carry large electrostatic charges due to the combustion process. This unit equalizes and quantifies the particle charge distribution. From the bipolar charger, the combustion products enter the electrostatic aerosol sampler which ionizes the aerosol present and intermittently precipitates the aerosol onto treated slides by use of an electric field force. Upon completion of a suitable number of cycles the treated slides are submitted for scanning electron microscopy (SEM).

The slides used for electrostatic precipitation of the particles are glass cover slips which are cleaned and coated with approximately 300 Angstroms of gold-palladium (Au-Pd) alloy. This Au-Pd serves as a viewing substrate for the SEM analysis. After collection of the aerosol, the slide is again coated with Au-Pd, about 200 Angstroms thick, which stabilizes the aerosol and increases the contrast for SEM analysis.

Analyzing Aerosols

SEM analysis is done by the University of Missouri - Rolla electron microscope laboratory located in McNutt Hall. The slides are examined and pictures of the slides are taken at magnifications ranging from 3500X to 75000X. Pictures are taken at locations chosen so that there is no bias towards the particles/area selected for analysis. Series of photographs are taken for particle analysis at higher magnifications. Figures 4-7 are examples of the electron micrographs analyzed.

In addition to analyzing areas in which particles are deposited, areas on the slides are covered as control regions. These areas are photographed in a duplicate manner as the actual test regions for analysis. A system control is performed as well in which the collection process is performed in its entirety except that combustion is not performed. This allows for a check of the entire system and the particles which may arise inherent to the system and not due to combustion. These control slides

are analyzed by the same procedure as the actual test slides with masked control areas and Au-Pd coatings.

A particle size distribution is desired from the electron micrographs to see the correlation between SEM analysis and size distributions readily obtainable from other aerosol characterizing methods already used and accepted. This correlation will provide information on the ability to use SEM analysis for the further characterization of carbon aerosols and their study. The method of determining this size distribution is to measure and record the diameters of the particles present on slides photographed by SEM and prepared using a lean burning laminar flow premixed combustion flame from JP-4 aviation fuel. This data follows; however it is important to note the criteria used for determining and measuring the particles.

A characteristic of the photographs is that an irregular background appearance occurs on almost all of the electron micrographs including those which are controls. See Figure 7. After consideration of what this could be - whether an actual particle collected or an anomaly associated with the Au-Pd substrate - it was decided to consider it something besides a particle layer. This decision arises from the fact that it occurs on control slides and the size distribution and appearance is not like that of a particle deposited to the surface. This will be discussed later with the following criteria used for determining which features constitute a particle as seen on Figures 5,6:

(1) the image appears distinct and defined at its edges,

(2) the image does not appear to be the size and shape of the background associated with the control pictures,

(3) the diameter of the particle is take as the outermost defined border of the particle,

(4) particles found on the edge of the micrograph are included provided the diameter can be determined.

The consideration and measuring of the particles is somewhat subjective to the person performing the task, however with these criteria it is believed that suitable guidelines are set for determining an accurate size distribution.

Data

The following data is taken from a single collection date in which carbon aerosols are collected from JP-4 aviation fuel. The fuel was burned with a lean (excess oxidizer), laminar flow premixed flame. Two slides were analyzed and the particles were counted from the micrographs obtained by SEM. This amounted to a total of 24 pictures for a total of 327 particles. The data is recorded as follows. Note all dimensions are in microns.

Photo Number	No. Part. Counted	Mean Diameter	Standard Deviation	Minimum Diameter	Maximum Diameter
SLIDE 1					
030904	10	0.072	0.014	0.056	0.094
030906	09	0.074	0.021	0.056	0.12
030908	19	0.065	0.019	0.036	0.11
030909	19	0.064	0.014	0.039	0.086
030910	16	0.070	0.021	0.042	0.12
030911	15	0.068	0.013	0.053	0.10
030912	10	0.068	0.023	0.044	0.12
030913	15	0.076	0.023	0.047	0.13
030915	12	0.064	0.014	0.044	0.094
030917	07	0.067	0.008	0.058	0.081
030918	18	0.065	0.012	0.047	0.099
030919	11	0.073	0.014	0.056	0.10
SLIDE 2					
030924	14	0.074	0.011	0.053	0.089
030926	12	0.080	0.018	0.050	0.11
030928	15	0.071	0.009	0.056	0.089
030929	20	0.075	0.016	0.056	0.11
030930	17	0.065	0.014	0.044	0.094
030931	10	0.068	0.021	0.036	0.10
030932	14	0.068	0.010	0.047	0.086
030933	10	0.068	0.021	0.042	0.12
030935	10	0.078	0.017	0.047	0.10
030937	09	0.093	0.012	0.072	0.11
030938	25	0.073	0.021	0.039	0.12
030939	10	0.075	0.013	0.047	0.089

The following is a summary of the particles counted from each slide and an overall summary for all particles counted. Again, all dimensional values are given as microns.

Summary of:	Number of Particles	Mean Diameter	Standard Deviation	Minimum Diameter	Maximum Diameter
Slide 1	161	0.068	0.017	0.036	0.13
Slide 2	166	0.073	0.017	0.036	0.12
Combined	327	0.071	0.017	0.036	0.13

Discussion

The summary of results gives an overall mean particle diameter of 0.071 microns with a standard deviation of 0.017 microns. This provides a particle range of 0.054-0.088 microns. This distribution is compared with the size distribution as measured by an alternate technique used in the Cloud Physics laboratory. This technique is based on particle electric mobility and uses the electric aerosol classifier (EAC) for sizing and the alternating gradient cloud nucleus counter (ALGR) for particle detection and counting. Data from the EAC/ALGR was computer processed to yield a size distribution with a mean particle diameter of 0.054 microns and standard deviation of 0.029 microns. This indicates a particle range from 0.025-0.083 microns. Good agreement is achieved between the size distributions determined by the two different methods.

Particle morphology (shape) is studied in addition to particle size. This is important as morphology affects particle hydration properties, mobility and scavenging rates which will affect the evolution of the particle in the atmosphere. Electron micrographs indicate that flame type is critical to particle morphology as shown on the attached figures. A yellow flame produces soot, hence large particle agglomerates, and a blue flame produces small spherical particles with little agglomeration. This production and analysis of small spherical particles validates the EAC/ALGR classification techniques which work well for spherical particles and not-so-well for large chain aggregates. It is seen from the data and the results of SEM that carbon aerosols can be produced and classified by several methods. These methods being SEM analysis and standard lab methods currently used such as EAC/ALGR counting of nuclei. The SEM method being studied is found to produce results which fall within the range of the currently accepted means of nuclei measurement. The studied method by SEM analysis not only produces a size distribution close to accepted methods but it provides additional information dealing with the morphology of carbon aerosols. This ability to characterize morphology opens many opportunities to explore particle characteristics based on the history of a particle. That is, the effects of an aerosol's exposure to moisture, chemicals, and storage conditions as functions of time. This information can then be used to determine the effects of carbon aerosols upon the environment.

Several areas of investigation remain with this study to improve and enhance the ability to characterize carbon aerosols by SEM analysis. These include (1) the collection process which may enhance particle collection efficiency, (2) improvement of SEM analysis by use of different substrates, (3) chemical analysis which can precisely describe the content of particles collected and (4) improvement of the quality of the micrographs, computer analysis equipment is available which can be used to collect data for size as well as area distributions.

This work will benefit future university aerosol research, e.g. environmental effects associated with jet engine exhaust by future high speed high altitude commercial aircraft as in NASA's high speed civil transport (HSCT) program.

Acknowledgements

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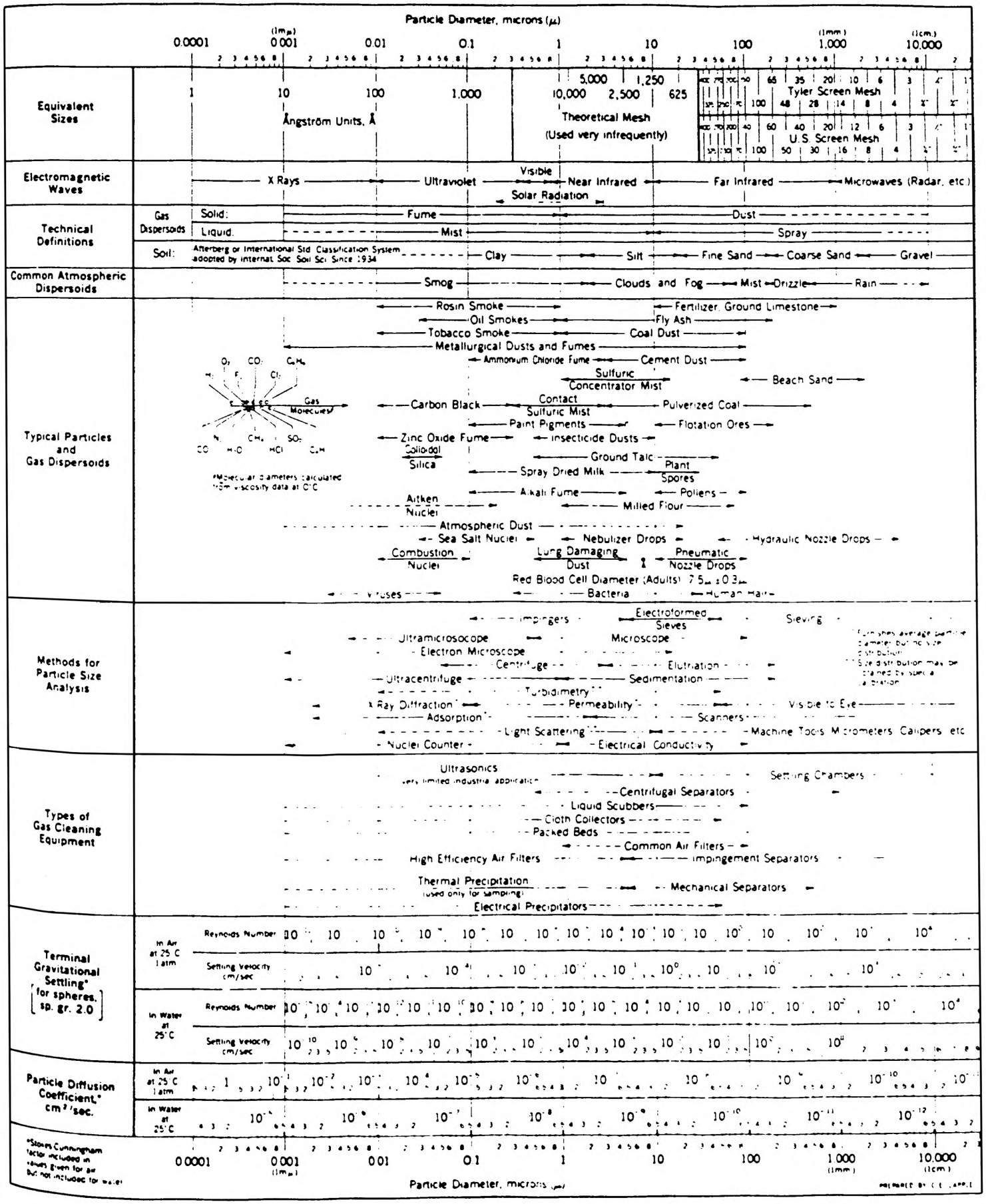

FIG. 1. Summary diagram of particle properties as a function of size, including measurements and gas-cleaning technology. (Courtesy of Stanford Research Institute.)

Combustion Aerosol Generation/Collection Diagram

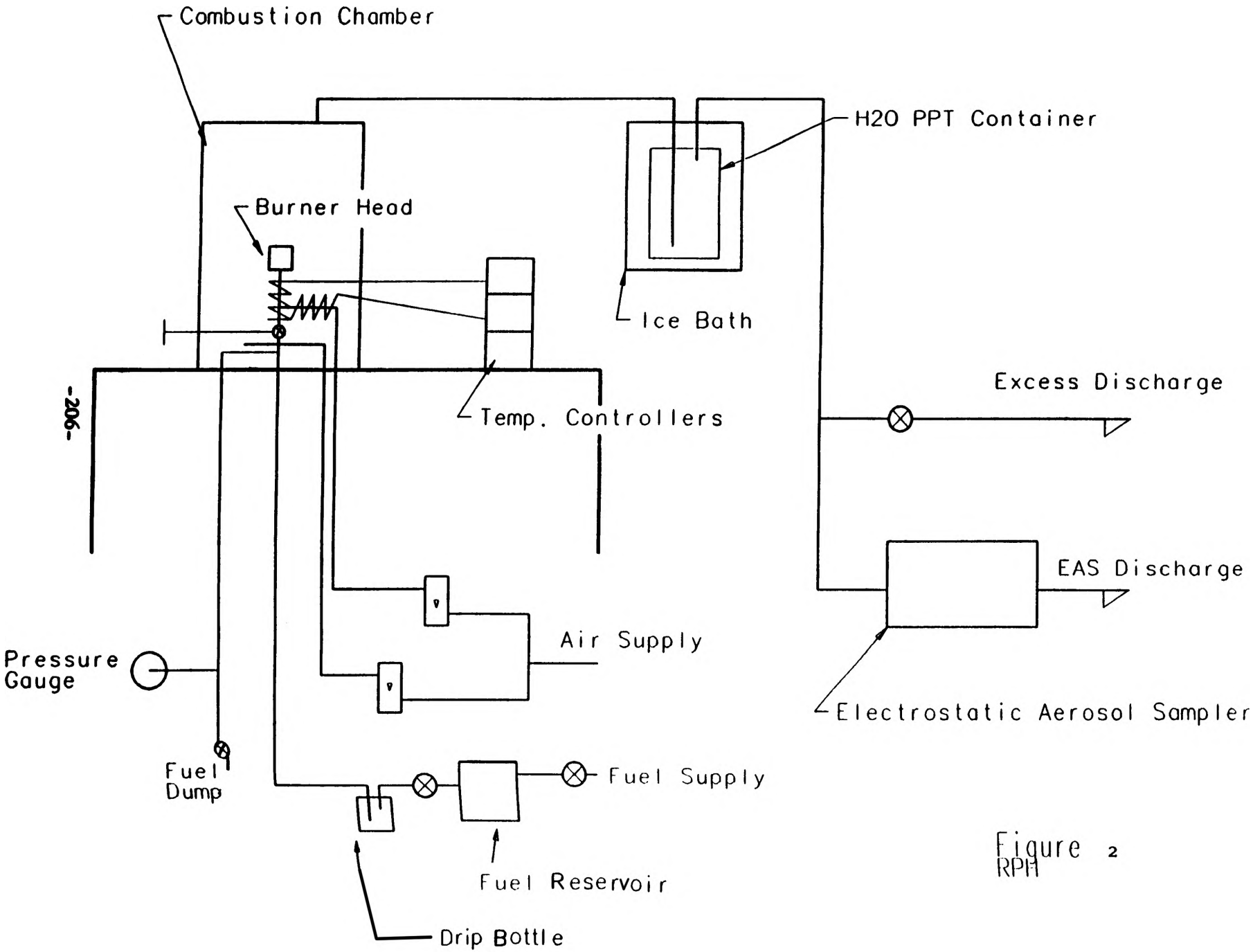

Figure 2
RPH

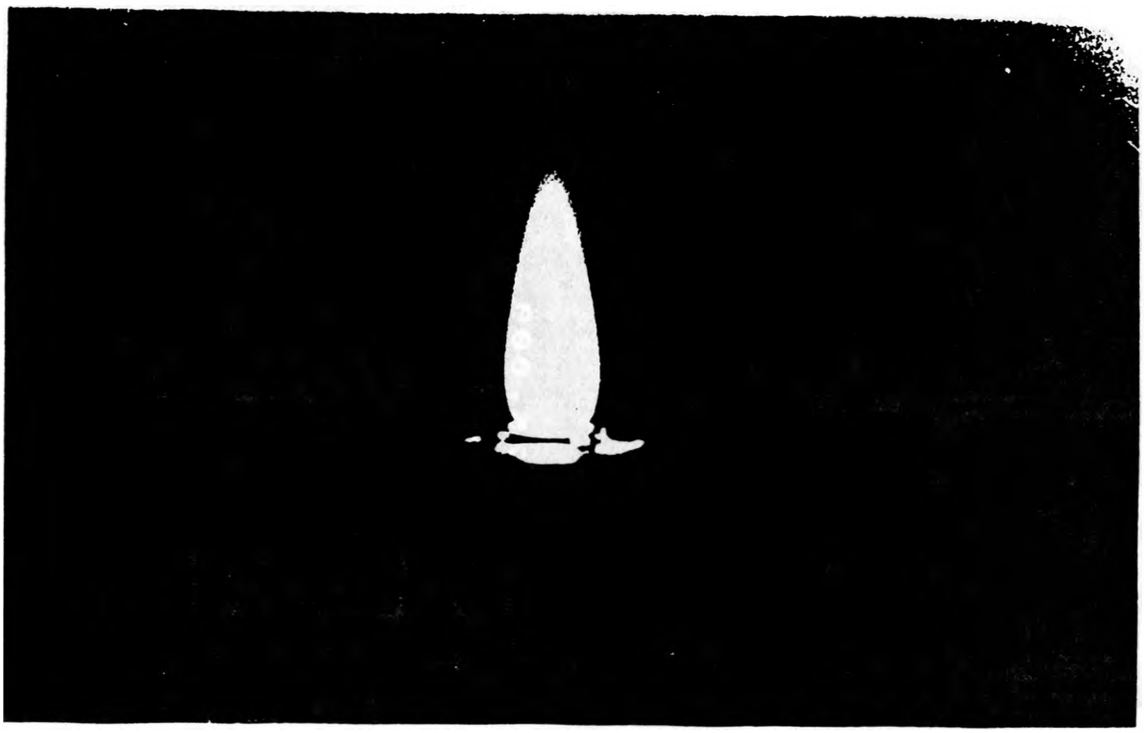

Fig. 3

Flame produced by improved torch

Fig. 4 - Large soot agglomerate (SEM)

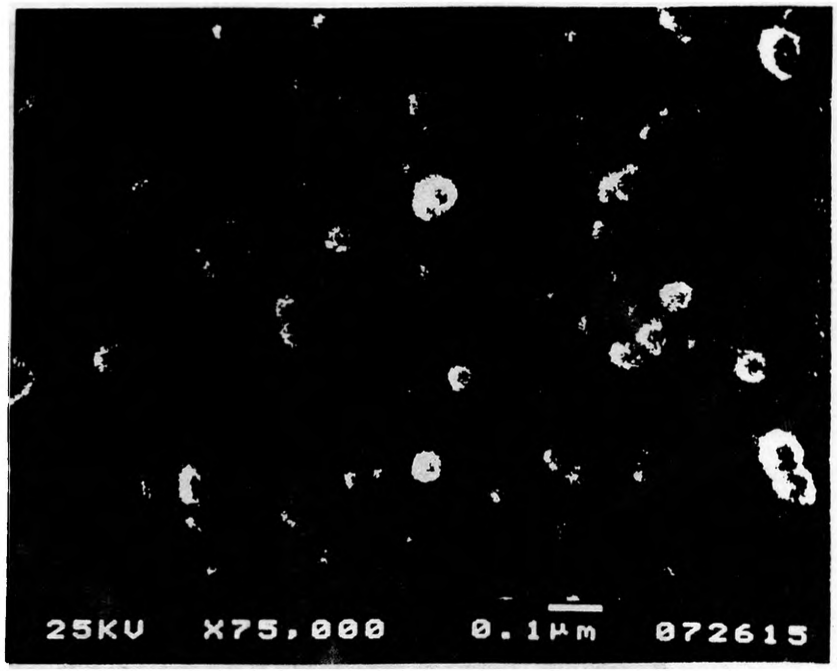

Fig. 5 - Single aerosol particles (SEM)