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Project Summary

Development of Sampling Methods for Source PM_{10} Emissions

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This report describes an investigation of the needs and available techniques for in-stack PM_{10} sampling. Discussion includes the conceptualization, development, documentation, and testing of two candidate methods. The first method, Constant Sampling Rate (CSR), is a procedural approach which adds particle size separation to sampling hardware that has been widely used in EPA Methods 5 and 17 but modifies the sampling protocol to accomplish the PM_{10} objectives. The second method, Exhaust Gas Recycle (EGR), is an equipment approach which accomplishes the PM_{10} objectives by using a modified sampling train to implement the concept of exhaust gas recirculation.

Six field studies indicated that these techniques were practical and compared well with one another and with more labor-intensive approaches. Laboratory investigations with monodisperse aerosols indicated that commonly used geometries for sampling nozzles could cause a decrease in the particle size cut of a closely coupled inertial sizing device. Nozzle geometries were also found which eliminated the observed shifts in particle size cut.

This Project Summary was developed by EPA's Atmospheric Research and Exposure Assessment Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

A size-specific PM_{10} ambient-air particulate standard has been promulgated by EPA. The Quality Assurance Division of the Atmospheric Research and Exposure Assessment Laboratory (AREAL) has initiated a research program to develop cost-effective source measurement techniques to support the PM_{10} standard. This report summarizes the source PM_{10} method development work performed at Southern Research Institute (SRI) under EPA Contracts 68-02-3118 and 68-02-3696 and EPA Cooperative Agreement CR-812274. Much of this material is described more fully in other reports, which are referenced in this report.

The extensive particle size sampling technology, developed as a result of research efforts associated with Inhalable Particulate (IP) matter from stationary sources and particulate control devices, provided valuable background information for the PM_{10} efforts. The technical difficulties in size-specific (PM10) particulate sampling are greater than, but similar to, those of total particulate sampling by EPA Reference Methods 5 or 17. In Methods 5 and 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. This type of bias is limited by specifying the minimum number of traverse points. Likewise, temporal variations due to process variations can also cause inaccurate or unrepresentative emission measurements. Thus, three traverses are required to limit this type of error. Another potential error in particulate measurements is duct/nozzle sampling bias. Unless the gas velocity entering the sampling nozzle (plug velocity) equals the local duct velocity, particulate matter will be selectively depleted or enriched in the sample gas stream because of inertial separation at the nozzle entrance. Percent isokinetic is limited to 100 ± 10% in Methods 5 and 17.

These potential errors are more difficult to control in PM_{10} sampling because of the additional requirement of aerodynamic size classification, which is achieved by inertial techniques involving aerodynamic drag on aerosol particles. Any errors in the inertial cutoff diameter, which is primarily determined by flow rate through the size separator, will lead to errors in the PM_{10} measurement by misclassification of particulate matter in the size range near 10 μm. Thus, this flow rate must be held constant. Without a sampling nozzle of continuously variable cross-sectional area, this requirement for a fixed flow rate precludes isokinetic sampling in the direct manner of Method 5 or 17.

Previous work on this problem led to the development of two candidate sampling methods, the Exhaust Gas Recycle (EGR) sampling train and the Constant Sampling Rate (CSR) traversing protocol. The EGR train maintains isokinetic flow of gas into the sampling nozzle and augments it with an adjustable amount of filtered, recycled stack gas upstream of the inertial sizing device. In this manner, the total flow through the EGR inertial sizing device is held to the constant value required for classification of particles larger and smaller than 10 μm. The CSR protocol is an alternate PM_{10} technique which uses existing sampling equipment without special gas recycle adaptations.

In the EGR train, stack gas is isokinetically extracted through the sample inlet portion of the EGR mixing nozzle into the size separation device of the sampling train. After passing the size separator and in-stack filter, the sample gas passes through the probe and condenser or impinger train and into the EGR flow control module. As in conventional Method 5 control modules, the gas flow rate entering the control module is controlled by coarse and fine control valves at the entrance of the sealed pump. At the exit of the pump and absolute filter, the total flow is measured with a laminar flow element. The gas stream is then split into the recycle and sample flow lines. The sample flow is monitored in the normal manner by using a dry gas meter and a calibrated orifice. The partitioning between sample and recycle gas is controlled by a third valve located in the recycle flow line. The recycle gas line, along with the sample and pitot lines, passes through the heated probe in which the recirculated gas is reheated to the duct temperature. Power to the heater is regulated by a proportional temperature controller with a thermocouple reference sensor located in the recycle gas stream.

The CSR is a procedural approach which simply adds a particle size separator (cyclones or cascade impactors) to the basic sampling train already in use. The objective of the protocol is to limit error due to anisokinetic sampling to the approximate range expected from the spatial and temporal variation of the emissions. Anisokinetic sampling bias is held within these limits in most sampling situations by performing a full duct traverse with a single nozzle. However, in the very unusual situation of large velocity variation within the sampling plane, the traverse may be synthesized from two or more partial traverses using a different nozzle for each partial traverse. Thus, the flow rate through the inertial sampler is held at the level required for a 10-μm size cut over the full traverse. The range of duct velocities over which a given sample nozzle may be used is such that the combination of nozzle inlet diameter and PM_{10} flow rate results in anisokinetic sampling errors less than ±20% for 10-μm particles. Since corresponding errors for particle sizes less than 10 μm are much smaller (decreasing proportionately to particle size squared) and since some of these errors are of opposite sign, actual anisokinetic sampling error for PM_{10} will be much less than 20% in magnitude.

It was decided that this program should primarily address utilization of a single-stage size separator. The largest cyclone (Cyclone I) of an existing five-series cyclone train was chosen. More detailed equipment descriptions and operating protocols for the EGR train and the CSR procedure are also given in the project report.

PM_{10} Field Studies

As a key part of the PM_{10} development program, six field studies were conducted at four emissions sources. In the course of these field tests, the two candidate methods were refined and tested, and PM_{10} measurements were performed at range of source conditions.

Since both PM_{10} methods hav hardware or procedural elements whic are different from other source samplir methods, the field studies were used a means of obtaining basic data abo these new procedures, as well a development, refinement, and validatic of the overall methods. The first objectiv of the field studies was to test and refin the procedures and sampling hardware o both PM_{10} methods. A second objectiv was to obtain comparison measurement of PM_{10} and total particulate concer tration by each method and by the bes available reference measurements. Th third objective was to measure the pre cision of each PM_{10} method at common source and compare these pre cision measurements to precision meas urements using Method 17.

To meet the objectives describe above, careful attention to test desig was required. While the detailed desig of each test varied according to the pri mary objective for the test and th specifics of the test site, certain element were common to several tests. These include use of independent measure ments as the best available reference or the accuracy of each technique, contro of external variables by maximum feas ible use of collocated and simultaneous sample protocols, and site selection fo significant challenge of the methods ove a range of source conditions.

Several conclusions may be drawn from the field data summarized in Table 1, as well as the more complete data sets given in the full project report. First, in every instance the average concentrations measured by different techniques agreed within the combined 95% confidence intervals. Since these intervals for some tests reflect a substantial degree of variation, presumably due mostly to source fluctuations, a more meaningful comparison can be drawn from paired-run analysis of the simultaneous measurements indicated in Table 1.

At both site 1 and site 4 in tests 1, 5, and 6, the EGR train measured less total particulate than Method 17 by a small but significant amount. Mean differences ranging from 5 to 13% were observed, and in each case these differences were larger than the 95% confidence limits. The reason for this small bias is not clear; however, since it does not exceed 15% for any of the sites tested, this bias is not considered detrimental. CSR total mass measurements at sites 1, 3, and 4 in test 2, 4, and 6 were not significantly different from the paired total mass measurements

Table 1. Percentage Differences in Particulate Concentrations Measured During Test Series[a]

Test Number	*Number of Replications*	*PM_{10}*	*Total Concentration*
1 EGR Initial Test: Site 1			
EGR Cyclone - Isokinetic Cyclone[b]	4	-8.3 ±27	9.0 ±29
EGR - Method 17[b]	8		-11.5 ±8.3
2 CSR Initial Test: Site 1			
CSR - Method 17[b]	3		-16 ±32
CSR - Isokinetic Impactors[b]	4	-1.8 ±22	-14.0 ±65
3 EGR/CSR Comparison Test: Site 2			
EGR Cyclone - CSR Cyclone[b]	5	-15.5 ±6.5	-9.2 ±8.5
EGR - Isokinetic Impactors	5-6[c]	-11 ±31	1.3 ±38
CSR - Isokinetic Impactors	7-6[c]	3.8 ±25	14 ±31
4 EGR/CSR Comparison Test: Site 3			
	Inlet		
EGR - CSR[b]	6	11 ±9.8	1.7 ±21
EGR - Impactor	6-5[c]	27 ±16	-9.8 ±16
CSR - Impactor	6-5[c]	16 ±16	-11 ±14
	Outlet		
CSR Impactor - Method 17	6-7[c]		-7.4 ±23
5 EGR Precision Test: Site 4			
EGR - Method 17[b]	6		-12.9 ±4.2
EGR - EGR_2[b]	6	-2.4 ±4.9	-0.9 ±4.3
6 CSR Precision Test: Site 4			
CSR - Method 17[b]	9		0.4 ±6.3
EGR - Method 17[b]	7		-4.8 ±1.7
EGR - CSR[b]	9	-15.8 ±7,8	
CSR_1 - CSR_2[b]	9	6.6 ±3.8	1.2 ±5.4

[a]All differences and confidence intervals expressed as percentages of the mean value. Confidence intervals represent 95% significant level.
[b]These comparisons were analyzed as pairs since the measurements were simultaneous.
[c]Where two numbers of replications are given, the first number corresponds to the first listed device and the second to the second device.

from Method 17 or other reference isokinetic sampling trains. Since the CSR technique is expected to be less accurate for total mass, these results are encouraging. When total mass data using the two techniques are compared, the results are mixed. At site 2 in test 3 the 9% EGR—CSR difference is marginally significant at the 95% confidence level. At sites 3 and 4 the EGR and CSR data are essentially the same.

The PM_{10} values measured by the two techniques differ at every site by more than 10% but less than 20%. At sites 2 and 4, the EGR PM_{10} value is about 15% less than the CSR value. At site 3, the EGR value is 11% greater than the CSR value. All three differences are statistically significant at the 95% confidence level. The results at site 3 reverse what would appear to be a trend at the other three sites for EGR PM_{10} values to be lower by about 5-15% than the CSR values, which are not significantly different from the individual isokinetic impactors. No clear reason was found for this test-to-test reversal.

At site 4, measurements with collocated pairs consisting of two EGR trains in test 5 and two CSR trains in test 6 indicated excellent reproducibility between the two trains. In only one instance of CSR PM_{10} concentrations does the mean difference in the measurements of two nominally identical trains exceed 2.5%, and even that low bias of 6.5% was found to be due to a systematic difference in cyclone flow rate between the two trains. For both PM_{10} trains, 95% confidence intervals were on the order of ±5%. By this measure, the precision of the PM_{10} trains was the same as that of the paired Method 17 trains operated during these tests.

Optimization of PM_{10} Cyclone and Sampling Nozzles

One further element in the testing and refinement of both candidate methods was the inertial sizing device itself. While the candidate PM_{10} cyclone had been used for several years in other applications, it had not been characterized either in the laboratory or field under conditions typical of PM_{10} operation. The versions of the cyclone which are commercially available have different exterior dimensions and nozzle designs from the SRI prototypes which were used on the initial studies. These differences prevented design of a single EGR nozzle system suitable for both versions of the cyclone. Several adaptations in both versions were necessary for use as a PM_{10} precollector for a single- or dual-stage sizing train. Prior to this work, it was also not clear how well a 10-µm cut could be predicted over a range of stack gas conditions with the cyclone, either in a gas recycle or a conventional nozzle configuration. During the test series, several of these potential difficulties were clarified or resolved.

Calibrations of Cyclone I were performed with a vibrating orifice aerosol generator (VOAG). The VOAG provides monodisperse dye aerosol of chosen particle size at a rate of about 60,000 particles/s. After lofting and drying, the aerosol is passed through the sampling train which includes an absolute filter. After the sampling run, all internal surfaces of the sampler and the filter are carefully washed with a measured volume of solvent. Spectrometry or fluorometry techniques are then used to determine the concentration of dye in the wash solutions and thus the collected aerosol mass for each surface and the mass captured by the backup filter. The dye particles utilized in this laboratory investigation were composed of dry ammonium fluorescein.

To simulate sampling from process streams, an apparatus for the calibration studies was designed that established a sample flow stream substantially larger than the diameter of the sample nozzles. The sample flow stream had a uniform velocity profile at (or near) the nozzle inlet and resulted in only minimum dilution of the VOAG aerosol. In addition, to understand better the effect of the nozzle geometry on the particle sizing performance of Cyclone I, a system was developed to obtain high-resolution velocity profiles at the cyclone inlet for each of the nozzle geometries calibrated. To correlate these data with the collection efficiency data, the velocity profile was measured at conditions which simulated each of the cyclone calibration conditions. The velocity sensing device used in the test section was a amall pitot made of two hypodermic needles (0.03-in. diameter) with beveled openings approximately 0.06 in. in length. For the purpose of traversing the test section in known increments, the pitot was mounted on a horizontal positioner attached to a vernier scale (reproducible to 0.001 in.).

In addition to the 1/2-in. nozzle, which has the largest sampling diameter and which was used as a reference, the other existing nozzles used for Cyclone I were classified into the following three types: tapered nonrecycle, large expansion nonrecycle, and recycle. Test results for Cyclone I collection, nozzle efficiency, and velocity profile were presented by nozzle type.

The tapered-nonrecycle nozzles have a small angle of expansion from the inlet diameter to the cyclone inlet diameter. The behavior of Cyclone I may be slightly different from that of the reference nozzle, but the cut size is changed by much less than 1 μm. Measurable nozzle losses did, however, occur. Losses in the larger nozzle increased with particle size from 3% (at 4 μm) to about 20% (at 10 μm). With the smaller nozzle, losses of about 22% were found, which decreased only slightly for the smaller particle sizes.

Cyclone I collection efficiency was measured for all nonrecycle large expansion nozzles, in which a large expansion angle within the nozzle is the sample aerosol pathway. The EGR nozzles with zero recycle air are included in this group since they present an abrupt expansion to the flow at the end of the nozzle sample tube. The cyclone cut diameter shifted down to about 6 μm with all of these nozzles. The highest nozzle deposition losses were incurred by the 0.138-in. nozzle, 30% at 4-um particle size, decreasing to 20% at 10 μm. The 0.155-in. nozzle had about a 10% loss at 4 μm and 15% at 10 μm. Nozzle optimization studies have minimized these problems.

Efficiencies were measured for recycle (EGR) nozzles at multiple recycle rates. In each instance, efficiency was higher (or cut size smaller) for the lower recycle rate. All three nozzle sizes caused cuts to vary from about 6 μm at 0% or 10% recycle to about 9 μm at 75% recycle. Nozzle losses with the 1/4-in. and 1/8-in. EGR nozzles were insignificant (at the <2% level) at all recycle rates studied. For the 1/8-in. EGR nozzle, losses were low (~3%) at the 75% recycle rate. At 0% and 10% recycle, the nozzle losses at the 4-μm particle size were about 20% and dropped to 2% for 10-μm particles.

Further measurements were performed to test approaches for eliminating the observed shift in cyclone cut size at the higher nozzle inlet velocities. The results obtained with the original nozzles indicated that the inertia of the higher velocity aerosol streams was not dissipated sufficiently to prevent additional impaction in the cyclone. Therefore, modified nozzles were tested which reduced the sample gas velocity prior to entering the cyclone. Two types of modified nonrecycle nozzles were tested. Both were extensions of nozzle lengths beyond the original nozzles of the same inlet diameter, one group having large expansion angles, ≥45°, and the other group having small tapered expansion angles of 7° and 14° (total included angle). One type of modification to the EGR nozzles was tested extensively. This was a simple extension of the nozzle length so that more distance was available for expansion.

The extended EGR nozzles gave cyclone behavior identical to the reference nozzle. However, these modified nozzles had substantially higher nozzle losses than the unextended EGR nozzles. The extended 1/8-in. EGR nozzle with expansion distance of 3.1 in. exhibited nozzle losses of 20% at the 4-μm particle size and 35% at 8 μm.

Nonrecycle 1/8-in. nozzles having large expansion angles and expansion distances greater than 2.2 in. improved cyclone efficiencies to those of the reference nozzle. The shorter of the two nozzles tested (2.2- and 3.1-in. expansion distance) exhibited lower nozzle losses at the 8-μm particle size, 36% compared to 47%.

The tapered-nonrecycle nozzles were compared to nozzles having large expansion angles and the same inlet diameter. The 0.16-in. nozzle with 7° angle eliminated the undesired effect on cyclone behavior caused by the original 0.154-in. nozzle that had an abrupt expansion angle and short length. The 0.16-in. nozzle with 7° angle had 5 to 10% higher nozzle losses, ranging from 10% at 4-μm particle size to 20% at 10 μm.

Cyclone behavior was not affected by the 1/8-in. nozzles with 7° or 14° tapers. Both of these had a total length of 3.2 in., i.e., the 14° nozzle had a straight section at its exit end. Nozzle losses for these two tapered nozzles were essentially the same. In contrast, the nozzle losses for the 1/8-in. nozzle with an expansion distance of 3.1 in. and an abrupt nozzle tube expansion were higher than losses for the two tapered nozzles of the same length, 11 and 13% higher at 4- and 8-μm particle sizes, respectively.

The laboratory data obtained in this PM_{10} program have major importance for PM_{10} methods in two ways. First, the data establish a basis for using Cyclone I as a PM_{10} size separator for a wide range of operating conditions. The efficiency curve for Cyclone I (D_{50} = 10 μm) has an acceptable geometric standard deviation of 1.4. Although the slope of the efficiency curve may decrease somewhat at elevated temperatures where Reynolds number is lower, it is expected to retain sufficient sharpness of cut to remain quite acceptable.

Second, the laboratory data establish the existence of, and point to a solution for, a significant effect of existing sampling nozzles upon cyclone behavior. It is expected that the solution found in this investigation could be optimized further. It is reasonable to assume that a similar nozzle effect occurs to some degree in all sizing devices used in process streams with high velocities. The observed effect of small nozzles on behavior was a shift in cut point from 10 µm to as low as 6 µm, the shift generally decreasing as nozzle inlet velocity decreased. If left uncorrected, this effect would cause measured PM_{10} to be lower than actual concentrations to a degree which depends upon the aerosol size distribution. The cause of the shift in D_{50} associated with some nozzles was attributed to high inertia associated with high-velocity gas streams. The shift in D_{50} was found throughout the data to correlate closely with the velocity of the gas entering the nozzle.

The effect of sampling nozzle on cyclone behavior and, hence, measured PM_{10} can be eliminated by causing the sample gas to decelerate after entering the nozzle and before entering the cyclone. In this present work, extending the nozzle inlet farther from the cyclone inlet regained the basic cyclone performance. However, nozzle losses were enhanced. The lowest losses occurred for tapered nozzles with expansion angles of 7° or 14°. These differences in losses between nozzle geometries were probably caused by flow separation accompanied by a region of flow recirculation with the larger expansion angles.

The analytical results based on these laboratory data show clearly that for further optimization studies the nonrecycle and recycle nozzles for Cyclone I should be redesigned with a smooth taper from the nozzle inlet diameter to the 0.5-in. diameter of the cyclone inlet. It appears that modifications of the EGR nozzle should also include modifying the recycle gas flow so that the recycle gas will have a higher average velocity but more uniform velocity profile. The data obtained thus far indicate that nozzle losses for these improved nozzle geometries will be significant for small inlet diameters or high stream velocities. Average losses for particulate diameters within the range studied here of 4 to 10 µm would be about 1% at 5 ft/s, to approximately 13% between 30 and 60 ft/s, and 30% at 88 ft/s. The velocity values relate to this laboratory study in which PM_{10} flow rate (for Cyclone I) was 0.45 acfm. The PM_{10} flow rate and the corresponding nozzle velocities are typically 20 to 30% higher in field measurements.

Conclusions and Recommendations

Six field studies have been performed to develop and characterize the methods. As measured by a modified dual-probe technique, the precision of each method is better than ±5%, comparable to that of EPA Method 17 at the same location. Comparability of the EGR and CSR techniques is within 16% at all sites tested. The EGR measured lower PM_{10} concentrations than CSR and other reference samplers at two sites, and higher than both at a third. All of these differences were statistically significant at the 95% confidence level.

Laboratory studies in this program indicate that decreases in particle size cut can occur for inertial sizing devices when the sampling nozzle has a small inlet diameter and is closely coupled with the inertial separation stage. Such shifts were observed to occur in Cyclone I, the current PM_{10} sizing device, which was tested with several of the current nozzles. Shifts were observed in particular with the three EGR nozzles and those nonrecycle nozzles which had an abrupt expansion within a short distance from the cyclone body. It is projected that this effect probably occurs in other available inertial samplers in this size range.

Optimization studies for sampling nozzles for Cyclone I indicate that the shifts in cyclone collection efficiency can be eliminated by lengthening the expansion zone in the nozzle. This lengthening, however, increases particle deposition in the nozzle. Nozzle losses averaged over particle sizes of 4 to 10 µm were observed to range from about 1% at low velocity, to near 13% at medium velocity, to 30% at high velocity. Although further research should be directed at the nozzle effects problem, the methodologies in their present form are usable with acceptable relative accuracy and precision for a wide range of sampling situations.

In view of these conclusions, the highest priority recommendation for further research is a more thorough design and characterization study to optimize the nozzles for use in a PM_{10} sampling method, in particular the EGR nozzles. While both EGR and CSR are usable in their present form with no modifications other than simple extensions of the smallest nozzles, the current methods appear likely to show a slight negative bias in measured PM_{10}, which increases with increasing duct velocity. Nozzle optimization and detailed specifications on nozzle design will probably be useful for measurement of PM_{10} at very high duct velocities.

Recommendations for further research of a somewhat lower priority can also be made. Reduction of approximate setup calculations for both methods to a form suitable for programmable calculators should be attempted. Extension of performance data of the PM_{10} sampling procedures to source conditions beyond the range currently studied is desired. Further field studies are suggested also to test any new nozzles from the recommended design study. Finally, extensive field studies are recommended to extend the number of source types tested by these methods.

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The complete report, entitled "Development of Sampling Methods for Source PM_{10} Emissions," (Order No. PB 89-190 375/AS; Cost: $21.95, subject to change) will be available only from:
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