Condensible Particulate Matter

Regulatory History and Proposed Policy

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Michael Pjetraj NC DAQ Stationary Source Compliance Branch Particulate matter is the most widely regulated air pollutant emitted from industrial sources. Health concerns regarding particulate matter concentrate on "fine" particulate matter - particulate matter less than 2.5 microns (PM2.5). Particulate matter exists in the solid and liquid physical states, and gases or vapors may also condense to form particulate matter. The latter, condensible particulate matter, is of great concern due to the inherently small size of condensation products; overwhelmingly, condensible particulate can be classified as PM2.5.

Federal Regulatory History:

When the EPA was mandated to develop emission standards for industries generating significant amounts of pollution, the EPA identified particulate matter as one of the regulated pollutants. In August of 1971, the EPA proposed emission standards for five source categories: Fossil Fuel Fired Steam Generators, Incinerators, Portland Cement Plants, Nitric Acid Plants, and Sulfuric Acid Plants. Limits for particulate matter emissions were proposed for three of the five initial New Source Performance Standard (NSPS) subparts and the test method to demonstrate compliance for these sources was also proposed.

The NSPS limits were developed from test data at multiple representative facilities. The initial tests were performed using a particulate method that would quantify the filterable and condensible particulate matter separately. The summation of these two values was then termed the total particulate catch. When the initial particulate limits were proposed, the limit was based on total particulate data. During the rule making comment period, there was dissent regarding the use of the condensible catch. One primary dissenting argument was that the SO₂ emissions would be quantified in the condensible fraction of the sampling train, thus double counting the SO₂ emissions. Also, arguments were presented that condensible emissions were a non-predictable phenomenon, and therefore, could not be predictably controlled. In December of 1971, the EPA promulgated the initial NSPS limits for particulate and the corresponding test method.

The promulgated Method 5 particulate matter test method omitted the analysis of the condensible catch and measured only the filterable particulate. To compensate for the exclusion of condensible particulate matter, the promulgated NSPS standards for particulate matter emissions from steam generators and incinerators were decreased.

Federal Register (FR) 12/23/71: Preamble to the Promulgated NSPS & Method 5

"Particulate matter performance testing procedures have been revised to eliminate the requirement for impingers in the sampling train. Compliance will be based only on material collected in the dry filter and the probe preceding the filter. Emission limits have been adjusted as appropriate to reflect the change in test methods. The adjusted standards require the same degree of particulate control as the originally proposed standards."

FR 3/21/72: Supplemental Statement in Connection with Final Promulgation:

"There has been only limited sampling with the full EPA train such that the occasional anomalies cannot be explained fully at this time. Accordingly, we determined that, for the three affected source categories, steam generators, incinerators, and cement plants, particulate standards should be based on the front half of the EPA sampling train with mass emission limits adjusted as follows:

	Originally proposed particulate standards, full EPA train	Recommended particulate standards revised sample method (front half only)
Steam Generators (Ib/mmBtu)	0.20	0.10
Incinerators (gr/dscf at 12% CO2)	0.10	0.08
Cement Kilns (lbs / ton feed)	0.30	0.30
Cement Coolers (lbs / ton feed)	0.10	0.10

The definition of particulate matter, pertaining to the compliance status of NSPS facilities, was any particulate matter caught by the Method 5 particulate test. Hence the method defined the pollutant. It is important to note that since the method defined the pollutant, the method's results also defined the NSPS emission standard.

Over the following years, the regulatory inclusion of condensible particulate matter was often discussed. Although condensible particulate matter was not included in the NSPS limits, a test method was eventually promulgated in 1991 that quantifies condensible particulate matter. (40 CFR 51, Appendix M, Method 202)

The NSPS regulations were designed to demonstrate the most effective control technology available at the time of subpart promulgation. Arguments have been made over time claiming that the control technology existing at the time of promulgation could not effectively control condensibles. If the EPA defined the pollutant by the test method and determined that condensibles would not be indicative of the efficiency of a control device, then the NSPS definition of particulate should exclude condensibles. The argument makes sense when a control efficiency is the end goal of the regulation or when the final emission limit is based on the effective control that a representative device can deliver. Since the NSPS limit was based on the post control emission rate, the measurement of a pollutant that was not controlled would be of no benefit.

FR 10/6/75: Preamble to the Promulgated Changes of the NSPS Subparts

"Several factors are of primary importance in developing the database for a standard of performance and in specifying the reference method for use in conducting a performance test, including:

a. The method used for data gathering to establish a standard must be the same as, or must have a known relationship to, the method subsequently established as the reference method. <u>The method should measure pollutant emissions indicative of the performance of the best systems of emission reduction</u>. <u>A method meeting this criterion will not necessarily measure emissions as they would exist after dilution and cooling to ambient temperature and pressure, as would occur upon release to the atmosphere.</u> As such, an emission factor obtained through the use of such a method would, for example, not necessarily be of use in an ambient dispersion model. This seeming inconsistency results from the fact that standards of performance are intended to result in installation of systems of emission reduction which are consistent with best demonstrated technology, considering cost. The Administrator, in establishing such

standards, is required to identify best demonstrated technology and to develop standards which reflect such technology. In order for these standards to be meaningful, and for the required control technology to be predictable, the compliance method must measure emissions which are indicative of the performance of such systems"...... "Subsequent to the promulgation of the standards of performance for steam generators, data became available indicating that certain combustion products which do not exist as particulate matter at the elevated temperatures existing in steam generating stacks may be collected by Method 5 at lower temperatures (below 160 C). Such material, existing in gaseous form at stack temperature would not be controllable by emission reduction systems involving electrostatic precipitators. Consequently, measurement of such condensible matter would not be indicative of the control system performance....... Nevertheless, to insure that an unusual case will not occur where a high concentration of condensible material, not controllable with an ESP, would prevent attainment of the particulate standard, the sampling temperature allowed at fossil-fuel steam boilers is being raised to 160 C."... "It is believed that this increase resulted in large part, if not totally, from SO3 condensation which would occur also when the stack emissions are released into the atmosphere. Therefore, where standards are based on emission reduction to achieve ambient air quality standards rather than on control technology (as is the case with the standards promulgated herein) a lower sampling temperature may be appropriate."

Early EPA statements indicate that the emission rates of condensibles vary with the emissions of filterable particulate. The degree of correlation was not stated, but the statement implies that there is some reduction benefit for the condensible emission rate from the typical control device. Obviously some processes strive to prevent condensation within the effluent in order to protect equipment. Thus, whenever the effluent is at an elevated temperature, there is the potential for condensation of some particulate matter when the effluent temperature decreases.

FR 3/21/72: Supplemental Statement in Connection with Final Promulgation:

"It is the opinion of EPA engineers that particulate standards based either on the front half or the full EPA sampling train will require the same degree of control if appropriate limits are applied. Analyses by EPA show that the material collected in the impingers of the sampling train is usually although not in every case a consistent fraction of the total particulate loading."

"Command and control" regulations are designed to reduce the emissions of pollutants. The reduction potential is determined by the control device deemed appropriate based on environmental and economic impact. However, the issue of generic control may be over ridden when ambient air quality standards are of concern. When sources and regulatory agencies are concerned with the effect of pollution on ambient air quality, source level measurements should include those pollutants that directly affect the ambient air quality.

FR 2/23/78: Preamble to the Promulgation of Subpart BB

"It should be noted that standards of performance for new sources established under section 111 of the Clean Air Act reflect emission limits achievable with the best adequately demonstrated technological system of continuous emission reduction considering the cost of achieving such emission reductions and any non-air quality health, environmental, and energy impacts. <u>State Implementation Plans (SIP's)</u> approved or promulgated under section 110 of the Act, on the other hand, <u>must provide for the attainment</u> and maintenance of national ambient air quality standards (NAAQS) designed to protect public health and welfare. For that purpose, SIP's must in some cases require greater emission reductions than those required by standards of performance for new sources."

An excellent example of two different types of particulate sampling trains measuring different types of particles can be found in the following Federal Register preamble. The Los Angeles Air Pollution

Control District particulate matter test method (LAAPCD Method) was an older test procedure that measured both filterable and condensible particulate matter. However, Method 5 data was again used as a bench mark to compare and determine control effectiveness from source to source during the development of this standard. The measurement of particulate matter via Method 5 does not necessarily indicate the total particulate loading attributable to a source.

FR 6/15/79: Preamble to Proposed Subpart CC-Standards of Performance for Glass Manufacturing Plants:

'Selection of Performance Test Methods: The use of EPA Reference Method 5 - Determination of Particulate Emissions from Stationary Sources' (Appendix A, 40 CFR 60, Federal Register, December 23, 1971) is required to determine compliance with the mass standards for particulate matter emissions. Emission test data used in the development of the proposed standard were obtained either by the LAAPCD sampling method or by EPA Method 5. However, results of performance tests using Method 5 conducted by EPA on existing glass melting furnaces comprise a major portion of the database used in the development of the proposed standard. EPA reference Method 5 has been shown to provide a representative measurement of particulate matter emissions. Therefore, it has been included for determining compliance with the proposed standards."......."This fabric filter installation was tested with the Los Angeles Air Pollution Control District particulate matter test method (LAAPCD Method), which considers the combined weight of the particulate matter collected in water-filled impingers and of that collected on a filter. EPA Method 5 also uses impingers and a filter, but considers only the weight of the particulate matter test a larger amount of particulate matter than does EPA Method 5, and, consequently, greater mass emissions would be reported for comparable tests."

The following preamble discusses the "fine" particulate concern with respect to this source type. There is also a discussion on the condensation of sulfuric acid in the sampling train and measurement as a particle.

FR 10/7/80: Preamble to Promulgated Subpart CC Standards of Performance for Glass Manufacturing Plants:

"With regard to the public's health and welfare, <u>the submicron size of most glass furnace generated</u> <u>particulates</u>, among other factors, is particularly important." "These particulates also have fairly long lives in the atmosphere and can absorb toxic gases, thus leading to potentially severe synergistic effects when inhaled."..... "Test Methods and monitoring: Some commenters stated that EPA Method 5, "Determination of Particulate Emissions from Stationary Sources" contains several sources of error when used to sample emissions from soda-lime glass melting furnaces. They stated that misclassification of particulate and gaseous species and inflated particulate emission values are errors which can be caused by the use of filter temperatures below the Sulfur Trioxide (SO3) dew point. When particulate matter is filtered at about 120 C, a significant amount of sulfuric acid, if present, can condense on the filter. <u>The measurement of this sulfuric acid by Method 5 does not constitute an error in the method because sulfuric acid is normally considered to be particulate matter.</u> However, the variability of the sulfuric acid content in the stack gas was not considered in developing the standards. As a result, the decision was made not to include sulfuric acid as part of these standards. Therefore the method was modified to allow operation of the filter and the probe at up; to 177 C, which is above the acid dew point and would prevent sulfuric acid mist from being collected by the filter."

Since the effects of sulfuric acid were not considered when the standard was developed, the EPA determined that an increase in the filter temperature was prudent. The preamble does not discuss an important point though. If the standard was derived from data when the sampling train was operated

at a lower temperature, increasing the filter temperature that can be used for compliance demonstrations may result in a relaxation of the standard.

When the EPA determined that a control device for a specific source category could control condensible particulate matter, the EPA created a method to determine the control efficiency of both filterable and condensible particulate matter. The method mentioned in the following preamble is different from other condensible methods in that it measures the total organic compound (TOC) concentration in the impingers. Other condensible methods measure the condensible catch gravimetrically upon desiccation.

<u>FR - 2/7/84: Preamble to the Proposed Subpart PPP Standards of Performance for New Stationary</u> Sources; Wool Fiberglass Insulation Manufacturing.

"The test method for determining compliance with the proposed standard would be the proposed Reference Method 5E for particulate emission. Proposed Reference Method 5E is a variation of Reference Method 5 that determines the amount of material caught in the front-half (nozzle, probe, and filter) of an EPA Reference Method 5 sampling train plus the amount of condensed particulate material caught in the impinger train as determined by measuring total organic carbon (TOC) using a TOC analyzer. Reference Method 5E is proposed because it reflects the performance of BDT [Best Demonstrated Technology] for this industry more accurately than does Reference Method 5." "It was known that some fraction of the particulate emissions results from condensation of organic compounds used in the binder. Therefore in evaluating emissions and control device performance, a sampling method was used that could collect and measure both solid particles and condensed particulate material. Test data show that 10 to 47 percent of the particulate emissions from wool fiberglass facilities are condensed particulate material. Test data also show that emissions of both solid particles and condensible particulate material can be effectively reduced by the use of certain control techniques. Therefore, the proposed standard would limit particulate emissions based on the measurement of both solid particles and condensible particulate material" "Total particulate catch (filter catch plus condensible fraction) is recommended as the test method because it best assesses performance that reflects the use of BDT for this industry. Wet control devices can effectively reduce the levels of solid and condensible particulate matter present in the emission stream from wool fiberglass insulation manufacturing facilities. However, because of the high moisture content of the exhaust gas stream from wet control devices, the sample gas stream must be heated to a temperature considerably higher than that of the exhaust gas stream to vaporizer the water droplets to prevent clogging in the front half of the sampling train. This heating may also vaporize compounds that are aerosols at the stack gas temperature. These vaporized compounds are then cooled to a temperature below that of the exhaust gas stream and are condensed and collected in the impinger train."

When NSPS Subpart PPP was proposed, a commenter reviewed historic test data and State emission limits for the source type and suggested that the proposed NSPS standard was not accurate. Based on a review of the data the commenter believed that the EPA had erred in their calculations. The actual problem lay in the fact that the proposed NSPS standard was coupled with the proposed method for compliance from these facilities. The proposed method (Method 5e) measures filterable and the TOC portion of the impinger catch. Some of the reviewed data and the State limits were derived from Method 5 "front half" only testing. Therefore, Method 5e results would differ from Method 5 results.

<u>FR - 2/25/85: Preamble to the Promulgated Subpart PPP Standards of Performance for New Stationary</u> Sources; Wool Fiberglass Insulation Manufacturing.

"Determining baseline emissions from wool fiberglass insulation manufacturing plants directly from the State regulations is inappropriate because a different test method was used in developing the standard than was used to determine allowable SIP emission limits. This modified test method is the reference method for determining compliance with the standard of performance for wool fiberglass insulation manufacturing plants. Most state regulations are based on a "front-half catch" test method and the standard is based on a test method that measures total catch. The total catch test method was developed and proposed along with the standard to account for certain factors that are peculiar to the wool fiberglass insulation manufacturing industry."

North Carolina Regulatory History

The North Carolina Administrative Code (NCAC) Title 15A, Subchapter 2D contains the regulations for Air Pollution Control Requirements. The subchapter contains, among other information, the definitions of pollutants, references to the applicable compliance test methods, and emission limitations for stationary sources.

Section .0501 defines the test procedures that shall be used to prove compliance with the limits contained within the rest of the subchapter. The following excerpt pertains to particulate matter:

.0501 COMPLIANCE WITH EMISSION CONTROL STANDARDS

(3) Sampling procedures for determining compliance with particulate emission control standards shall be in accordance with Method 5 of Appendix A of 40 CFR Part 60. Method 17 of Appendix A of 40 CFR Part 60 may be used instead of Method 5 provided that the stack gas temperature does not exceed 320 F. The minimum time per test point for particulate testing shall be two minutes and the minimum time per test run shall be one hour. The sample gas drawn during each test run shall be at least 30 cubic feet. <u>A number of sources are known to emit organic material (oil, pitch, plasticizers, etc.) which exist as finely divided liquid droplets at ambient conditions. These materials cannot be satisfactorily collected by means of the above Method 5. In these cases the Commission may require the use of Method 5 as proposed on August 17, 1971, in the Federal Register, Volume 36, Number 159.</u>

The line referring to Method 5 proposed on August 17, 1971 (M5'71) was specifically incorporated into this part of the regulation in order to allow for the measurement of condensible particulate emissions. There has been some debate revolving around the frequency and under what circumstances the NC DAQ should require the use of M5'71. It has been suggested that the use of M5'71 should have been the default test procedure since it would measure and quantify the filterable and the condensible particulate separately. Over time, the "need" for quantifying condensible particulate matter shifted from a scientific basis to an aesthetic basis. Essentially, condensibles were not deemed a problem from a process unless the visible emissions were high enough to warrant complaints and Notice of Violations (NOVs) for opacity exceedances. Although a process could have significant condensible emissions, the requirement to test for condensibles was issued infrequently. Thus, the default became testing for condensibles only when there was a problem with the visible emissions. It should be noted that extreme visible emissions may be indicative of condensible particulate. However, the absence of extreme visible emissions does not necessarily indicate the absence of condensible emissions.

Another factor that must be considered in any determination of this kind is that atmospheric pollution is a relatively new science. When the original NSPS were written, there was a concern for total particulate emissions. The specific regulation of PM10 and condensible emissions, as well as test methods, were not addressed until years later. Now, the air quality science is focusing on deleterious chronic health effects due to PM2.5. When we gain new knowledge about a pollution phenomenon, the information should be applied. Since condensible emissions are primarily within the PM2.5 size range, it would be prudent to require condensible measurements.

Part of the history of particulate matter regulation in North Carolina is the definitions of particulate matter in 15A NCAC 2D .0501. None of the definitions for particulate matter specifically exempts condensible particulate matter. The definitions state that the pollutant shall be measured "*by methods*"

specified in this Subchapter". M5'71 is specified as a method in the subchapter.

Particulate definition			
<u>NCAC 0101</u>	DEFIN	<u>DEFINITIONS</u>	
	(23)	"Particulate matter" means any material except uncombined water that exists in a finely divided form as a liquid or solid at standard conditions.	
	(24)	"Particulate matter emissions" means all finely divided solid or liquid material, other than uncombined water, emitted to the ambient air as measured by methods specified in this Subchapter.	
	(27)	"PM10" means particulate matter with an aerodynamic diameter less than or equal to a nominal 10 micrometers as measured by methods specified in this Subchapter.	
	(28)	"PM10 emissions" means finely divided solid or liquid material, with an aerodynamic diameter less than or equal to a nominal 10 micrometers emitted to the ambient air as measured by methods specified in this Subchapter.	

An appropriate review of the NCAC emission standards should also occur. The standards in 2D .0500 were adopted from other agencies rather than testing at facilities within North Carolina. The exact origins of some of the limits is not clear. However, the intention of the State Implementation Plan is to ensure attainment of the NAAQS. Therefore, the limits should be linked to an acceptable ambient pollutant loading.

Memo from Thom Allen 12/97:

"As stated in the January, 1972 edition of the air quality rules, the purpose of this Rule and other particulate rules is ...to establish maximum limits on the rate of emissions of air contaminants into the atmosphere. All sources shall be provided with the maximum feasible control. (Rule 1.00, Purpose, Section IV, Emission Control Standards) This Rule follows the Air Control Advisory Council recommendation on emission control standards for suspended particulates and sulfur oxides: The emission control standards for all sources be based on the principle of providing the maximum feasible control. (Report and Recommendations of Air Control Advisory Council on Emission Control Standards for Suspended Particulates and Sulfur Oxides October, 1970, p.6.)"

Specifically for 2D .0515, the limits were derived from Los Angeles Air Control District regulations.

The limits were discussed by Mr. Kennedy of du Pont on 12/9/70: "The genesis of this California regulation is described by Dr. McCabe in <u>Industrial and Engineering</u> <u>Chemistry</u> in 1949. He was the Los Angeles Air Control Director. It was generated to control the metallurgical industry only and the emission limits were chosen on the basis of detailed study of the technical and economic feasibility of those operations only."

The 2D .0515 rule is basically a "catch all" for any sources that do not fall under a specific regulation. The rule was derived for a specific industry and applied to a broad group since the emission limits, dependant on process rates, are relatively large. Thom Allen's memo answers a question regarding the inclusion of condensible particulate matter during compliance demonstrations with the 2D .0500 rules:

Memo from Thom Allen 12/97:

"Conclusion. A review of the extant ancient records does not give a specific answer to the question

whether or not condensible particulates are included in along with the noncondensible particulates when determining compliance with 15A NCAC 2D .0515. However, the rules do suggest that condensible particulates are included because the test methods include testing procedures for condensibles (the Federal Register reference contained in the rule) and mention the emissions of organic materials that exist as liquid droplets at ambient conditions."

The final analysis of the history of condensible particulate emissions will need to answer what we *should* regulate. Throughout the early history of the Clean Air Act, the emphasis of source testing dealt with compliance demonstrations for NSPS sources. Particulate was defined by the Method 5 sampling train and therefore the emissions measured from the Method 5 train was then considered the absolute particulate value. Since particulate matter is temperature dependant, Method 5 offers a standard for defining and measuring particles. When the initial promulgation of the NSPS rules was concerned about the reduction of particulate matter emissions, a standard measurement was necessary to determine across the board reduction and compliance with command and control regulations.

Over time, the regulatory emphasis for particulate control has shifted to fine particles. PM10 emissions standards were introduced and newly promulgated PM2.5 ambient standards indicate the particle size of greatest concern. The historical testing using EPA Method 5 will capture filterable particulate down to approximately 0.3 microns. The EPA has stated that condensibles are a significant portion of PM10 emissions and therefore should be quantified for emissions inventories purposes.

PM-10 Emission Inventory Requirements, Final Report 9/94

"2.1.2 Condensible PM-10

Condensible particulate matter (or condensed particulate matter, as it is synonymously described) can be broadly defined as material that is not particulate matter at stack conditions but which condenses and/or reacts (upon cooling and dilution in the ambient air) to form particulate matter immediately after discharge from the stack. Condensible particle matter forms in a few seconds in the stack exhaust due primarily to immediate cooling and air dilution. Condensible particulate matter is of potential importance because it usually is quite fine and thus falls primarily within the PM-10 fraction. <u>As a consequence, condensible particulate matter should always be included in the emission inventory</u>.

2.1.3 Secondary PM-10 (PM-10 Precursors)

Secondary particulate matter can be broadly defined as particles that form through chemical reactions in the ambient air well after dilution and condensation have occurred (i.e., usually at some distance downwind from the emission point). An example of this phenomenon is the formation of sulfate particles in a plume from the oxidation of sulfur dioxide by one of several atmospheric transformation mechanisms. Generally, secondary particulate matter can be distinguished from condensible particulate matter by the time and/or distance downwind from the stack required for formation. Precursor emissions contributing to secondary particulate matter should not be included in the PM-10 inventory except where EPA and the State determines that the sources of PM-10 precursors contribute significantly to PM-10 levels which exceed the PM-10 NAAQS in the area. This determination will be based upon air quality analysis in which States assess the contribution of precursors.

If precursors contribute significantly to nonattainment, States will need to consider both the source-receptor relationship and the significance of precursor contributions to overall nonattainment. In making a determination regarding significance, EPA will rely in part on the technical information contained in the State's submittal which could include filter analyses, an assessment of the relative contribution of precursors to overall nonattainment, and the State's Reasonable Available Control Technologies (RACT)/Reasonably Available Control Measures (RACM) strategy. States are encouraged to submit additional material for consideration, since all findings will be made on a case-by-case basis."

If we are now concerned with those particles that can contribute to ambient PM10 and PM2.5 loading, we will need to gather condensible particulate matter data.

Condensible Methods

The effect of temperature on the state of particulate matter is important in understanding the methods of regulating particulate. Standards that relate to the control technology define particulate based on the performance of the device and the compliance method. Other regulations that are more concerned with the overall environmental impact and ambient loading of particulate matter may require a more encompassing definition of particulate matter.

Particulate matter is dependent on the temperature of a given effluent. As the effluent temperature changes, the physical state of the particulate constituents may change as well. Therefore, it is difficult to define particulate matter without a reference temperature. The current Method 5 particulate matter test defines particulate as any material that condenses at or above 248 degrees F. This temperature is set so that water vapor will not condense on the filter and clog the sampling train. (There is an allowable variation of 25 degrees and specific NSPS regulations may define particulate matter at a different temperature.) This definition begets the questions: What about particles that condense below 248 degrees? Since ambient temperature is always below 248 degrees, will "other" particulate matter condense and contribute to ambient particulate loading?

<u>Development of Methodology to Measure Condensible Emissions from Stationary Sources</u> "The change in the basis of the ambient air particulate concentration standard from total particulate concentration to PM10 may result in the need for updating source inventory information to include PM10 emissions. Because condensibles are likely to represent a greater fraction of the primary emissions in terms of PM10 than was the case for total emissions, the need arises for developing a source test method which includes the condensibles component of the emissions."

The test method for measuring condensible particulate that we have discussed is the 8/17/71 proposed EPA Method 5 sampling train (M5'71). There have been other suggestions for the measurement of condensible emissions including the use of a dilution apparatus. In 1990, when the PM10 test method was promulgated, the measurement of condensibles was addressed:

Fr- 4/17/90: Preamble to Promulgated change to 40 CFR 51

"Because of the need for a source test method for PM10, the Agency is adding two PM10 methods, Method 201 and 201A, for the measurement of in-stack PM10 emissions from stationary sources, to appendix M, 40 CFR part 51. Appendix M is a newly designated repository for recommended test methods for SIP's. The agency also is revising subpart K, 40 CFR part 51, to direct states to appendix M and to reiterate the fact that each SIP must include enforceable test methods with each emission limit in the SIP, including PM10. The PM10 source test method shall be Method 201, 201A, or an acceptable alternative. If the state intends to require the measurement of condensible emissions, then an enforceable method for the measurement of condensible emissions shall also be included in the SIP. As promulgated, the methods measure only in-stack PM10. <u>However, the EPA recognizes that condensible emissions are also PM10, and that emissions that contribute to ambient PM10 concentrations are the sum of in-stack PM10, as measured by Method 201 or 201A, and condensible emissions. Therefore, for establishing source contributions to ambient concentrations of PM10 for emission inventory purposes,</u> EPA recommends that source PM10 measurements include both in-stack PM10 and condensible emissions."

Method 202 was promulgated in 1991. It was created as a method to quantify the condensible particles that contribute to ambient PM10 loading. The method describes the analytical measurement of condensibles after testing has been performed. The actual testing procedure entails performing an EPA Method 5, 17 or 201 particulate measurement test. In order to quantify condensibles, the impinger catch from the sampling train of the method used is analyzed. The analysis includes dividing the catch into an organic extractable and an "inorganic" portion. The extracts are then desiccated and the residual particulate is gravimetrically measured. An important addition to Method 202 that was not in M5'71 is the nitrogen purge procedure. Due to concern of SO₂ oxidizing to sulfates in the impingers and then being counted as a particulate, the impingers are purged with nitrogen in order to remove the SO₂, prior to sulfate formation. By purging the SO₂, quantifying the SO₂ as particulate should not occur. However, other sulfate species should be quantified in the analysis.

FR 12/17/91 - Preamble to the Promulgated Method 202

"One commenter suggests that EPA determine the chemical composition of the material collected in the sampling train to verify that it will form ambient condensibles. The EPA believes that material will collect in the impingers only by condensation or dissolution. Dissolved gases will evaporate during analysis and will not be measured unless the gases react to form a solid or liquid while they are in solution. The EPA has designed Method 202 to prevent the formation of reaction materials from dissolved gases. The EPA believes that any remaining material collected and measured by Method 202 represents the material that would condense in the ambient air. Additional analysis of chemical composition is not necessary." "Another comment raises the concern that the method may collect some portion of the sulfur dioxide (SO_2) as condensible. The dissolution of SO₂ in water does not lead immediately to the formation of sulfuric acid (H2SO4), but tends to lower the solution pH, which further inhibits sulfate or H2SO4 formation. The method includes a purging procedure which effectively removes SO₂ before significant oxidation occurs." "One commenter requests that EPA clearly state that Method 202 should not be used for assessing compliance with emission limits set on the basis of data derived from a different measurement approach. The EPA agrees that a violation must be shown, in the first instance, by means of measurements made with the applicable test method. Once such a showing is made, however, section 113(e) of the Clean Air Act allows the Agency to rely on any credible evidence, including evidence other than the applicable test method, to establish the duration of the period of noncompliance for the purposes of assessing a penalty."

Method 202 and M5'71 are virtually the same with respect to the condensible portion of the sampling train. In fact, Method 202 allows for steps that may reduce the condensible catch when compared to M5'71. The organic extraction in Method 202 is performed with methylene chloride rather than the ether / chloroform extraction required in M5'71. The reasons for the switch include: safety, efficiency of extraction, and ease of procedures. Ultimately the methods can be described as functionally equivalent when the nitrogen purge is applied to both methods.

Proposed Policy:

The use of condensible particulate matter data is dependent on the purpose of the test. Analyzing a control device for the removal efficiency of particulate at 248 F may not require the use of condensible data. When a control device is designed to remove filterable and condensible particulate matter (as is the case with Subpart PPP sources) the use of condensible data is imperative. If the purpose of the test is to determine the ambient impact of source emissions, then the filterable and condensible particulate matter data should be used. This logic can lead to a sensible question: Are we always concerned with the impact a source has on the ambient particulate loading? Since PSD regulations are concerned with the ambient air and the guidance from the emission inventories document requires the inclusion of condensible matter, among other reasons, we can say that the end result of Air Quality regulatory work is the protection of ambient air quality. If this is the case, then condensible particulate matter should be measured and counted as part of the total particulate catch.

It is possible for a source to make measurements of the "filterable" and "condensible" catches via a single sampling train and then use the data as necessary. Since a source may be required to determine the efficiency of a control device in terms of filterable particulate only (NSPS), the data from the filterable catch alone could be used. Then, in order to determine the contribution to ambient loading, the filterable and condensible catches could be combined. Thus, the source can complete one test program with one test method, and produce enough data to satisfy the command and control regulations as well as the air quality regulations.

There will always be concerns and disagreements about specific source types and their respective emissions. Is the condensible catch double counting a pollutant? Are the measured condensibles actually artifacts due to reactions in the impingers? Would the condensible matter truly condense in the atmosphere? These questions have been addressed to some degree in this paper. A general framework to decide whether condensible particulate matter should be measured is needed. However, a regulatory agency will always need to answer specific questions on a specific basis. At the same time, the burden of proof for the exemption of data from a reference test method lies squarely on the source requesting the exemption.

Based on the investigation performed by the Stationary Source Compliance Branch into the subject of condensible particulate measurement and regulation, I believe that there are a number of options that the DAQ can take at this time and in the near future:

Condensible Particulate Measurement Implementation Options:

1) Ignore condensible particulate emissions:

Since condensible particulate contributes to ambient PM10 and PM2.5 loading, this option is unacceptable. EPA's emission inventory guideline document states that condensible particulate emissions must be included in PM10 inventories.

2) Require measurement of condensible particulate at "problem" sources, or when we believe there is a "problem."

This option is probably the easiest to implement (we already are) but leaves the DAQ open for legal action. What is the basis for defining a source as a "problem" source? High opacity would be a starting point. How do we then determine which sources are not a problem? At what percentage opacity do we define problem? If a specific source is required to test for condensibles and measures large amounts of condensible particulate, what should we require of other sources in the same industrial category? What does this approach do for consistency state wide?

3) Require condensible particulate measurement from all facilities performing M5 sampling over the course of a year.

With the new PM2.5 NAAQS we have approximately 3 years to determine significant contributors to PM2.5 loading. Since condensibles will overwhelmingly fall into the PM2.5 category, investigating condensible emissions statewide, *now*, should give us a good database to start identifying problem areas. This would be a proactive step to determine where problems exist. Also, from this data, we can define which source categories would need to continue sampling for condensibles.

4) Require condensible particulate measurement from all sources.

This procedure would eliminate any claim of bias by a specific company since the regulatory treatment would be equitable. However, sources with negligible (needs to be defined) or no condensibles would then rightly argue that they no longer need to waste their money quantifying a pollutant that is not present.

Condensible Particulate Emission Data Application:

The North Carolina DAQ has the following options with respect to actions evolved from the condensible data:

(The data will not be used for NSPS purposes, unless it is required by a specific subpart. For particulate regulated under an NSPS subpart, the subpart defines the test method for particulate determinations. Due to the physical phenomenon of particulate matter, the test method measures a defined particle state.)

1) The condensible data will only be used for emission inventory purposes

2) The condensible data will be used for emissions inventories, permit classifications, and fees.

3) The condensible data will be used for emissions inventories, permit classifications, fees, and for Prevention of Significant Deterioration purposes.

4) The data will be used for all of item 3 AND for compliance purposes with respect to the 15A NCAC 2D .0500 rules.

Item 3 would seem to be the minimum usage of the condensible data. Item 4 may also be considered viable since the state emission standards, like PSD, are linked to the ambient air loading rather than performance of a control device.

It is the recommendation of this study that the state of North Carolina require all facilities to measure condensible emissions for a period of 1 year. After that time the DAQ can determine a criteria for percentage of condensible particulate that will require sources to continue testing in the future. The data should be used for emission inventories, permit classifications, fees, PSD, and compliance with the North Carolina standards.

NSPS sources are exempt from adding condensible emissions to filterable particulate when determining compliance with the NSPS standard. However, the NSPS sources will still need to quantify and report condensible emissions for emission inventories, permit classifications, fees, and PSD.

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