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ARB Contract A3-092-32
RECOMMENDED METHODOLOGY FOR THE DETERMINATION OF
PARTICLE SIZE DISTRIBUTIONS IN DUCTED SOURCES
EXECUTIVE SUMMARY

Prepared by

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Conclusions:

Three identifiable current and future needs with respect to particle size related sampling of stationary sources were found. These were as follow:

- 1) A method for obtaining particle size distribution information for satisfying needs related to emissions control and downwind transport.
- 2) A method for obtaining size fractionated bulk samples of sufficient quantity to permit chemical analysis which could identify the particle size fractions in which certain toxic, hazardous, or other environmentally significant elements or compounds might be concentrated.
- 3) A method for measuring PM_{10} emissions from stationary sources in support of the ambient PM_{10} regulations and possible future PM_{10} source regulations.

Inertial sizing using cascade impactors was selected as the method for the measurement of particle size distributions. The equipment and protocol provide for measurement of five to eleven size fractions, depending upon immediate goals, over the size range from about 0.1 micrometers to about 15 micrometers. Series (or cascade) cyclones were selected as the method for obtaining size fractionated bulk samples for chemical analysis. The selected equipment and protocol for the latter purpose will provide samples in six size fractions with fractionation points in the range from about 0.25 micrometers to 10 micrometers. Finally, a method was selected for PM_{10} measurements. This method is currently under development by the US EPA. The method uses the technique of emission gas recirculation to provide full flexibility in meeting sampling traverse requirements while maintaining a 10 micrometer particle cutoff diameter and general compatibility with the current particulate sampling method (Method 5).

PERFORMANCE SPECIFICATIONS

SUMMARY OF AGENCY NEEDS

The particulate sizing needs of the ARB were reviewed and assessed using: the original request for proposal on which the contract was based, a meeting with personnel of the ARB in Sacramento, a meeting with personnel of the South Coast Air Quality Management District, and a telephone survey of various ARB personnel and its contractors. The purpose of the review was to define a set of performance criteria on which to base the selection of particulate sizing methods for application mainly by the ARB, but it is expected that local air quality management districts, industries, and consulting firms within the state of California will also become users of the methods as well. As determined from the meetings and surveys, potential end uses of the data to be obtained by the particle sizing methods include the following: basic research regarding source emissions, potential health and toxicity effects related to specific elements and compounds which might be concentrated in particular ranges of particle size, development of particle size related emission inventories, and possible compliance methods for potential particle size related emission standards (e.g. PM_{10}). It was anticipated that three or four judiciously selected methods would cover the bulk of the agency's particle size related sampling needs.

The more important factors to be considered in the review included such items as the sizing ranges and resolutions needed to accomplish the goals of various parts of the agency, the particle size conventions that were most appropriate for the agency's applications, possible needs for chemical analyses of size-fractionated particulate matter, and condensable and/or reactive components of the gas streams. Other items to be considered in arriving at the specifications were such things as test duration (sampling time and number of samples required) that might be needed to characterize a source, ease of handling of equipment, applicable concentration range, ease of sample recovery, applicable range of source conditions, interferences, and accuracy.

Broadly speaking, one can divide the ARB current or potential needs with respect to particle sizing into three classes: (1) regulatory, including setting of emission standards and compliance testing; (2) control strategy development (emission inventories) and permitting (control device selection, etc.); and (3) basic research and development. Of course, considerable overlap exists in the types of information needed for each of these activities.

As currently foreseen, possible regulatory action on emissions may take place based on one or both of two particle size classes. The first, and more likely, of these possible regulatory actions is related to the PM_{10} class (particles having aerodynamic diameters smaller than 10 micrometer, μm) for which a state ambient air regulatory standard has already been set. The second class for possible action concerns fine particles, say those particles having aerodynamic diameters smaller than 2.5 μm . In either case, the regulations may be chemical species and/or industry or process specific as well as particle size specific. If particle size specific regulations are set, compliance test methods would be a concomitant necessity. Development of an emissions inventory would be a preliminary activity prior to such regulatory action - such an inventory is

currently being constructed within the ARB for the PM_{10} class based on such information as is now available. The number of size classes (and the resolution) required for these activities is obviously limited - only one or two size cuts are needed and relatively simple and inexpensive techniques are desirable if they are to be used as compliance tools.

Greater resolution than that needed for compliance testing is desirable for activities related to permitting. The performance of many (or most) particulate control devices can be predicted for a given source from a broad base of experimental data and models provided that the gas stream conditions and the particle size distribution of the material to be collected are known. In most cases, the critical size range for estimating the probability of achieving a required level of control in this fashion is from about $0.1 \mu m$ to $20 \mu m$. Resolution into about eight size classes, evenly spaced in terms of the logarithm of particle diameter, over the latter range is generally sufficient. In cases in which specific target chemical species may not be homogeneously distributed with respect to particle size are of interest, size segregated samples suitable for chemical analysis may be needed in addition to data for overall size distribution. The size resolution for this application need not be as great - three to five size fractions may be adequate.

The needs of the agency with respect to basic research at the present time appear to fall into three areas. The first is providing support for the activities previously described; the second is the development of a data base characterizing the principal types of industrial emissions in the state; and the third is concerned with particle chemistry. The main concerns in the latter area appear to be primary emissions of toxics and emissions of catalysts which may play roles in secondary aerosol formation.

SPECIFICATIONS (Performance Criteria)

The specifications were subdivided by usage rather than attempting to generate a uniform set of requirements for all sampling methods. Some general specifications can be listed which are common to all sampling requirements. In addition to these general specifications, separate specific recommendations were made regarding specifications on which to base the selection of methods for each of three classes of measurements: (1) PM_{10} , (2) moderate to high resolution particle size distribution measurement, and (3) sampling to collect size fractionated material for chemical analysis. Separate specifications were better suited for purposes here since the requirements for each of these classes were different and in some cases conflicting. The subject of condensables is treated separately since problems related to them will probably require special treatment(s) in a fashion(s) that can be applied in common to all of the three particle sizing methodologies. In arriving at the specifications, attempts were made to confine them to properties that would insure that the methods finally selected would be optimal for ARB's purposes without making them so unduly restrictive that little likelihood would exist of finding methods that could actually achieve them.

First, it was necessary to clarify the nomenclature used for defining particle diameter. A number of conventions are used as bases for the presentation of particle size distributions with respect to both the definition of

particle size and the property of the distribution presented. Particle size is most often defined in terms of a "diameter" implying that the particles are being treated as spheres - depending upon the circumstances this may be rigorously true or only a useful approximation. The most frequently used diameter bases in air pollution work are:

True diameter - the actual diameter of the particle. Useful only if the particles are spherical.

Stokes diameter - the diameter of a sphere of the same density and settling velocity in air as the particle in question. (Equals the true diameter of spherical particles). This definition is often used as an approximation for estimating the volume or surface area of irregular particles.

Aerodynamic diameter - the diameter of a unit density sphere which has the same settling velocity in air as the particle in question.

Volume equivalent diameter - the diameter of a sphere having the same internal volume as the particle in question.

Surface equivalent diameter - the diameter of a sphere having the same total surface area as the particle in question.

Area equivalent diameter - the diameter of a sphere having the same projected area as the particle in question.

No set conventions exist for selecting the diameter basis for data presentation, however, certain bases are favored for use in particular applications. For instance, the aerodynamic basis is the preferred choice in work related to inhalation and health effects and in wet scrubber technology; while the Stokes diameter is favored for work related to light scattering (opacity) and in electrostatic precipitation. In most cases, convenient transformations exist for changing from one basis to another, however, this may not be the case if the particles are highly irregular in shape. Because of their widespread use in research related to health effects, visibility, and control device technology, the methods best suited to ARB's purposes will be those for which the diameter bases are either aerodynamic or Stokes. Therefore the ability to provide data on an aerodynamic diameter basis was set as a requirement for each method.

The conditions under which industrial source sampling are carried out in themselves dictate certain specifications which will be shared in common for all methods to be selected. Among these are features related to portability, support services required, operational temperature limits, corrosion resistance, sampling platform and port dimension requirements, and applicability over the range of particulate concentrations expected for industrial sources. These common specifications are as follows.

1. Provide a measure of total particulate loading.
2. Provide usable samples from sources having any concentration within the range from 0.005 to 50 grains per cubic foot.
3. Provide measurement of the weight fraction of particles smaller than any (the) specified size to within 10 percent of the stated size, with 95 percent confidence.
4. Be applicable in stacks having -5 to +20 inches of water pressure differential to ambient.
5. Be applicable at sources having stack gas temperatures in the range of 0 to 450° Celcius.
6. Be capable of obtaining a representative sample from stacks having gas velocities in the range from 10 to 100 feet per second.
7. Have a maximum single component weight of 50 pounds.
8. Be resistant to corrosion by acids and alkalis.
9. Require port dimensions no larger than four inches in diameter. (Three inches preferably.)
10. Be capable of traversing the stack.
11. Require no greater electrical service than that needed for EPA Method 5.
12. The maximum length of any single component should be six feet (probe excluded).

Devices meeting these specifications are man-carryable, amenable to use in the normal physical environment under which source tests must be carried out, and capable of withstanding exposure to the (frequently) hot, corrosive stack gases. The velocity and traversing requirements are necessary in order to insure that representative samples can be obtained, even when the particulate matter is stratified within the duct or stack.

Specifications for the Source PM₁₀ Method

The PM₁₀ methodology can be expected to be used by the ARB, local control districts, and industrial and consulting organizations. Because the method is to be developed in support of the California PM₁₀ ambient air quality standard, which is based on aerodynamic classification, the aerodynamic diameter basis is appropriate for it as well. If it is to be used as a compliance method, it should be applicable by personnel having the same level of expertise as is currently required for ARB Method 5. The method should be modeled after ARB Method 5, except that the sampling nozzle shall be followed immediately by an inertial collection device having an efficiency of 50% at an

aerodynamic diameter of 10 μm . Deviations from Method 5 should be limited wholly to those required to achieve the 10 μm size cutoff. The sharpness of cut of the collector should be such that its collection efficiency curve when plotted in log-probability coordinates has a geometric standard deviation less than or equal to 1.7. The latter value matches that specified for ambient PM_{10} samplers.

Specifications for the Size Distribution Method

Moderate to high resolution particle size distributions are needed in research applications of ARB and for use as a basis for estimating expected efficiencies of control devices for control device selection and permitting. Experience has shown that for most applications the critical range over which size distribution data is needed is from about 0.2 μm to 10 μm , together with total concentrations for the ranges smaller than 0.2 μm and larger than 10 μm . Sufficient resolution for modeling the effects of control devices, estimating overall control device efficiencies, predicting stack opacities (for noncondensing stacks), and characterizing the fractional collection efficiencies of operating control devices can be provided by separating the aerosol particles into about six to eight size classes within the 0.2 μm to 10 μm size range. The latter range also includes "respirable" particles and consequently is of special importance in health effects. The actual size distributions of most natural and industrial aerosols are such that they can best be described by distribution functions in which the logarithm of the diameter is the argument (for instance the log-normal distribution). Thus the resolution specification for the method can best be given in terms of log (diameter). Size fractionation at steps of 0.25 to 0.333 in log (diameter) over the 0.2 to 10 μm range is expected to be adequate for most foreseeable needs of the ARB and other potential users of its methods. The sharpness of cut provided by the classifier(s) should result in separation efficiency curves having geometric standard deviations of less than 1.5. The size fractionation must be well characterized with respect to performance changes produced by changes in operating conditions.

Specifications for the Sized Chemical Analysis Method

At present the needs regarding size fractionated material for chemical analysis fall almost entirely in the area of basic research, and more especially in the area of research on primary emissions of toxic materials and priority pollutants. Interests in toxics include metals (e.g. chromium, beryllium, and cadmium), PAH's, and dioxins and furans. In the case of the metals, concentrations within specific valence states are sometimes of greater importance than just the total concentrations (e.g. chromium). Aside from toxics, materials which can serve in the formation of secondary aerosols as catalysts for chemical transformations and in the formation are of interest.

Discussions of sampling for chemical analysis can be broken into two parts; particulate and condensables. The first particulate is comprised of sampling for materials that are in solid or liquid form at stack conditions. Many toxics however, - both metals (e.g. arsenic, selenium, and mercury) and

organics - are materials which can have substantial vapor phase concentrations at flue gas conditions. Treatment of sampling for analysis of condensables following section devoted exclusively to the subject of condensables is discussed in the following section.

The number of size fractions needed to meet the goals of the various potential users of this methodology is somewhat less than that needed for particle size distribution determinations. The need to collect sufficient material in each size fraction to satisfy the analytical needs makes it desirable to minimize the number of fractions. Obvious cut points for health effects research include the fine particle fraction (2.5 μm) and the PM_{10} (10 μm) fractions. Isolation of a frequently occurring submicron combustion mode is also desirable, indicating the need for a cut near 0.25 μm . Interest in studies of catalysts have indicated a desire for a cut at 1 μm . A sharpness of cut specification of 1.7 or less for the geometric standard deviation in the efficiency curve should be suitable here.

The sample must be collected in an unadulterated, contaminant free form if valid analyses are to be obtained. Discussions with personnel at analytical laboratories who have provided analyses of particulate matter for the ARB in the past revealed that sample quantities substantially larger than a few milligrams are needed for many, if not most, of the analyses which they have been called upon to perform. Therefore the method must be capable of providing sample quantities in the tens or hundreds of milligrams for the individual size fractions.

Specifications Regarding Condensables

The term condensables as used here includes all materials which are wholly or partially in the vapor phase at stack conditions but will be driven to be attached to particles by thermodynamic processes upon discharge to the atmosphere. Thus any material which becomes attached to the particulate matter by condensation, sublimation, prompt reaction, or sorption is considered part of the condensables.

The transfer of material to and from the vapor and solid/liquid phases is a complex dynamic process for which the rates and equilibria depend on the concentrations, temperatures, and mixing rates of the stack gases and the ambient air. Rigorous duplication of the actual plume conditions, even at a single defined set of atmospheric conditions, including matching of mixing rates, mixing ratios, etc. becomes a practical impossibility. Therefore the development of a usable method for sampling condensables will require that a number of compromises be made in order to obtain a workable approximation of plume conditions.

The simplest method for sampling condensables is that used in the California Method 5, in which the condensables are defined as the material collected in bulk in chilled liquid filled impingers. This choice serves a useful regulatory purpose in that the Method 5 "back half" is not likely to underestimate the total condensable emissions. However, this method cannot provide information regarding the ultimate distribution of the collected

materials with respect to particle size, nor can it provide accurate data on material which would attach to the particles by sorption as most of the condensation takes place on surfaces other than those of the particles. Further, by placing the collected material in liquid solution or suspension, it becomes possible for reactions to occur which might not take place if the materials had condensed or sorbed on the surfaces of particles in the atmosphere. Therefore impinger collection is not a useful technique for applications in which it is important to determine the effects of condensable vapors on the size distribution of the final aerosol, or for applications in which it is necessary to apportion the condensable contribution to specific size classes. For these applications, we recommend that sampling methods utilize a form of air dilution which simulates the mixing processes that take place in the plume. Suggested specifications for a condensable materials sampling method are:

1. Dilution of a flue gas sample stream with conditioned ambient air be used to simulate the plume processes.
2. A standard dilution condition be used for all sources (e.g. 25:1 dilution ratio, with filtered dilution air at 25° C and 40% RH).
3. The sampling system capable of being interfaced with currently used ambient air particle sizing and collection devices.

FINAL RECOMMENDATIONS FOR THE NUMBER AND TYPES OF METHODOLOGIES TO BE DEVELOPED

In summary, a total of four sampling methodologies appear to be needed. The first method would be intended to support PM₁₀ regulatory work and would provide size fractionation at only 10 µm with the method being generally consistent with current total particulate methods. The second methodology would cover the entire size spectrum but provide detailed information only within the range from about 0.2 µm to 10 µm. That range being the most critical for health effects studies, control device selection and evaluation, and opacity/visibility related work. The third method would be intended to provide fairly large samples, with less size resolution, for chemical analysis. And finally, the fourth method would provide a means of quantifying the contributions of condensibles in as realistic a fashion as circumstances permit short of calling for actual sampling in plumes.

After these recommendations were submitted, a project review meeting was held at the ARB offices in Sacramento. The result of this meeting was a consensus that the first three methodologies described above should be pursued further. However, development of a condensibles method would be deferred and such a method would not be developed on this contract.

METHODS REVIEW AND EQUIPMENT SELECTION

Task 2 of the project called for a literature survey of potentially relevant sizing methods and associated commercial instrumentation using the respective methods. This survey was conducted independently of the establishment of the performance criteria in order to compile a comprehensive list of all possible approaches to the source measurement problem. A summary of the information gathered in the literature survey is given in an Appendix to the final report. The specifications for the three methods were applied to the results of the literature survey and used to select recommended test methods and instrumentation for each of the three specified ARB Methods: (1) Source PM_{10} Method, (2) Size Distribution Method, and (3) Sized Chemical Sample Method. The recommended methods are summarized in Table 1 and described in the following paragraphs.

SIZE DISTRIBUTION MEASUREMENT METHOD

Moderate to high resolution particle size distribution information is needed for research applications and for control device selection and permitting in that this information provides a basis for estimating expected efficiencies of control devices. Experience has shown that for most applications the critical range over which size distribution data is needed is from about 0.2 μm to 10 μm , together with total concentrations for the ranges smaller than 0.2 μm and larger than 10 μm . Sufficient resolution for modeling the effects of control devices, estimating overall control device efficiencies, predicting stack opacities (for noncondensing stacks), and characterizing the fractional collection efficiencies of operating control devices can be provided by separating the aerosol particles into about six to eight size classes within the 0.2 μm to 10 μm size range. The range also includes "respirable" particles and consequently is of special importance in health effects.

After reviewing all available methods for measuring particle size distributions, the method of inertial separation using cascade inertial impactors was selected as the preferred technique (standard method) for measuring particle size distributions of effluents from stationary sources. This method, of all those available, most nearly met all of the specifications previously given.

For years inertial impactors have been commonly used to determine the particle size distribution of particulate matter suspended in industrial process gases, especially those emitted to the atmosphere. Impactors have several advantages over competing equipment: they are compact, they can be inserted directly into gas ducts (avoiding the problems associated with extractive sampling), they are fairly accurate, and they produce information which has been widely used and understood. The majority of the particle-size distribution data available on industrial process streams have been taken using cascade impactors covering a diameter range of 0.3 to 20 μm . These devices consist of serial configurations of several impaction stages. Each stage of the impactor removes particles of a characteristic diameter, starting with the largest and progressing to smaller diameters. The popularity of these devices is due not only to their simplicity of design and operation but also to their

Table 1. Equipment Selection Summary
Advantages and Disadvantages

Stationary Source PM₁₀ Method:

Recommended: Cyclones:

Advantages: Sharp 10 μm cut, no bounce or overload problems

Disadvantages: Flow limitations imposed by 10 μm cut, high temperature performance theory not well established

Option: Impactors:

Advantages: 10 μm data with minimal flow limitations, theory well established

Disadvantages: Particle bounce, overloading, operator skill level

Size Distribution Method:

Recommended: Impactors:

Advantages: Good size resolution, light stage loadings

Disadvantages: Operator skill level, manhour intensive, overloading

Option: Cyclones:

Advantages: No overloading at high concentrations, simultaneous bulk chemical sample

Disadvantages: Limited size resolution, require large catches, manhour intensive, high temperature performance not well established.

Sized Chemical Sample Method:

Recommended: Cyclones:

Advantages: Large bulk sample, contamination free

Disadvantages: Manhour intensive

Option: Impactors:

Advantages: Simultaneous size distribution data

Disadvantages: Light samples only, substrate contamination, operator skill level, particle bounce, manhour intensive

Plume Condensibles Method (with sizing capability):

Recommended: Plume Simulator:

Advantages: Reasonable estimate at minimal cost, uses ambient instrumentation

Disadvantages: Operator skill, prototype equipment only

Option: Require Plume Sampling:

Advantages: Accurate condensibles assessment

Disadvantages: High cost (airplanes, etc.), limited time in the plume, weather dependency

portability and adaptability to a large variety of aerosol streams. When used properly, cascade impactors are capable of providing particle size distribution measurements extending from below 0.5 μm to diameters of 10 μm and above.

Figure 1 is a schematic diagram illustrating the principles of particle collection common to all inertial impactors. The sample aerosol is constrained to pass through a circular hole or rectangular slit to form a jet that is directed toward an impaction surface. Large particles will possess sufficient inertia to cross the gas streamlines and impact on the collection surface. Particles having lower momentum will follow the gas stream past the collection plate. In a cascade impactor the gas stream passes sequentially through several impaction stages designed to remove successively smaller particles, thus collecting the airborne particulate matter in a series of discrete size fractions.

The probability of collection in an impactor stage typically varies with particle size as shown in Figure 2. Ideally, an impaction stage would provide complete collection of all particles larger than a known size and pass all smaller particles. In other words, the ideal collection efficiency curve would be a step function. In practice, the real stage collection efficiency curves such as the one schematically illustrated have sharp enough transitions to be useful for aerosol size distribution measurements. The behavior of a real stage in operation is then described in terms of a characteristic particle diameter (D_{50}) which is collected with 50 percent efficiency for the operating conditions used.

Impactors with a wide variety of geometrical configurations have been observed to have the qualitative behavior described above. Impactor stages have been constructed with one to several hundred holes or rectangular jets, depending on the desired jet velocity and volumetric flow rate. The number of jet stages ranges from one to about 20 for various impactor geometries reported in the literature; most commercially available impactors use 5 to 10 stages. Three commonly used impactors are shown in Figure 3. The University of Washington Mark V Cascade impactor was specifically recommended for the purposes of the ARB. This device is illustrated in Figure 3.

OBTAINING SIZE FRACTIONATED SAMPLES FOR CHEMICAL ANALYSIS (Sized Chemical Sample Method)

The environmental effects of particulate emissions from stationary sources depend on both the composition and particle size distributions of the emissions. Although composition information obtained from bulk samples can be used in estimating potential effects, the possibility - indeed, probability in many cases - of the composition of emissions being inhomogeneous with respect to particle size makes it desirable to have the capability of determining composition by particle size. Redispersion and separation of bulk samples might be used for this purpose; however, if all or part of the aerosol material were capable of coalescing it would be impossible to reconstitute the original size and composition distributions. Further, even in the case of dry solids, it becomes difficult or impossible to remove submicron particles from the surfaces of larger ones once contact has been made. Therefore, redispersion and subse-

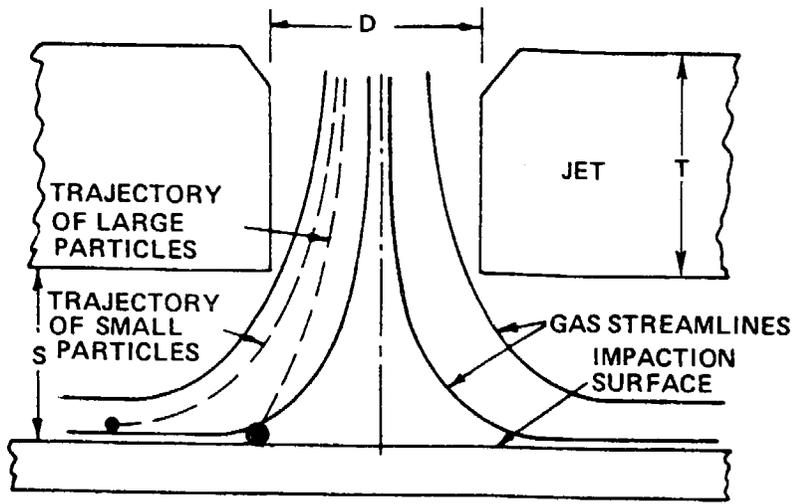
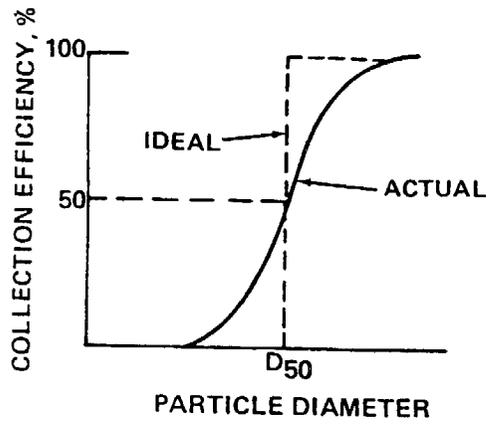


Figure 1. Typical impactor jet and collection plate.



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Figure 2. Generalized stage collection efficiency curve for a cascade impactor.

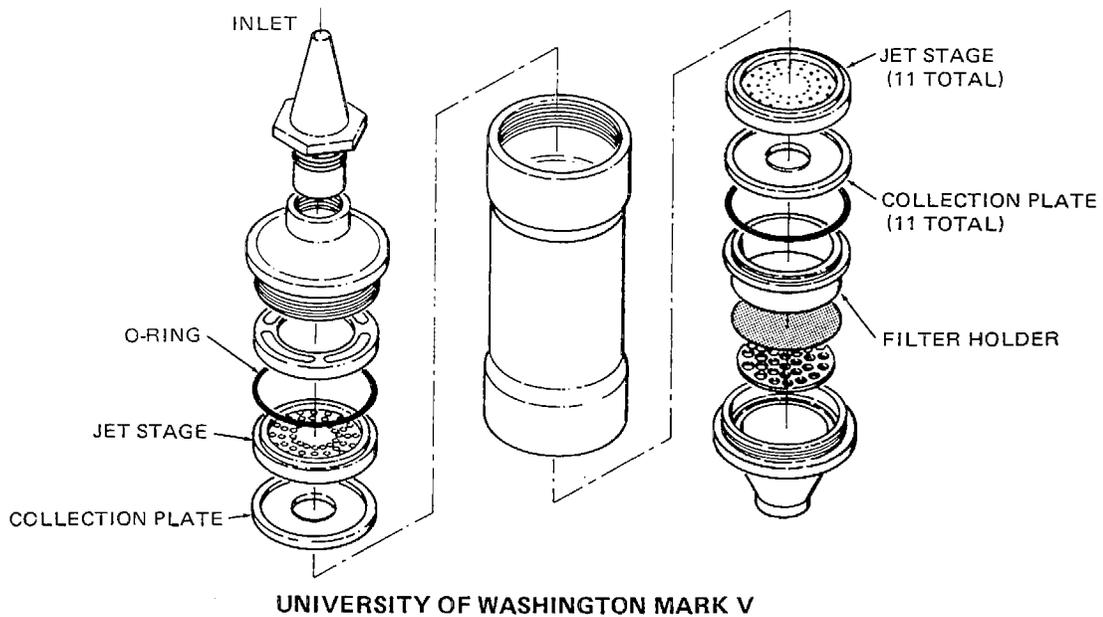
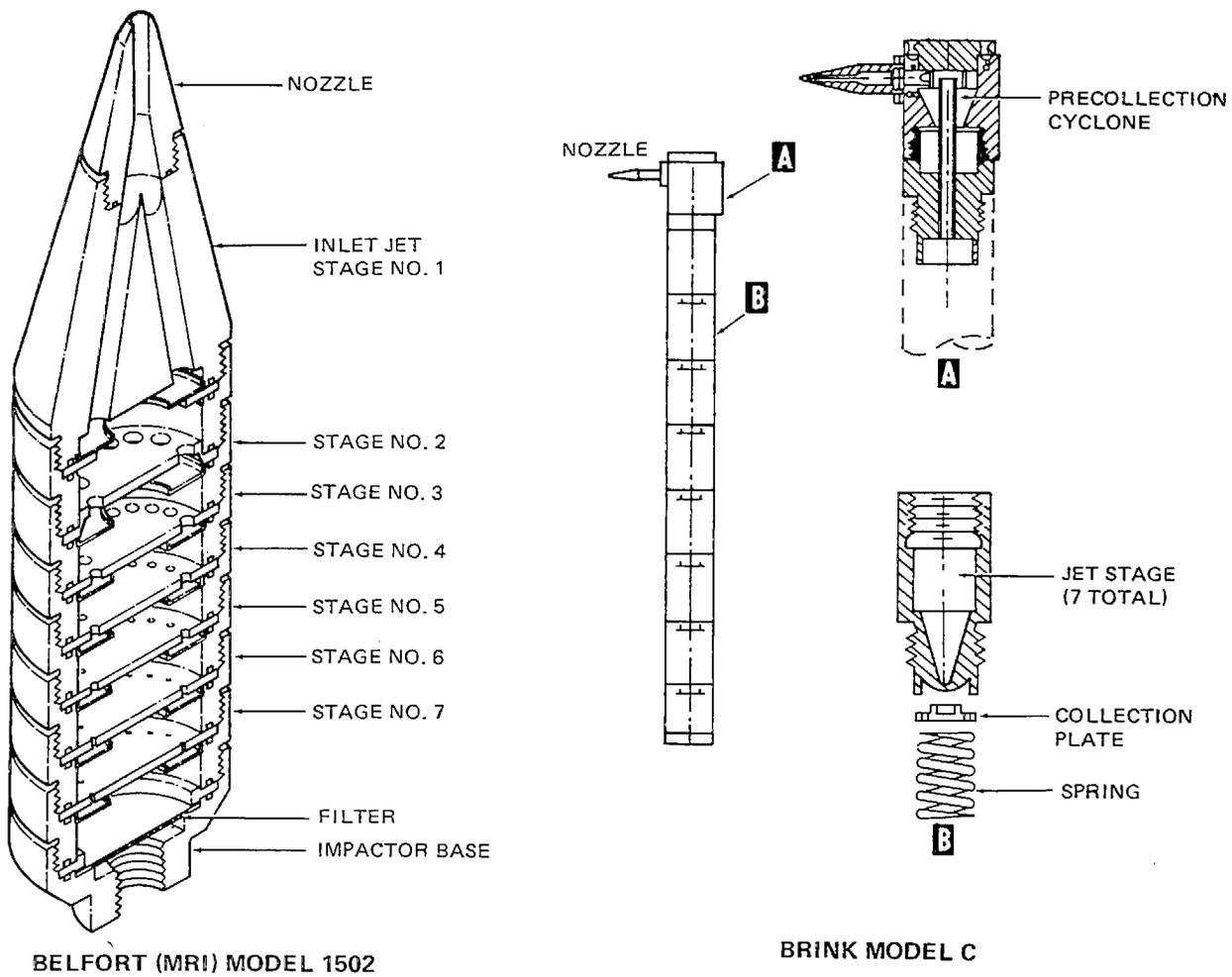


Figure 3. Schematics of three typical commercial cascade impactors.

quent partitioning by size of a bulk sample is a poor choice for the purpose. A method in which the particles are separated by size as they are collected is needed.

Two forms of inertial particle size separators are commonly used in source sampling - cascade impactors and cyclones. Impactors have a number of advantages for use in measuring particle size distributions and were selected for use by the ARB for that purpose, but they are not well suited for providing samples for chemical analysis. The quantities of material that can be collected on an impactor stage are limited to a few milligrams as most stages in an impactor sample will have catches of only fractions of a milligram. (The fact that only small amounts of material can be collected is not a problem in size distribution measurements, in fact, the need to collect only small amounts of particulate can be advantageous because it reduces the time required to perform the sampling.) Omission of stages in impactors frequently results in operational failures because of particle bounce, consequently this approach cannot be used to reduce the number of fractions and increase catch sizes. Moreover, the adhesives or surface coatings used to increase particle retention in impactors can result in intolerably high backgrounds and/or interferences. Cyclones, on the other hand, do not suffer problems from particle bounce or retention, have capacities of the order of grams, and require no surface coatings which might lead to interferences. Therefore, they were selected as the means to obtain size fractionated samples for chemical analysis. Cyclones may also be used to good advantage for measuring particle size distributions when the particulate concentration in the stream is very high. In such circumstances cascade impactors (which are normally used for size distribution measurement) may be impractical because of rapid overloading.

Cyclones are forms of centrifugal particle collectors in which the circulation of a particle laden gas stream about a central axis is induced by conversion of forward motion through the cyclone inlet. The gas enters through the inlet tube from whence it passes into the cylindrical body of the device. Here it acquires a spiral motion, descending along an outer spiral toward the base of the cyclone for some distance; it then undergoes an abrupt change of direction and continues upward to and through the exit tube. The length of the exit tube is made great enough that the gas does not simply "short-circuit" the intended flow path and flow directly from the inlet to the outlet tube. The flow in the central core of the cyclone, after the gas has turned upward, may proceed in a tighter inner spiral, or may flow in a rectilinear path to the outlet tube. The latter is believed to be the case when the Reynolds number of the flow in the outlet tube is low. Large particles, because of their inertia, will tend to cross the curving flow streamlines and thus drift towards the wall of the cyclone body where they become disentrained upon contact. Additional separation may take place by impaction on the wall opposite the inlet and at the bottom of the outer spiral where the reversal in flow direction takes place.

The use of cyclones is not without drawbacks. First, particulate catches are frequently distributed over rather large surface areas within the cyclone. This makes sample recovery difficult, especially if the recovery is to be sufficiently complete so that the data can be used reliably for concentration measurement. This is not a problem if the catches are large, but long sampling

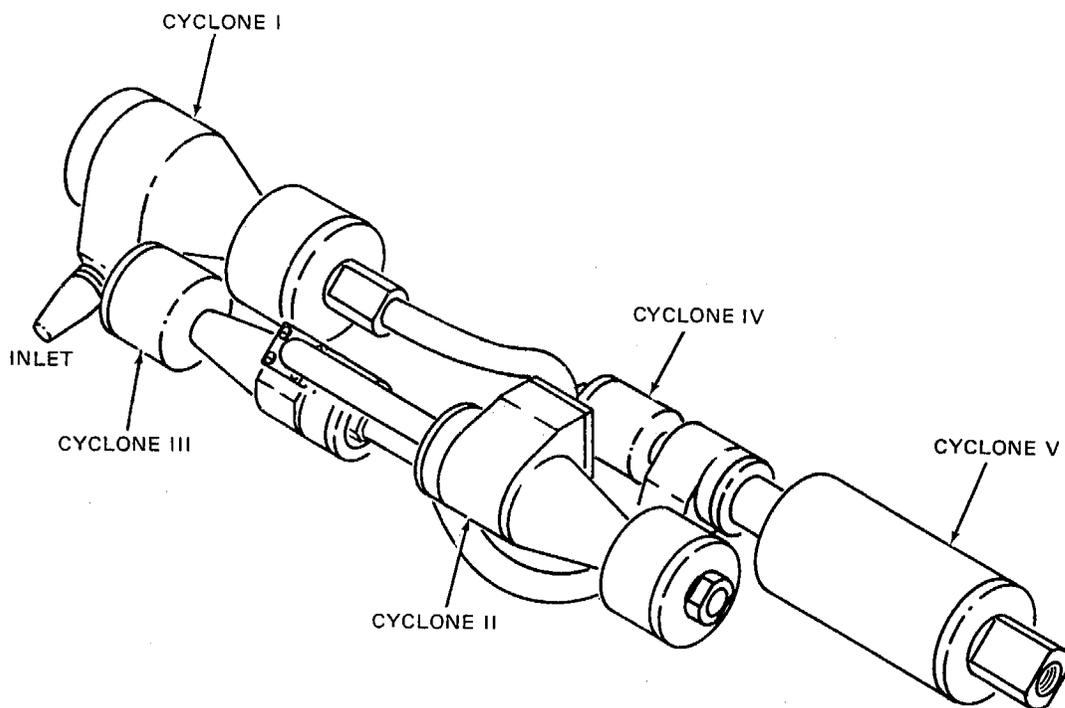
durations may be required to obtain large catches. Second, current theories on cyclone operation are rudimentary at best, and none do well in predicting cyclone performance from geometrical and flow considerations. Therefore, extensive calibrations are needed so that good empirical relationships can be developed for use in calculating the sizes of the collected particles. This results in less flexibility in construction and operation being available to the user than is the case with impactors.

Target cutoffs for particle size fractions for chemical analysis were set as follows: A cut at or near a diameter of ten (10) micrometers was desired to provide samples which conform to the PM₁₀ ambient air standard. Isolation of the fraction commonly designated as respirable suggests that a cut near 2.5 to 3 micrometers is desirable. Isolation of the one (1) micron and smaller fraction was desired by researchers working in the field of catalytic effects in the atmosphere, and finally, a cut near 0.25 micrometers was suggested since many combustion aerosols contain a sub-micron mode in their distributions which is known to differ chemically from the remainder of the material and which almost always falls below that size. The sampler which most nearly met these specifications was the SoRI/EPA designed Five Series Cyclone. These cyclones are available commercially from a number of manufacturers both in and out of the United States.

The design goal for the SoRI/EPA Five-Stage Cyclone Sampler, shown in Figure 4, was five equally spaced particle size cuts (D_{50}) on a logarithmic scale within the range of 0.1-10 μm . This in situ sampling system operates at a nominal sample flowrate of 28.3 L/min (1 ACFM), and is compact enough to fit through a 10 cm (4 inch) diameter port. The sampler consists of five cyclones and a backup filter connected in series. Since the backup filter is a separate unit, either a flat filter mat can be used or, in the case where there is a large percentage of fine particulate present, a thimble type filter can be used. The first of the five cyclones, Cyclone SRI-I (which separates the large fraction of particulate matter from the gas stream), accepts a range of nozzles to facilitate isokinetic sampling.

SOURCE PM₁₀ MEASUREMENT METHOD

Procedures for measurement of size-specific particulate emissions (such as PM₁₀) are more complex than, but similar to, those for total particulate sampling by EPA Reference Methods 5 or 17. Potential sampling biases exist due to variations in the spatial distribution of particulate concentrations across the sampling plane defined by the duct cross-section. Likewise, temporal variations in particulate concentrations due to process variations can cause inaccurate or unrepresentative results. Also, if the gas velocity entering the sampling nozzle is not the same as the local duct velocity, particulate matter will be selectively depleted or enriched in the sample gas stream due to inertial separation at the nozzle entrance. EPA Reference Methods 5 and 17 deal with these problems by placing specifications on the sampling location to minimize stratification and by sampling isokinetically at an array of points spanning the sample plane. Isokinetic sampling is accomplished by measurement of the duct velocity at each point and adjustment of the sample train flowrate so that it is proportional to velocity.



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Figure 4. Environmental Protection Agency-Southern Research Institute Five-Stage Cyclone.

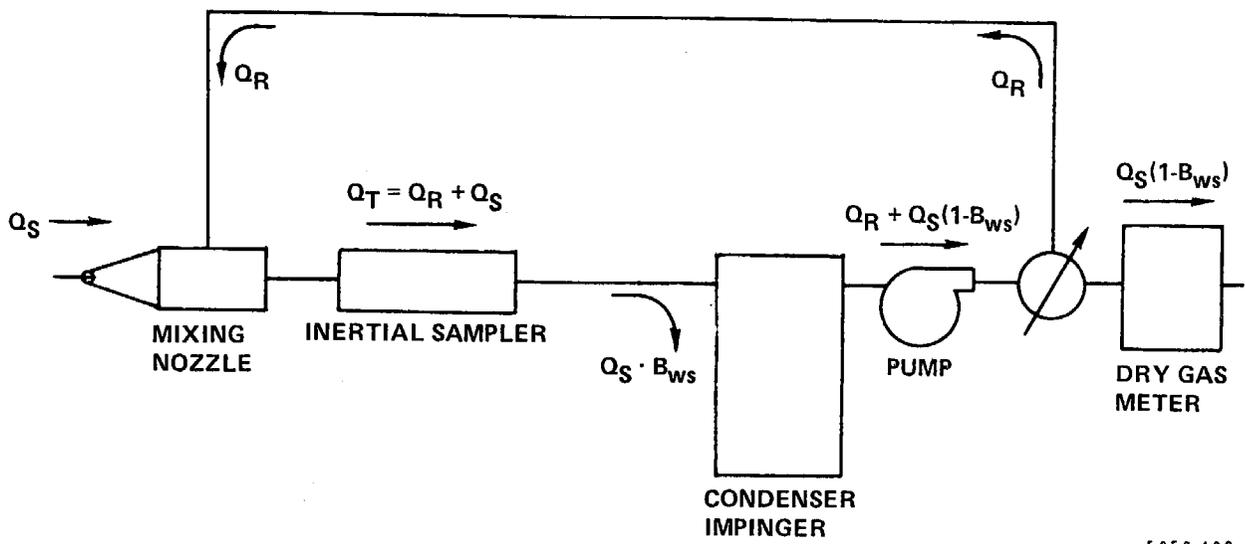
Size-specific emission measurements are accomplished by sampling through an inertial size-separation device like those already described. The procedure and equipment must deal with the same problems described above while, in addition, maintaining a constant flowrate through the separator to provide the desired size cut ($10\mu\text{m}$ for the present purpose). This constraint on flowrate causes additional complexity in sampling methods for PM_{10} as compared to those for total particulate matter. The constraints of isokinetic sampling and a specified flowrate for the size cut directly conflict, resulting in a requirement for additional hardware.

The method selected uses a special sampling nozzle which allows isokinetic sampling while maintaining the specified flowrate for a $10\mu\text{m}$ size cut provided by an instack particle classifier. The sampling train incorporates the principle of emission gas recycle (EGR). It allows a variable fraction of conditioned and filtered exhaust gas from the sampler to be added to the sample stream between the sample nozzle and the inertial classifier. This allows a preselected constant flowrate to be maintained through the inertial classifier while the gas flowrate into the sampling nozzle is adjusted to remain isokinetic with the local duct velocity. This system currently exists only in prototype form.

The principle of operation of the EGR train is illustrated in Figure 5. Stack gas is extracted isokinetically at volumetric flowrate Q_s . If the stack moisture fraction is B_{ws} , the sample flow consists of $Q_s \cdot B_{ws}$ moisture and $Q_s(1-B_{ws})$ dry gas flow. At the mixing point a flow, Q_r , of dry recycle gas is added to the sample stream to bring the total flowrate to the predetermined constant level, Q_t . In the impingers or condenser, the moisture content $B_{ws} \cdot Q_s$ is removed. After the pump and total flow metering element, the recycle flow Q_r is diverted by means of adjustable valves. By mass balance, in a leak-free system, the remaining flow which passes through the dry gas meter and orifice will simply be $Q_s(1-B_{ws})$, exactly as would occur in an isokinetic sampling train without gas recycle.

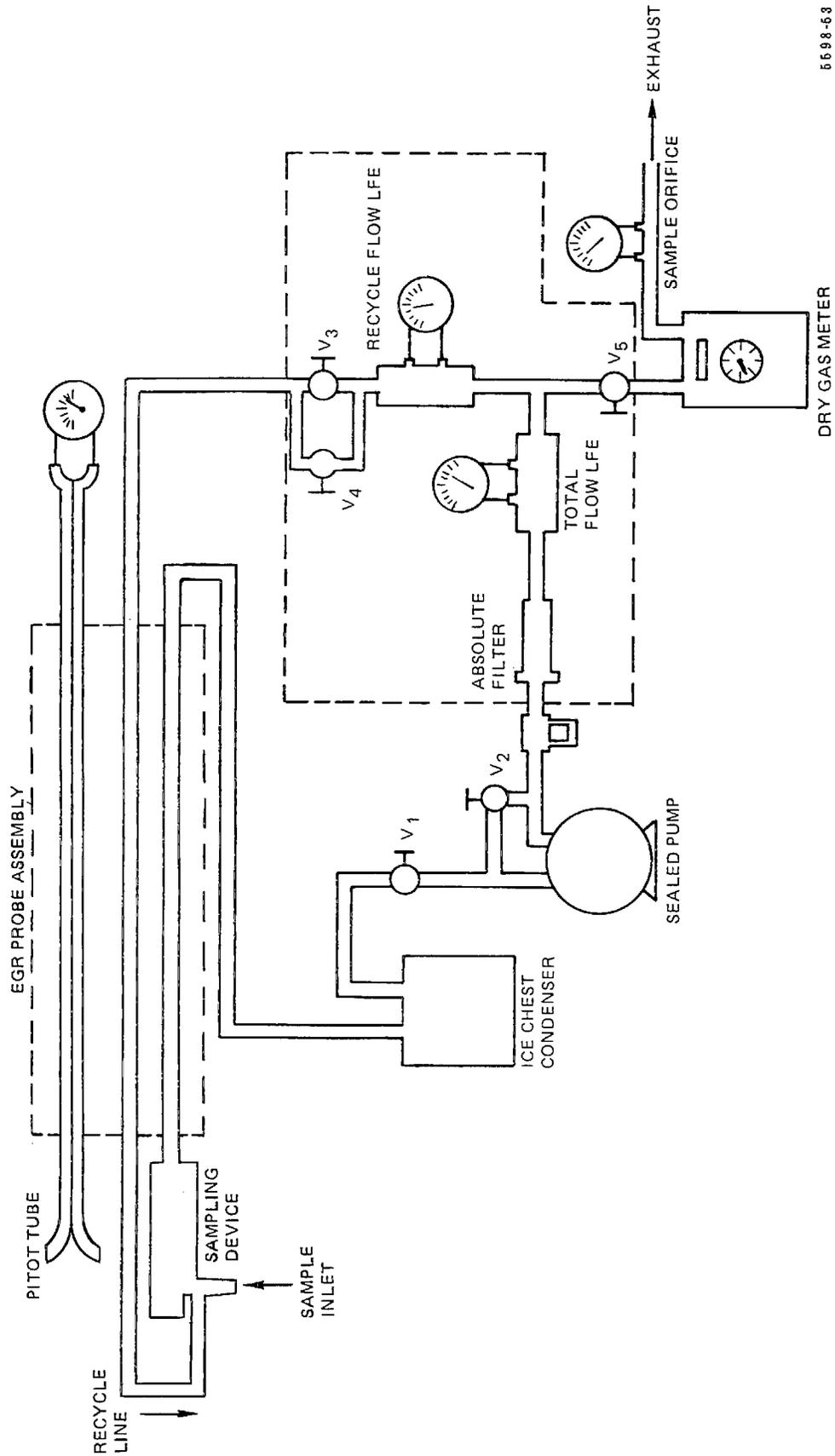
A block diagram of the prototype train is shown in Figure 6. The gas sample which contains particulate matter enters through the sample inlet of the mixing nozzle. Oversize particles are removed by the classifier, after which the desired particulate sample is collected on the sample filter. The gas then passes through an impinger train or ice-cooled condenser, followed by a sealed pump controlled by valves V_1 and V_2 for coarse and fine flow adjustment, respectively. From this point in a standard isokinetic sampling train the gas would pass directly to the dry gas meter and sample orifice and finally be exhausted. In the train as modified for EGR, after the gas exits the pump (sealed, oilless) it passes through an absolute (HEPA) filter and the first of two laminar flow elements where the total flow is measured. The gas stream is then split into the recycle and sample lines. The recycle gas flow is controlled by valves V_3 and V_4 , and measured by a second laminar flow element. The sample flow is monitored in the usual manner, using a dry gas meter and a calibrated orifice. Valve V_5 , at the inlet to the dry gas meter, was added to the system to extend the range of control to higher recycle percentages by adding back pressure to the sample flow line.

The PM_{10} size-fractioning cyclone used in conjunction with the EGR system is shown in Figure 7. The cyclone chosen as the PM_{10} sizing device was the



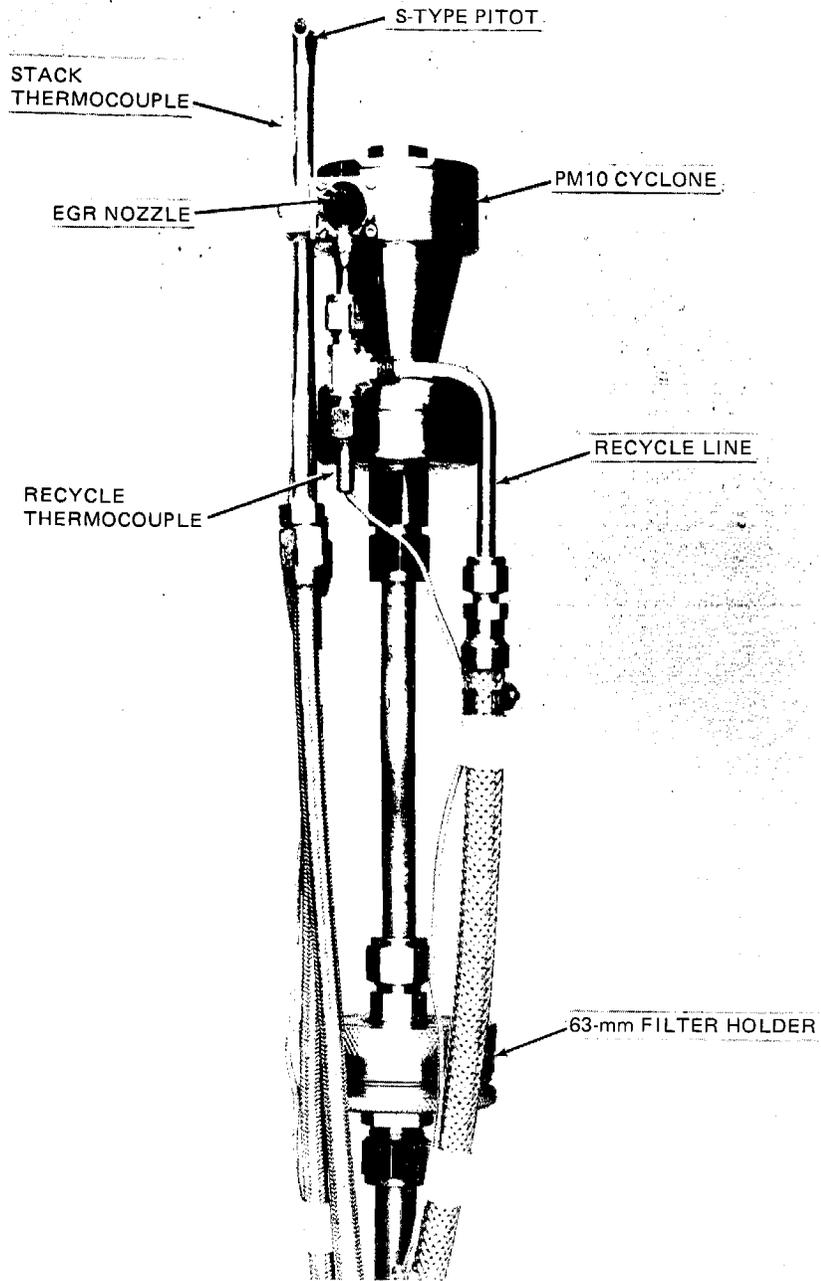
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Figure 5. Gas flow in emission gas recycle (EGR) train (Harris, et al., 1981).



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Figure 6. Schematic of the exhaust gas recirculation train (Williamson, et al., 1984).



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Figure 7. EGR-PM10 Cyclone Sampling Device (front view).

commercially available version of SoRI/EPA's Cyclone I (of the 5-Stage Series Cyclone). Potential problems which were considered in the design and selection of hardware for the EGR train, were deposition of particles smaller than 10 μ m upstream of the sizing device, bounce and reentrainment of particles larger than 10 μ m and an inaccuracy in the cutoff diameter due to errors in setting the total flowrate.

The EGR sampling system as described above consists of an in-stack classifier and filter combination. In this configuration, operation of the system is analogous to that of Method 17 used for total particulate emissions measurement. It should be noted, however, that the system could be assembled to operate in a geometry similar to that of Method 5 sampling with the PM₁₀ cyclone mounted directly onto the sampling probe and the filter mounted on the opposite end of the probe contained within an exterior oven. This mode would make it possible to maintain maximum similarity between the EGR sampling procedures and Method 5.

SAMPLING PROTOCOLS AND DATA ANALYSIS

Comprehensive documentation for each of the three methods is provided as attachments to the final report. A separate complete document has been prepared for each of these methods. Included in each document are discussions of the theoretical basis for the method, calibrations and other empirical proofs of performance, discussions of problem areas and interferences, complete operational protocols, and data reduction and analysis protocols. The data reduction techniques are described in detail for each method and numerical examples illustrating the data analysis procedures are provided. In addition, computer programs for carrying out all calculations that are needed in conjunction with both field sampling and data analysis were provided on floppy disk for the Apple II series of microcomputers. These programs are written in the BASIC computer language and can be adapted to other microcomputers. Documentation for the programs is included as an appendix to the procedures documents for each of the respective methods.



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