

**COOPER ENVIRONMENTAL SERVICES LLC (CES)
GENERAL COMMENTS TO**

**U. S. ENVIRONMENTAL PROTECTION AGENCY
Docket ID No. EPA-HQ-OAR-2002-0051**

**NATIONAL EMISSION STANDARDS FOR HAZARDOUS AIR
POLLUTANTS FROM THE PORTLAND CEMENT MANUFACTURING
INDUSTRY**

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Proposed Rules 40 CFR Parts 60 and 63, (OAR – 2002 – 0052; FRL – 8898 – 1) RIN 2060 – AO15

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Cooper Environmental Services, LLC is a small business that develops and manufactures multi-metals continuous emissions monitors, ambient multi-metals monitors and other related equipment. CES and its staff have been conducting research in this field for over 40 years.

In addition to any formal response published in the Federal Register, it is requested that the EPA respond directly to the authors of these comments, at their earliest convenience.

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1. Introduction

EPA proposed amendments to the National Emissions Standards for Hazardous Air Pollutants (NESHAP) from the Portland Cement Manufacturing (PCM) Industry in the May 6, 2009 Federal Register (Vol. 74, No. 86, pages 21136 through 21192). This EPA proposal includes new emission limits for particulate matter (PM) and mercury (Hg) as well as other standards and modifications such as inclusion of emissions during startup, shut down and malfunctions (SSM) in the average emissions calculations, new Performance Specifications (PS) for mercury Continuous Emission Monitoring Systems (CEMS, PS 12A) and on-going quality assurance procedures for mercury CEMS (Procedure 5).

The Clean Air Act (CAA) requires the EPA to review NESHAP standards every 8 years “taking into account developments in practices, processes, and control technologies.” U.S.C. § 7412 (d) (6). It has been more than 8 years since EPA’s last general NESHAP review of this industry and **circumstances have changed significantly** since the last review. For example, the D.C. Court of Appeals has required emissions during SSM events be included, and new perspectives have been provided on the use of surrogates such as PM and volatile mercury to represent HAP metals and total mercury. *Sierra Club v. EPA*, 551 F.3d 1019 (D.C. Cir. 2008). In addition, over the past decade, new methods of monitoring metal emissions have been developed that provide the ability to continuously measure **all** phases (PM and vapor) of **all** HAP metals including mercury. These methods are commercially available, proven (more than 5 years in operation), EPA award achieving, state and EPA-approved for compliance demonstration, and listed in EPA’s Other Test Methods (EPA Other Test Methods 16 – 21). These new circumstances significantly impact EPA’s proposed rules for monitoring all HAP metals, CEMS performance specifications (PS), quality assurance (QA) procedures, proposed emission limits and SSM requirements. In addition, this new monitoring methodology can provide more complete and direct measurement of HAP metals, and is also the simpler, lower cost option to the alternative of continuously monitoring both PM and mercury vapor.

Cooper Environmental Services, LLC (CES) is submitting detailed comments on key specific analytical issues associated with Performance Specification 12A and Procedure 5 in a separate document. Our comments submitted below are of a more general nature.

It is CES’ contention that:

- **EPA needs to consider advances in multi-metals monitoring technology** when using surrogates, setting standards and defining performance specifications.
- **Use of PM as a surrogate for HAP metal emissions is no longer appropriate** given the changed circumstances that include a) the Courts requirement to include SSM emissions, b) availability of proven, multi- metal CEMS technology, c) PM does not meet the Court-defined criteria for surrogacy, d) the need to evaluate the efficacy of metal-reducing options, e) the need to evaluate residual risks for specific metals, f) meeting the CAA

mandate to use enhanced monitoring methods, and g) multi-metals CEMS represent the simplest, lowest cost monitoring option.

- **Use of vapor phase mercury as a surrogate for total mercury emissions is no longer appropriate** given the changed circumstances that include a) the Courts requirement to include SSM emissions, b) availability of proven, total mercury CEMS technology, and c) vapor phase mercury does not meet the Court-defined criteria for total mercury surrogacy.
- **SSM HAP metal emissions should be included** and can be measured with proven, contemporary monitoring methods.
- **PS 12A should NOT be generalized to other unspecified sources** in part because it defines the form (vapor) of future mercury emissions standards and inhibits development of innovative methods of measuring total mercury.
- **PS 12A should be re-written to require measurement of total mercury (PM and vapor)** and replace contemporary-technology-based requirements with performance-based requirements that can stimulate the development of improved CEMS.
- **Procedure 5 should be re-written to include total mercury** and replace contemporary-technology-based requirements with performance-based requirements.
- **Calibration drift sections of PS 12A and Procedure 5 should be re-written to separate the calibration drift check function from system integrity and accuracy functions.**

2. Circumstances have changed significantly since the last review

2.1. Inclusion of SSM emissions necessitates inclusion of PM and vapor phases of all HAP metals including mercury

The Court of Appeals for the District of Columbia Circuit recently vacated the SSM exemption. *Sierra Club v. EPA*, 551 F. 3d 1019 (D.C. cir. 2008). This Court action has the following key, relevant implications:

- **CEMS will be required** to measure emissions during randomly occurring malfunction events.
- **PM is not expected to be a good surrogate for HAP metals during SSM events** in which emissions, control of these emissions, the chemical state of the emissions and the partitioning between phases could and in realistic circumstances would be expected to be different from those during normal operation.
- **All physical phases of all HAP metals, including mercury need to be measured during SSM events.** For example, if mercury is captured on powdered activated carbon, which in turn is captured on a fabric filter, the percentage of mercury in the PM fraction downstream of the fabric filter during malfunctions could and would be expected to be significant.

2.2. Proven monitoring methodology now commercially available

In the past decade since the EPA first proposed the use of PM as a surrogate for HAP metals, a new monitoring methodology has been developed, proven and approved by the EPA for HAP

metals compliance demonstration. This methodology is based on reel-to-reel sampling using chemically reactive filter tape that traps both particulate and vapor phases of all HAP metals including mercury. HAP metals in the resulting deposit are quantified using X-ray fluorescence analysis as defined in EPA Compendium of Inorganic Methods, Method IO 3.3. [Other Test Method 16 (OTM 16) - Specifications for X-Ray Fluorescence Based Multi-Metals CEMS at Stationary Sources], (Yanca et al 2006). (Note: Since Method IO 3.3 is non-destructive and is unable to measure beryllium, this element would need to be measured off line by other methods.)

This methodology has been approved by the EPA for compliance demonstration (EPA, 2006), received EPA's Clean Air Excellence Award (EPA 2007a) and has been operating downstream of a wet scrubber on a hazardous waste incinerator for over five years (Lilly, 2009a). It has demonstrated its applicability to monitor HAP metals including mercury downstream of a wet scrubber on an EPA coal-fired boiler (EPA 2007b), and has passed an independent RATA on a coal-fired utility boiler stack down stream of dry controls (Cooper 2009). The applicability of this methodology to other sources than those noted is discussed in material previously submitted to the EPA (Lilly 2005).

2.3. EPA's cost argument no longer applicable

EPA suggested in *National Lime* that using PM as a surrogate would eliminate the cost of performance testing to comply with numerous standards for individual metals. *National Lime Association v. EPA*, 233 F. 3d 625, 637 (D.C. Cir. 2000). This is no longer relevant with the advent of this proven multi-metals methodology because by monitoring the metals directly in emissions, it would not be necessary to conduct performance tests mentioned by EPA. In fact, this proven multi-metals CEMS is the simpler, lower cost option when compared to the alternative of needing both PM and mercury CEMS. This proven, commercially available multi-metals CEMS simultaneously measures over 20 elements that have the potential to be used to assess effectiveness of emission reduction options, apportionment of ambient impacts, and process optimization. In addition, since most of the major elements present in cement kiln dust can be measured with this new technology, it should be possible to calculate the PM concentration based on the measured concentration of the major elements present in PM like Ca, Ti, Mn, Fe, Zn, Sr, Ba, etc.

2.4. Improved understanding of the behavior of HAP metal emissions

Our current understanding of the behavior of HAP metals in high temperature processes and PM controls clearly indicate that none of the Court-defined surrogacy criteria are expected to be met. (See Subsections 3.1 and 3.2 for detailed discussion.)

3. Use of PM as a surrogate for HAP metals no longer appropriate

3.1. Contemporary understanding of PCM supports expected differences in behavior of PM and HAP metals

The EPA concluded in *National Lime* that it had very little HAP metal data on cement kiln emissions. *National Lime* at 639. When the EPA referenced studies, it referred to studies conducted on coal fired utility boilers (CFUB) and their PM control efficiencies; e.g. an EPA "Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units –

Final Report to Congress Volume 1, EPA-453/R-98-004a, February 1998 (II C, page 11 of Court summary and page 31916 of June 14, 1999 (1999a) Federal Register in final rule now vacated).

The behavior of trace metals, including HAP metals in CFUB processes is well understood based on extensive studies summarized in the above EPA reference as well as others such as “Trace Elements – Emissions from Coal Combustion and Gasification” (Clark and Sloss, 1992), “Fine-Particulate Emissions From Stationary Sources and Control Technologies,” (Pan, 1987) and “Air Pollution Engineering Manual” (Buonicore and Davis, 1992). A clear and consistent picture of the behavior of trace HAP metals in the CFUB and PCM processes emerges from a review of this material that includes the following key features:

- HAP metals such as Hg, selenium (Se), arsenic (As), cadmium (Cd), lead (Pb), etc. vaporize at the high temperatures present in the combustion chambers of CFUB and PCM kilns.
- Downstream of these high temperature zones, the less volatile HAP metals cool to form and/or condense on submicron fine particles that are as a result enriched in HAP metals while the coarse particles are depleted in these metals.
- The more volatile metals (Hg, Se, Ar, Cd, Pb etc.) remain in the vapor phase to varying degree depending on the chemical form, temperature and other potential reactants available in the process stream.
- PM controls have higher removal efficiencies for larger particles resulting in lower removal efficiencies for the fine particle fraction of PM enriched in PM HAP metals.
- ESP and fabric filters have similar overall PM removal efficiencies (about 99%), but fabric filters are superior at controlling fine particulate matter that are enriched in HAP metals and thus should be superior to ESP units at controlling HAP metals
- Partitioning between vapor and PM phases can depend on temperature as noted above as well as such factors as the presence or absence of species either present in the feedstock or added to the process. Examples include the addition of powdered activated carbon (PAC) to sequester mercury; addition of chemicals to oxidize elemental mercury to increase removal efficiency in scrubbers; addition of chemicals to inhibit deleterious processes such as corrosion, poisoning of catalyst, etc. The addition of these various chemicals can impact the form and behavior of HAP metals and their removal efficiency, particularly the more reactive and semi-volatile species such as arsenic, selenium and cadmium.

This picture is further supported by a substantial body of research as summarized in the above noted summaries as well as the following specific reports: Beishon et al 1989, Boron and Wan 1990, Chadwick et al 1987, Dale and Chapman 1991, Davis and Pakrasi 1992, Gossman 2007, Gutberlet 1984 and 1988, Hicks 1991, Karlsson 1986, Kauppinen and Pakkanen 1990, Laudel et al 1991, McKenna and Furlong 1992, Meij 1989 and 1992, Meij and Vaessen 1991, Meij and Winkel 1991, Meij et al 1990 and 1991, Meserole and Chow 1991, Morrison 1986, Neme 1991, Obermiller et al 1991, Schiffner and Hesketh 1992, Sligar 1991, Smith 1980, Smith et al 1999, Tumati and Devito 1991, Turner et al 1992, Yokoyama et al 1991.

3.2. PM does NOT meet any of the three Court-defined criteria for HAP metal surrogacy

The EPA’s proposed rule incorrectly continues the use of PM as a surrogate for HAP metal emission limits without reconsidering whether continuing to use this surrogate is still

“scientifically reasonable.” *National Lime* at 637. The EPA relies on *National Lime* where the Court upheld the EPA’s use of PM as a surrogate for HAP metals in the original Portland Cement NESHAP promulgated in 1999. In a 2004 case and others, the Court of Appeals established a three part test for surrogacy based on their earlier ruling in *National Lime*. *Sierra Club v. EPA*, 353 F.3d 976 (D.C. Cir. 2004)(“*Copper Smelter MACT*”).

All three criteria must be met to use PM as a surrogate for HAP metals. These criteria are:

- “...HAP metals are invariably present in cement kiln PM...” (Subsection 3.2.1)
- “...PM control technology indiscriminately captures HAP metals along with other particulates.” (Subsection 3.2.2)
- “...PM control is the only means by which facilities “achieve” reductions in HAP metal emissions...” (Subsection 3.2.3)

Copper Smelter MACT at 984, quoting *National Lime*.

However, despite the Court of Appeal’s decision to uphold the use of a surrogate in 2000, the Court has repeatedly cautioned the EPA both in *National Lime* and more recent cases to reconsider whether PM is an appropriate surrogate for HAP metals. See *Natural Resource Defense Council v. EPA*, 489 F. 3d 875, 882-883 (D.C. Cir. 2007) (“*Brick MACT*”); See also *Cement Kiln Recycling Coalition v. EPA*, 255 F.3d 855, 863-865 (D.C. Cir 2001); See also *Copper Smelter* at 985. Specifically, the Court has directed EPA to consider whether PM is still a good surrogate in light of the potential impact upon emissions that changes in inputs to the cement manufacturing process can have, especially the possibility of fuel switching. In *National Lime* the Court states that:

“In considering the role of inputs, the EPA must also assure itself that the fuels and other inputs affect HAP metal emissions in the same fashion that they affect the other components of PM. For example, PM might not be an appropriate surrogate for HAP metals if switching fuels would decrease HAP metal emissions without causing a corresponding reduction in total PM emissions.”

National Lime at 639. This latter possibility noted by the Court is clearly the case since reducing HAP metal concentrations from their current levels in feed and fuel to zero would essentially reduce HAP metals to zero in the emissions (reducing HAP metals concentrations by many orders of magnitude) while having essentially no impact on PM emissions. It is clear that the Court intends for the EPA to consider all aspects of the PCM processes including primary input feedstock as well as fuels and additives to reduce such things as corrosion and improve control efficiencies for both PM and vapor phase species like mercury.

It needs to be emphasized that the Court in reaching its conclusion supporting the EPA’s use of PM as a surrogate for HAP metals, specifically qualified its support by defining the kind of HAP metal; i.e. non-volatile HAP metals. *National Lime* at 628 and 637. In addition the EPA used this qualifier in its arguments to the Court (EPA, 2000) and the only way the EPA’s justifying statements in its brief (EPA, 2000) could be technically correct is by restricting the discussion to non-volatile HAP metals; i.e. those metals having an insignificant (<<1%) fraction of its mass in the vapor phase at the temperatures of PM control. It needs to be noted that in this current proposal, the EPA has defined the specific metals it is referring to: arsenic, beryllium, cadmium

and lead, calling them non-volatile HAP metals. (Section II A) In Section II B it justifies its use of a surrogate (PM) for non-volatile HAP metals referring to the *National Lime* case. It is thus important to note that the EPA has restricted its reference to non-volatile HAP metals, but the elements it herein defines as non-volatile (arsenic, beryllium, cadmium and lead) have not been previously referred to as non-volatile by the EPA, nor has the EPA provided herein or else where data that can justify classifying these elements as non-volatile.

On the contrary, the EPA has previously referred to cadmium and lead as semi-volatile HAP metals, arsenic and beryllium as low volatile metals, and published reports supporting this latter classification. (EPA, 2008) In fact the EPA has even referenced tables in its own reports that would suggest that none of these metals could be classified as non-volatile by the above definition. (Tables 13-6 and 13-7, EPA 1998a) In the 18 ESP tests conducted on coal-fired utility boiler emissions, the lowest trapping efficiencies ranged from 94, 27, 0, 93, 34, 95, and 0% for arsenic, beryllium, cadmium, chromium, lead, manganese and mercury while the mean trapping efficiencies were 98, 94, 80, 97,93, 98 and 25% respectively. For fabric filters, the minimum reported trapping efficiencies were 97, 94, 0, 75, 97, 95, and 0% while the mean trapping efficiencies were 99, 99, 72, 94, 99, 98, and 36%, respectively. This EPA data clearly suggests a significant mass fraction of these metals is present in the vapor phase. In addition, there has been a continuing concern for vapor phase arsenic poisoning DENOX catalyst (Gutberlet, 1988). As such, **the classification of these elements as non-volatile HAP metals is incorrect**, both based on the EPA's own data as well as EPA's previous use of these terms and the published literature. (Germani and Zoller, 1988; Clark and Sloss, 1992). The above would suggest there is no court support for using PM as a surrogate for low or semi-volatile HAP metals; i.e. arsenic, beryllium, cadmium and lead. However, even if the non-volatile classification were to apply, none of the specific criteria the Court has laid out to guide the EPA are satisfied for all non-mercury HAP metals as will be shown below, thus eliminating any justification for using PM as a surrogate for urban HAP metals.

3.2.1. HAP metals are NOT invariably present in cement kiln PM

A significant fraction of HAP metals such as Hg, Se, As, Cd, Pb etc. can be found in the vapor phase. As such, they will be removed in PM controls with varying efficiencies as noted above. The fine particle fraction of PM will be enriched in these HAP metals, which in turn is removed in PM controls with lower efficiency than coarse particles. In a process-controlled operation like PCM, trace species like HAP metals can vary by orders of magnitude in the feed material while major species such as calcium, silicon, iron, etc. are controlled in the process and will vary by only a few percent. Thus, even though PM removal efficiency can be relatively constant, minimizing individual HAP metals in emissions will require a complex optimization of feed material, control of processes as well as selection of optimum control technology for both PM and vapor phases. As a result, even though urban HAP metals including Hg, Se, As, Cd, Pb, etc. can be found in PM, their individual concentrations will be highly variable and depend on the above noted conditions.

3.2.2. PM control technology does NOT indiscriminately capture HAP metals along with other particulates

PM control technology preferentially removes larger particles over fine particles and vapor.

As noted above, HAP metals are expected to be enriched in the fine particle fraction of PM and to have a significant fraction (>1%) of its mass in the vapor phase. This is indeed what was shown in the above noted reference and tables. (EPA 1998a) While the HAP metals exhibited widely varying removal efficiencies (beryllium from 27 to 99%, cadmium from 0 to 98%, lead from 34 to 99%), the PM capture efficiencies varied only from about 96 to 99+%. Therefore, as PM passes through the control systems, larger particles are removed more efficiently than smaller particles and vapor. In other words, PM control technology does not indiscriminately remove PM, but instead the control technology has a bias in favor of larger non-vaporized particles.

3.2.3. PM control is NOT the only means by which facilities achieve reductions in HAP metal emissions

EPA notes in its proposal that “*There are two potential feasible process changes that have the potential to affect mercury emissions. These are removing CKD from the kiln system and substituting raw materials, including fly ash, or fossil fuels with lower-mercury inputs.*” (Page 21149) Presumably, if this might be the case for mercury, the most volatile HAP metal, it would also be the case, but more so, for the less volatile HAP metals. Thus, even the EPA seems to feel there are other means than just controls by which facilities can achieve reductions in HAP metal emissions such as additives to improve PM and vapor phases of HAP metals trapping efficiencies. Furthermore, the impact of future process modifications on HAP metals cannot be predicted. For example, the addition of catalyst and corrosion inhibitors, and/or additives to oxidize mercury such as halides might have significant impacts on the volatility of other HAP metals. Since the volatility of HAP metal halides is substantially greater than most oxide forms, converting mercury to a halide may convert other HAP metals to their more volatile halide. Individual HAP metal emissions must be monitored to assess the impact of these possible future process modifications.

Thus, none of the Court-defined criteria for using PM as a surrogate for HAP metals are met. Furthermore, the reader is reminded that in the *National Lime* case, the Court sided with the EPA only in the case of non-volatile HAP metals, none of which are listed by the EPA in this proposal. Now, as demonstrated above, PM is not even an appropriate surrogate for non-volatile HAP metals. That is, monitoring PM is not an appropriate surrogate (substitute) for monitoring each individual HAP metal, nor is setting PM emission limits an appropriate surrogate (substitute) for setting emission limits for each individual HAP metal. Individual HAP metals must be measured directly to understand the impact on HAP metal emissions of various PCM process and control options. PCM processes such as recycling cement kiln dust; type and operating procedures of PM controls; use of additives to extend the lifetime of catalyst, control corrosion, capture mercury and other vapor phase metals; and modification of the characteristics and composition of input materials and fuels will all have varying degrees of impacts on the emissions of the eleven different HAP metals.

3.3. Court-defined criteria for PM surrogacy are NOT expected to be met during SSM events

As noted above in Subsection 2.1, this has several key, relevant implications as noted below:

- The only way these emissions can be included is by making emission measurements on a continuous basis with CEMS so as to include emissions during random malfunction events.

- During these events, emissions, control of these emissions, the chemical state of the emissions and the partitioning between physical phases could and in realistic circumstances would be expected to be different from those during normal operation. Under these conditions, one would not expect the PM surrogacy criteria to be met. On the contrary, the conditions during SSM events are such that the criteria for PM surrogacy established by the Court are even less likely to be met than during normal operating conditions. CEMS need to be able to measure all physical phases of HAP species including mercury PM since a significant amount could be associated with this fraction. (Gossman 2007)

3.4. Direct measurement of HAP metals is essential for the EPA to comply with the spirit and intent of the Clean Air Act.

Subsection 112(d)(3) of the CAA requires the EPA to set standards for existing sources based on the emission averages of the best performing 12% of current facilities. Further, subsection 112(d)(6) requires that the EPA review and revise these standards at least every 8 years. Thus, it is clear that Congress intended the EPA to periodically adjust emissions standards as industry standards improved. In order for the EPA to accurately revise the standards for HAP metals they need to monitor for HAP metals directly to better understand how the "best performers" reduce HAP metal emissions. That is, the efficacy of various HAP metal emission reduction options cannot be assessed unless each HAP metal is measured because each has unique and wide ranging chemical and physical properties dictating their presence and behavior under various conditions. This will be particularly important if processes and/or chemistry changes through the addition of reactants to facilitate other aspects of the process such as minimization of corrosion and catalyst poisoning, enhancement of collection efficiency of other species like mercury, etc.

Failure to monitor HAP metals directly will significantly impair the EPA's ability to revise emissions standards in the future and would not be in keeping with the intent of the CAA to ensure that emissions standards are updated every 8 years based on improvements that the best performers have implemented.

3.5. Direct measurement of HAP metals is required to evaluate residual risks

HAP metals have wide ranging health and environmental impacts. Unless each is monitored directly and continuously, these impacts cannot be fully assessed. Certainly, simply monitoring PM provides essentially no information for assessing these potential health and residual risk impacts. PM is a particularly poor surrogate for assessing the health risks associated with HAP metals such as Hg, As, Pb, Cd, Cr, etc.

In addition, section 112(k)(1) of the CAA encourages the connection between source emissions and ambient exposure to effectively reduce public health risks such as cancer associated with source emissions. In 112(k)(2), congress clearly states its intent to have both the ambient air and source emissions monitored for metals to characterize the source emissions and define the contribution of these sources to public health risks from HAP. Defining and quantifying the contribution of sources to ambient risks is greatly aided by the direct measurement of specific HAP metals as well as other metals that can be provided by the new monitoring methodology noted in Subsection 2.2.

3.6. Congressional mandate in the CAA to use enhanced monitoring

In section 114 of the CAA, congress gives EPA a mandate to require enhanced monitoring (42 U.S.C. 7414 (a) (3)). Major sources are required to use enhanced monitoring such that there is reasonable assurance of HAP control. Clearly, periodic testing does not provide reasonable assurance of control nor does monitoring PM provide reasonable assurance of urban HAP metal control. Similarly, monitoring only the vapor phase of mercury does not provide reasonable assurance of control under all conditions that are now to include SSM events. This is particularly the case when there is a proven and approved technology available that measures all phases of mercury as well as all phases of all urban HAP metals.

Clearly, direct monitoring of all phases of all HAP metals is an enhancement over the monitoring of a surrogate (PM) for some urban HAP metals and one phase of mercury.

To emphasize the importance of requiring this type of enhanced monitoring on all sources, we note an event that was recently recorded in East St. Louis, Illinois. On April 13, 2009 a continuous ambient metals monitor in the vicinity of schools recorded a two hour period in which the average arsenic concentration recorded was 2,300 ng/m³. For perspective, OSHA suggests that no worker should be exposed to arsenic concentrations greater than 2,000 ng/m³ for longer than 15 minutes. Clearly, a nearby source emitted arsenic at much higher concentrations than those measured in the community. These emissions went unrecorded because there was no requirement for the possible source to continuously measure metals in its emissions. Because the winds were changing during this monitoring period, it is uncertain how long the emissions lasted, but if a multi-metals CEMS or fence line monitor had been in place, the problem might have been identified early enough such that the emissions could have been minimized. In addition, had the emissions been more significant, the nearby population could have been alerted had there been a multi-metals CEMS or fence line monitor in place. Clearly, requiring enhanced monitoring is a real and significant issue that the EPA should consider seriously in its comment review.

3.7. Availability of new monitoring technology

As noted above in Subsection 2.2, a new, proven multi-metals CEMS is commercially available and accepted by the EPA. Thus, use of PM as a surrogate for HAP metals monitoring is no longer needed.

3.8. HAP multi-metals CEMS is the lower cost, simpler option

With the Court ruling requiring the inclusion of emissions from SSM events in emission averages, it will be essential that CEMS be available that can measure emissions of all phases of HAP metals including mercury PM. As such, the use of multi-metal CEMS methods would allow all of the urban HAP metals including mercury and all phases to be continuously monitored with a single CEMS. This option would be simpler, more reliable and have lower initial and on-going costs than the only other option consisting of two CEMS, one for PM and one for mercury vapor. In addition, these multi-metal CEMS can provide emission measurements for over 20 metals including all 11 HAP metals. Measurement of the non-HAP metals might be used for process diagnostics, source apportionment in the ambient environment and other possible applications not yet realized.

EPA suggested using PM as a surrogate would eliminate the cost of performance testing to comply with numerous standards for individual metals. This is no longer relevant with the advent of the new multi-metals monitoring methodology (Subsection 2.2) because by monitoring the metals directly in emissions, it is not necessary to conduct the performance tests mentioned by the EPA.

3.9. Public has a right to know HAP metal emissions in plain language

If the EPA has decided that emission of HAP metals is sufficient to require regulation, the public has the right to know what those individual HAP metal emissions are in plain language; that is in a language the public can relate to ambient measurements. The EPA uses different units to define emission limits for different HAP. The EPA should use reasonably common units at least in form and common with typical ambient measurement units such as ng/m³.

4. Startup, shutdown and malfunction Events should be included

The Court of Appeals for the District of Columbia Circuit vacated the SSM exemption. (*Sierra Club v. EPA*, 551 F. 3d 1019 (D.C. cir. 2008)).

CES is supportive of the inclusion of these emissions, feels it is a step in the right direction, will help plants better manage their emissions and will provide local stakeholders with the assurance that these emissions will be minimized in general as well as recognized early during malfunctions and thus minimized. (See last paragraph in Subsection 3.6.) New HAP metals monitoring methodologies certainly make this possible for all urban HAP metals as noted above. As such, CES strongly encourages the EPA to include these emissions as the Court has directed.

As noted above in Subsection 2.1, inclusion of these emissions has significant implications. Although it is now technically possible to include these HAP metals, it may not be possible for all HAP species. However, such limitations should not prevent the EPA from moving forward with the inclusion of these emissions for those HAP species for which it is now technically feasible (HAP metals) and will in fact encourage the development of technologies for those HAP for which it might not be feasible at this time.

Emissions from these events should be included because current methodology allows these emissions to be monitored at no additional costs and if these emissions are significant, they should be included and if they are not significant it doesn't matter since the cost of including them is insignificant with this proven multi-metals CEMS technology.

5. Mercury emission limits should include all mercury phases (PM and vapor)

5.1. CAA specifies mercury NOT vapor phase mercury

It is inappropriate to set emission limits for just vapor phase mercury. Nothing in the CAA says the EPA may set emission limits for only one physical phase of a HAP metal. This is contrary to the intent of the CAA and could result in the omission of a significant fraction of mercury emissions. What the EPA appears to be suggesting is that vapor phase mercury can be used as a surrogate for total mercury emissions. A review of the Court-defined criteria noted in

Subsection 3.2 of this document clearly indicates that vapor phase mercury does not meet these surrogacy criteria.

5.2. Vapor phase mercury does NOT meet the Court-defined criteria for total mercury surrogacy

Although historical measurements of emissions from CFUB would suggest a small fraction of the mercury mass emissions is in the PM fraction, EPA's own data (EPA 1998a) suggests that about 30% (not insignificant) of mercury is captured in PM controls and over 90% might be captured in PM controls if powdered activated carbon is used to control Hg emissions. Clearly, during SSM events and more specifically during malfunctions in PM controls, a significant fraction of the Hg mass could be in the PM fraction of emissions, which would be excluded under EPA's current proposal to regulate only the vapor phase of mercury.

However, more relevant to the EPA's current proposal is what the EPA says about the PCM process on page 21150. *"Among kilns that waste CKD, the percentage reduction in mercury emissions by wasting CKD ranged from 0.13 percent to 82 percent, with an average of 16.5 percent and median of 7 percent."* The EPA went on to state that *"Mercury speciation may affect the extent to which mercury accumulates in the CKD, with particulate and oxidized mercury more likely to accumulate ..."* In addition, the EPA says that *"Reducing mercury emissions through the wasting of CKD may be feasible ..."* *"However the degree to which CKD can be used to reduce mercury emissions cannot be accurately estimated..."* and *"We do not have data that would allow us to quantify the effect of mercury speciation."*

Clearly, volatile mercury does not meet the surrogacy criteria to represent total mercury emissions (PM and vapor). That is:

- Mercury vapor is NOT invariably present in cement kiln total mercury emissions; i. e. mercury vapor is not a constant fraction of total mercury at various stages of the process and emissions control, but is also found in significant quantities in the PM phase.
- Mercury vapor control technology does NOT indiscriminately capture total mercury emissions
- Mercury vapor control is NOT the only means by which facilities can achieve reductions in total mercury emissions.

Based on the above, CES requests the EPA change its emission limit from one based on vapor phase mercury to one based on total mercury that includes both PM and vapor phases.

5.3. Total mercury can now be measured with proven methods

It is now possible to measure total mercury emissions (all phases including PM) continuously using proven and EPA accepted monitoring methodologies. (Subsection 2.2). Regulating and monitoring total mercury (vapor and PM) would be an enhancement over just regulating and monitoring vapor phase mercury.

This requested change is important and necessary to account for mercury emissions during SSM events. During these events, a significant fraction of mercury emissions might be in the PM fraction. In addition, as noted above, PM-bound Hg is significant during normal PCM processes

including PM control operational cycles as PM trapping efficiencies can change during these cycles.

5.4. Total mercury monitoring represents enhanced mercury monitoring

This would also be consistent with congressional wishes that the EPA require enhanced monitoring where it is appropriate to assure continual compliance with emission limits (Total mercury monitoring is enhanced monitoring over monitoring just one phase of mercury, vapor phase.) This recommended change would benefit the EPA and other stakeholders by providing more complete information assuring compliance and if a multi-metals monitor were used could provide compliance demonstration information for all phases of all urban HAP metals while simultaneously demonstrating compliance with a total mercury emission limit. This monitoring would be simpler and achieved at a lower cost because it would replace two CEMS, mercury vapor and PM. In addition, the plant, agency and public would benefit from the non-urban HAP metals monitored with the same CEMS that might provide diagnostic information to improve plant operation, data on all HAP metals not just for selected HAP metals and source apportionment information, all at a lower cost than the alternative of using two CEMS providing only two parameters (PM and vapor phase mercury) instead of over 20 metals

6. PS 12A should NOT be generalized to other, unspecified sources

The EPA states in its summary that these “...specifications would be generally applicable and so could apply to sources from categories other than and in addition to, Portland cement ...”

Because emission standards and measurement methods are not written independent of each other, defining the measurement method in effect defines the emission limit/standard for other, unspecified sources. This is particularly the case for mercury since the EPA is proposing to use volatile mercury as a surrogate for total mercury emissions. By generalizing this measurement technology to vapor phase mercury, the EPA appears to want to generalize the emission standards for other, as yet to be specified sources to vapor phase mercury. By doing this, the EPA greatly inhibits the development of new methods that might measure total mercury.

CES believes the EPA has not provided data to justify the extension of this technology to other sources or why it might be a reasonable measure for compliance demonstration for either this source (See Section 5 above) or other unspecified sources. As noted above in Section 5, vapor phase mercury is clearly not an adequate indicator of compliance with the CAA or an appropriate surrogate for total mercury emissions for PCM sources.

As such, CES requests the EPA not approve this monitoring technology for either PCM sources (See below) or other unspecified sources.

7. Performance Specification 12A should be re-written to replace contemporary-technology-specific requirements with generalized measurement-objective driven requirements

7.1. Contemporary-technology-specific requirements

CES requests the EPA modify Performance Specification 12A (PS 12A) to eliminate those requirements that are “contemporary-technology-specific” (CTS) as discussed in more detail below. The requirements currently in PS 12A are not solely based on performance, but instead include CEMS construction requirements and tests that are specific to a particular technology. This is important because as written, PS 12A fixes future technology and emission standards to the capabilities of the measurement technology available when PS 12A was written, stifles innovation and **UNNECESSARILY** requires NIST certified standards or traceable vapor phase mercury generators to conduct linearity and drift tests. The plant, public and EPA all benefit when unnecessary barriers to improvements are removed.

The primary function of performance specifications should be to demonstrate initial validity of results from a CEMS (an automated sampling and analysis system). This validity is established by demonstrating the CEMS meets **performance-based** analytical requirements that generally include tests to demonstrate its precision, accuracy, linearity and stability (calibration drift).

The current PS 12A is confusing in part because these basic functions are not clearly delineated and in some cases combined as summarized below in Table 1.

Table 1. Summary of EPA Proposed Performance Specification 12A (Appendix B, page 21171)
Specifications for initial evaluation of acceptability/certification of total vapor phase mercury CEMS

QA Parameter	Ref.	Objective	Method	Metric	Freq.	Acceptance Criteria	Comments
Linearity	3.6, 8.3, 13.1	Demo. linearity Demo. quant. range	Ref. gas comp.	% span	once	< 5% of span (2 times std.)	Requires NIST stds. Emiss. Std. not defined in µg/dscm
Calib. Drift	3.7, 8.4, 13.2	Demo. anal. stability Demo. sys. integrity Demo. sys. accuracy	Ref. gas comp.	% span	7 daily	< 5% of span (2 times std.)	Requires NIST stds. Standard not defined in µg/dscm Not mentioned but implied Not mentioned but implied
Accuracy	3.8, 8.5, 13.3	Demo. Accuracy	RATA	% RA % RA Conc. diff.	once	<10% if RM>10 µg/dscm <20% if RM < 10 µg/dscm < 1 µg/dscm if RM < 5 µg/dscm	RM comp. at operating cond.
Sys. integrity	ND	System integrity	ND	ND	ND	ND	Not specifically mentioned

ND: Not defined

These issues and specific examples of non-performance based requirements (CTS requirements) in PS 12A include, but are not limited to the examples presented and discussed in the following subsections.

7.2. Analyte should be redefined as total mercury including PM mercury (Subsection 1.1)

EPA’s selection of vapor phase mercury as the analyte in PS 12A is the clearest indication of a CTS requirement. The current PS 12A is based in part on the perceived monitoring capabilities available in the middle of this decade when the Clean Air Mercury Rule for CFUB was promulgated. At that time, the only mercury monitoring capability appeared to be that based on sensing of mercury in its elemental vapor form. This technology was well established in ambient air monitoring and had been used on some sources in Europe. Even though congress had specified mercury in the CAA, not vapor phase mercury, the reasoning was that the vast majority of mercury in CFUB emissions was in the vapor phase and thus measurement of the vapor phase should be a reasonable approximation of total mercury. However, this assumption did not include SSM events, PCM sources, etc. Clearly, the defined analyte in PS 12A is based **not** on a

generalized measurement objective (mercury as specified in the CAA) but specifically on what was perceived as **measurable with available technology** (vapor phase) when written; i.e. a clear CTS requirement.

Thus, as noted above in Sections 5 and 6, the emission limit should be based on total mercury including PM and vapor phases, and the PS should be modified to reflect this because:

- The CAA specifies mercury, not vapor phase mercury
- Inclusion of SSM events requires the ability to measure total mercury (PM plus vapor) continuously
- Vapor phase mercury does not meet the Court-defined criteria for total mercury surrogacy. (See Subsection 5.2.)
- A proven and EPA-approved method to measure total mercury on a continuing basis is commercially available.

In addition, the **default position should be to monitor total mercury**. If the EPA chooses to propose using vapor phase mercury as a surrogate for total mercury, the burden of proof should be on the EPA to show why vapor phase mercury is an acceptable surrogate by presenting data showing that it meets the Court-defined criteria for surrogacy. The EPA has not provided such data, and as such, the vapor phase requirement should be modified to read total mercury.

7.3 Linearity (Subsections 3.6, 8.3 and 13.1)

Linearity is defined by the EPA in Subsection 3.6 as the absolute difference between a CEMS reported result and a reference gas (NIST traceable or certified) value expressed as a percent of the span value when the entire CEMS including the sampling interface is challenged. The first problem with this definition is that the acceptance criterion is a percent of the span. The span is defined as two times the standard (limit), which is 43 tons per million tons of clinker for existing sources. However, the span in $\mu\text{g}/\text{m}^3$ would be different for each plant and could vary over time. In addition, it is clear that by requiring a reference gas to establish linearity, the EPA is assuming that only gas phase mercury sensors are available; i.e. a CTS criterion.

From CES' perspective, the primary performance evaluation function of the linearity test should be to demonstrate a linear response of the CEMS mercury sensor over a concentration range defined by the operator; i. e. the range of quantitation. If linearity is demonstrated over this range, quantitation and system integrity over this range can be demonstrated by establishing accuracy at a single point with passing a RATA as required by the current PS 12A. Requiring NIST certified or traceable standards is unnecessary to define linearity. Any secondary standard whose concentration can be traceable to a NIST standard would be adequate. It is standard laboratory practice to use secondary standards to perform routine functions such as establishing an instrument's linearity or working curve. More rigorous and expensive NIST standards are generally used to establish accuracy or traceability to NIST standards. However, the EPA proposed challenge of the CEMS with NIST traceable standards **cannot establish accuracy** because it does so in a clean gas, not in the presence of stack gas as would be done in the case of a dynamic spiking test.

In addition, the current PS 12A defines a specific location in a hardware configuration where the standard is to be introduced. Again, this is specific to contemporary technology which may be

different for different types of CEMS such as *in situ* based CEMS. In addition, linearity is primarily a function of the analytical sensing component of the CEMS and it isn't necessary to challenge the entire CEMS to evaluate/test the CEMS linearity. One approach to assuring system integrity may be to challenge the entire system with a NIST-certified traceable gas, but it is certainly not the only method nor should this function (system integrity) be required to be demonstrated in this linearity demonstration performance specification.

CES requests the EPA re-write this subsection to meet the intended generic linearity objective. This specification is clearly a CTS requirement. For example, requiring different chemical forms of mercury would be unnecessary for *in situ* mercury measurements with X-ray based sensors (response is independent of chemical form, there would be no catalyst or reactant involved, and there would be no transport of mercury). In the specific case of an *in situ* X-ray based CEMS noted above, only sensor linearity would need to be checked, which could be done with a solid or gas of any chemical form. CES recommends EPA clearly define the analytical objectives of this linearity check and specify performance based linearity requirements. CES further recommends the EPA omit "filtration system" and add Hg PM; replace "reference gas" with "reference sample" and "zero" with "low"; and use an acceptance metric expressed as % of mean value at each level. This is particularly important since the span is not well defined and the valid range needs to extend well beyond the standard value to include concentrations that might be experienced over short monitoring intervals during SSM events.

7.4. Calibration drift (Subsections 3.7, 8.4 and 13.2)

The calibration drift is defined in Subsection 3.7 as the difference between a reference gas (NIST certified/traceable) concentration and the CEMS reported concentration expressed as percent of span value when the entire CEMS is challenged after a period during which no maintenance or adjustments have taken place. This definition suffers from the same limitations noted above for the linearity tests; i. e. the acceptance criterion is based on a variable standard, unnecessarily requires testing with NIST certified/traceable standards and requires specific location for testing. Again, it is clear that the calibration drift check/test as defined in PS 12A is based on perceived CTS requirements and unnecessarily includes by implication a system integrity check and an accuracy demonstration function that significantly complicates the method and inhibits the development of improved CEMS. These CTS requirements include:

- Requirement to use of a NIST certified/traceable reference gas to check calibration factor drift.
- Requirement that this calibration check also serve the function of system integrity check.
- Requirement that this calibration check serve the function of accuracy check.

Each of these requirements/functions are considered to be CTS, fix the CEMS technology to the perceived technology capabilities at the time of writing the PS and significantly inhibits development of new, innovative approaches to this CEMS technology.

7.4.1 Analytical measurement background

Most analytical procedures and certainly most CEMS are based on either *in situ* analysis or extractive methods. Extractive methods include two primary steps, extraction of a representative sample of stack gas coupled with an analysis step. The analysis step in both approaches is typically based on a comparison of a mercury sensor's response to a standard of known

concentration to the sensor's response to an unknown sample. The instrumental response to a standard (response/concentration) is commonly referred to as a calibration factor. This calibration factor is typically determined separate from the determination of the instrument response to an unknown. Inherent in this comparison is the assumption that the calibration factor is the same during the unknown analysis as when it was originally determined minutes to years earlier for the standard.

The primary function of a calibration check is to provide documentation that the original calibration has not drifted or changed between the time of calibration and unknown analysis. This can be done simply by analyzing a sample of **constant concentration** at the time of calibration and showing that the instrumental response to this constant sample has not varied. This is routinely done in the laboratory by re-analyzing a solution whose concentration is stable (secondary standard) before and after unknown samples are analyzed or non-destructive analysis of a solid sample (secondary standard). It needs to be pointed out that this function in the laboratory does **not** require a standard, much less a **NIST-traceable** standard, but simply a sample that is reliably known to be constant. For example, most laboratory methods for analysis of solution samples using atomic methods (fluorescence, absorption, emission; e.g. EPA Method IO-3.2) require daily instrument calibration (determination of daily working calibration curves) with periodic calibration checks over a day of analysis. Laboratory X-ray fluorescence analysis instruments are much more stable requiring calibrations no more frequently than every year or two with calibration **"checks"** on a daily basis (EPA Method IO-3.3).

7.4.2 Reference gas required for calibration check

This requirement is CTS in that it assumes a gas is required to check the calibration factor drift with all current and future mercury CEMS. It is clearly designed to check the calibration of a CEMS based on a sensor that measures mercury in the vapor phase and a sensor whose response is different for different forms of mercury vapor. As such, it is technology specific and not based on performance; i.e. stability of the CEMS calibration factor. Other CEMS methods that can meet calibration factor stability performance criteria should not be limited to demonstrating calibration stability by a method optimized for other sensors. **Clearly, it would be unreasonable to ask a method based on atomic fluorescence or absorption to check its calibration with a thin film standard that might be optimum for an X-ray fluorescence based method.**

CES strongly recommends that the calibration drift requirement be re-written to accomplish only the primary stated function –calibration drift check. The system integrity check and accuracy functions should be in separate requirements.

7.4.3 System integrity check better defined in separate, independent performance specification

The EPA in its current calibration drift test has included by implication a system integrity check function by requiring the entire system from probe to analyzer be challenged. CES recommends that these two functions be separated. This will greatly simplify the calibration check and allow the system integrity check to focus on demonstrating the stability and/or validity of the entire CEMS since the last RATA. As noted above, a certified, NIST-traceable reference gas standard or mercury generator is not required to demonstrate calibration factor stability.

A system integrity check can be accomplished in several ways including periodically comparing stack gas concentrations with CEMS readings, parameter monitoring, periodic challenges to the entire system by spiking into clean air, dynamic spiking into stack gas, etc. or a combination of the above. By separating this system integrity check from the calibration drift check, each function can be accomplished more efficiently and at the same time encourage innovations appropriate to specific technologies.

In addition, if the accuracy check function is separated from both the calibration and system integrity check functions, neither would require NIST-traceable standards to perform these checks/tests, greatly reducing their complexity and costs.

Another important feature of the EPA defined calibration drift test is the period between drift checks. The EPA requires that these tests be performed daily and in so doing assumes this frequency is both adequate and not unnecessarily excessive and **thus sets the requirement based on perceived limitations or capabilities of contemporary monitors.**

In writing a new system integrity check, CES strongly recommends that the EPA not require it to demonstrate accuracy as well, but only demonstrate the integrity of the system has not changed since the last RATA. Again, using NIST-traceable standards for this function is but one possible approach; however, one that is far more complicated than required. Unless the system integrity check includes dynamic spiking of the stack gas, the system integrity check will not serve as an adequate check of the system accuracy. System accuracy assurance should be left to other requirements as defined in RAA or RATA.

7.4.4 CEMS accuracy already adequately defined with relative accuracy audits

CES recommends the EPA accomplish the function of assuring the initial and on-going accuracy of CEMS reported results with initial and periodic accuracy audits. One of the best ways to assure accuracy is with a RATA using an EPA reference method such as 30B because it challenges the entire CEMS system under actual stack gas conditions including all interfering species. Although it is not economically practical to conduct frequent RATA, other RAA using standards and/or surrogate reference methods might be available to demonstrate continued system integrity and accuracy between RATA.

Clearly, by unnecessarily including a requirement to use a certified, NIST-traceable mercury generator in the calibration check, the EPA has unnecessarily complicated the existing CEMS systems. In doing so, it has required the development of not only complicated mercury generators that then must be certified and checked for drift, but also has required the development of complicated procedures to trace generator emissions to NIST and establish the stability of the emissions from these mercury generators. By insisting that this NIST-traceability be accomplished in this way, the EPA appears to be attempting to duplicate the accuracy demonstration function more appropriately provided by the RATA. In addition, the same tests the EPA is proposing in its “Interim EPA Traceability Protocols for the Qualification and Certification of Elemental and Oxidized Mercury Gas Generators” (EPA 2009 b, c, d, e) to validate NIST certified generators might just as well be applied directly to the CEMS. By

requiring these generators to be used in this performance specification, all the EPA has done is to transfer the stability certification step from the CEMS to a NIST certified gas or generator.

7.5 Accuracy (3.8, 8.5 and 13.3)

The test based on a RATA comparing a FRM measured stack gas mercury concentration with a reported CEMS measured concentration is both appropriate and adequate to establish both accuracy and system integrity at the stack gas concentration. If the CEMS has also passed a linearity test as well as a calibration drift test, traceability of the accuracy can be extended to the entire analytical concentration range as well as the period for which the calibration check continues to demonstrate stability.

CES strongly recommends that the EPA also consider allowing the operator to establish accuracy using dynamic spiking methods. This is important because the technology to do so is now available. For example, using a quantitative aerosol generator (QAG) to dynamically spike a multi-metals aerosol into stack gas (EPA Other Test Method OTN-18), it should be possible to distinguish between the spiked mercury and the native stack gas mercury if a multi-metals CEMS is used to monitor the total mercury.

7.6 System integrity

System integrity checks are not required by the proposed performance specification during this initial validation. The linearity and calibration tests validate the analytical components while the RATA validates the entire sampling and analysis system.

7.7 Reagents and standards (Subsections of EPA proposal: 6.2, 7.0 and 7.1)

Subsections 6.2, 7.0 and 7.1 appear to be technology specific. It is implied in these subsections that the only method to verify system integrity, linearity, etc. is by challenging the CEMS with NIST-traceable gas standards. Again, this may be the case for the commercially available vapor phase mercury CEMS in the middle of this decade, but this is no longer the case. For example, calibration drift and system integrity checks might be accomplished by periodically running simultaneous stack tests with a reference method (RM) or other approved stack test. Although this might be impractical for relatively unstable systems, it might be more practical for more stable systems. However, it should be noted that this type of comparison would be more challenging, more stringent and more realistic; i.e. it relates to actual operating conditions, not conditions when analyzing clean air spiked with a NIST-traceable standard as required by the current PS 12A. It should also be noted that the accuracy of the CEMS is established by the initial and periodic relative accuracy test audits (RATA), which is the most realistic check of accuracy. Another example of where RM comparisons might be more appropriate than challenges with NIST-traceable gas standards would be for a total-mercury monitor that might be based on *in situ* measurements using laser or X-ray based sensors. Current requirements as specified in PS 12A would inhibit this type of innovative development.

In addition, the requirement to use NIST-traceable standards for periodic system and calibration checks is unnecessary. What is required is a reference source that is stable over time so that the accuracy established at the initial certification and/or periodic RATA can be extended to the time period between RAA and RATA. To do this does not require a NIST-traceable standard, only a stable source of mercury that can be run during each RATA and periodically between the RATA

to demonstrate continued stability of the CEMS relative to the two RATA bounding the run period. Requiring NIST-traceability of these “check” materials greatly increases the complexity of the system, requires the creation of an entire instrument and industry around this requirement, and is again contemporary-technology specific. For example, use of any stable standard of reasonable quality (in contrast to NIST-traceable) for periodic checks that would be run both during periods of RATA checks for accuracy and periods in between RATA should provide more than adequate quality assurance the accuracy established during the RATA is being maintained during the periods between RATA.

It should also be noted that if the standard were to be changed to total mercury (PM plus vapor), a QAG is available and has been approved by the EPA (EPA Method OTM-18). With this QAG, it is possible to challenge a system with an aerosol containing both PM and gas phase mercury species. However, even if the PS 12A is not changed to total mercury, PS 12A would still be CTS since more stable systems might be challenged at the low, medium and high concentrations in different ways from NIST-traceable gas standards such as the use of a QAG generated aerosol to challenge the entire system.

Since the EPA has given the operator the responsibility to calibrate, maintain, and operate the CEMS “properly” in Subsection 1.2.2, CES recommends the EPA give the operator the responsibility of defining an operating procedure (approvable by the EPA) that meets EPA’s general QA/QC objectives of accuracy, stability, precision and linearity (performance based specifications) developed around EPA Method 301. This would in effect, remove barriers to new, innovative technologies while maintaining the accuracy and quality of the CEMS reported concentrations.

7.8 Span value (Subsection 3.5)

The span value which is currently tied to the emission limit should be re-written to define a value an operator selects as the upper end of the range for which results will be validated and accepted by the agency. Along with this, the agency should define the consequence of reported values that might exceed this defined upper end of the range. The operator should be required to replace any value that exceeds this range with a default value that might be defined as some multiple of the span value. This is important in this particular application (current proposed rule) because the measured values over the CEMS short sampling and measurement intervals are expected to be substantially greater than the proposed 30-day rolling average emission standard. As such, limiting the span to two (2) times the emission standard is unnecessarily overly restrictive and likely to result in many CEMS results exceeding the span value as defined.

8. Procedure 5: On-going quality assurance should be re-written to replace contemporary-technology-specific requirements with generalized measurement-objective driven requirements

The general features of Procedure 5 are summarized below in Table 2. Essentially all of the comments noted above for PS 12A (Sections 7) are applicable to Procedure 5. Key among these are CTS requirements and the inclusion of multiple functions in the calibration drift check requirement. As can be seen from Table 2, the calibration drift check requirement has within it

implied system integrity, accuracy and linearity check functions. CES strongly recommends that EPA modify Procedure 5 by separating these four QA/QC functions as discussed in Section 7.

Table 2. EPA Proposed Procedure 5 (Appendix F, page 21180)
On-going QA/QC requirements for total vapor phase mercury CEMS

QA Parameter	Ref.	Objective	Method	Metric	Freq.	Acceptance Criteria	Comments
Linearity	ND	ND	ND	ND	ND	ND	Implied with CD requirements
Calib. Drift	2.4, 4	Demo. anal. stability	Ref. gas comp.	% span	daily	<10% of span (2 times std.)	Requires NIST stds. Standard not defined in µg/dscm
	ND	Demo. sys. integrity	ND	ND	ND	ND	Not mentioned but implied
	ND	Demo. sys. accuracy	ND	ND	ND	ND	Not mentioned but implied
	ND	Demo. sys. linearity	ND	ND	ND	ND	Not mentioned but implied
Accuracy	2.6, 5	Demo. accuracy	RATA	% RA % RA Conc. diff.	annually	<10% if RM>10 µg/dscm <20% if RM < 10 µg/dscm	RM comp. at operating cond.
			RAA		quarterly	< 1 µg/dscm if RM < 5 µg/dscm < 15% of audit sample or < 7.5% of std.	Standard not defined in µg/dscm
Sys. integrity	2.5, 3.0 (3)	Not stated	Ref. gas comp.	ND	ND	ND	

ND: Not defined

Each of these requirements/functions (system integrity, linearity and accuracy) are considered to be CTS, and cements the CEMS technology to the perceived technology capabilities at the time of writing the PS. This significantly inhibits development of new, innovative approaches to this CEMS technology. (See comments in Subsections 7.3.1 through 7.3.4)

However, the most significant of these is the proposed method of **establishing accuracy traceability** between RATA as discussed in more detail below.

DEMONSTRATING ON-GOING ACCURACY TRACEABILITY

As noted above in Table 2, the EPA appears to be proposing to establishing on-going accuracy traceability with periodic (annual) RATA where the CEMS results are compared to RM results. Although not stated directly, the EPA also appears to suggest that accuracy between these annual RATA can be demonstrated by challenging the CEMS with clean air spiked with NIST-traceable mercury from a NIST-certified generator. As noted above in Subsection 7.5, spiking clean air is not the same as a dynamically-spiked stack gas challenge and is only suggestive of accuracy in an interference-free and unrealistic condition. This is not a minor issue, particularly when the EPA requires all CEMS meet this same, less than ideal accuracy traceability requirement. As such, it is important to understand the origin of this requirement and its assumed necessity for all CEMS to demonstrate on-going accuracy traceability.

CES believes that at the foundation of this requirement is the assumption that all CEMS are inherently unstable and need to be re-calibrated frequently, on the order of daily. This is important because each time the CEMS calibration drifts enough that it needs to be re-calibrated, the chain of accuracy traceability to the most recent RATA is broken. Thus, each time the CEMS is calibrated, there is a need to re-establish accuracy traceability, if not to a recent RATA, then to a NIST standard. Thus, the frequency with which this accuracy traceability must be re-established depends on the frequency with which the CEMS needs to be re-calibrated because of excessive drift.

It is important for the EPA to note that not all CEMS are as unstable as those CEMS around which the current Procedure 5 appears to have been written. For example, CEMS based on X-ray fluorescence are extremely stable and require re-calibration typically no more frequently than annually. A demonstration of this stability is illustrated below in Figure 1.

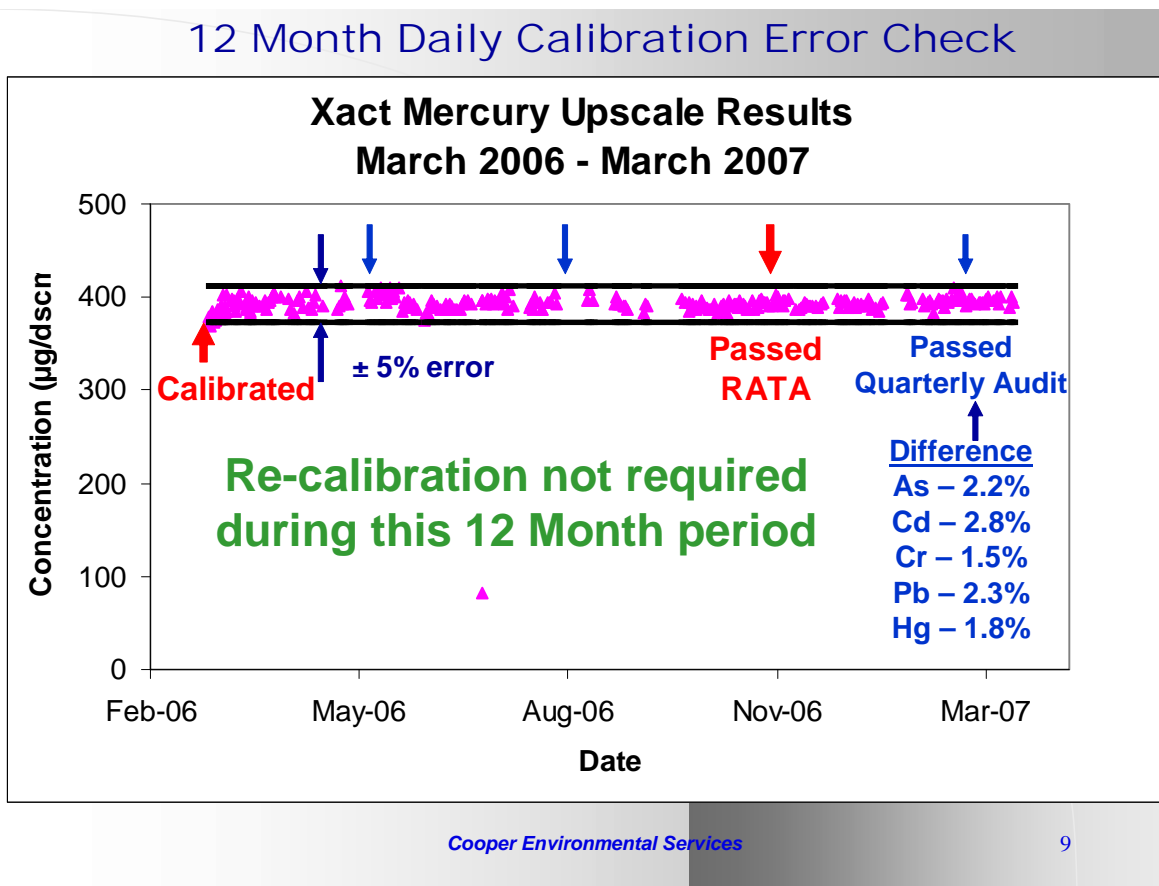


Figure 1. Plot of daily calibration error “check” for a CES Xact multi-metals CEMS monitoring emissions from a hazardous waste incinerator showing calibration stability within $\pm 5\%$ over a period of about one year during which the chain of accuracy traceability was not broken with a CEMS recalibration.

This figure shows a plot of daily calibration error “check” for a CES Xact multi-metals CEMS monitoring emissions from a hazardous waste incinerator. The calibration error check is **NOT** a recalibration, but simply a re-analysis of a stable sample that was analyzed the last time the CEMS was recalibrated with standards traceable to NIST standards. This plot demonstrates that the CEMS calibration was stable to within $\pm 5\%$ over a period of about one year during which the chain of accuracy traceability was not broken with a CEMS recalibration. This same period in which the CEMS was not recalibrated, the CEMS passed three quarterly audits and a RATA.

The generic, measurement-objective-driven performance specification should be to establish accuracy traceability for periods between RATA. This can be accomplished in several ways one of which might be to challenge the CEMS on a daily basis using reference gases from a NIST

certified generator. Although this might be necessary for relatively unstable CEMS, it is CTS and should not be required of all CEMS technologies, particularly those with calibration stabilities on the order of years.

CES recommends the EPA re-write Procedure 5 to eliminate CTS requirements such as calibration drift checks with NIST-traceable gases and specified frequencies of these checks. CES further recommends the EPA allow the CEMS operator to define the frequency and then demonstrate the system stability during its proposed calibration check period.

9. Other comments and suggested edits

9.1. Units –

CES recommends the EPA use plain language and units that can be related to ambient measurements for its emission limits. The units currently proposed cannot be related to ambient measurements, requires modifying the direct CEMS output with inputs under the control of the plant, which make the results un-interpretable by anyone wanting to make comparisons with ambient measurements or health effects; or the CEMS concentration readings should be required to be posted/reported.

9.2. Use of coal ash

Coal ash is enriched in other urban HAP metals besides Hg. These enrichments over coal are on the order of 5 to 10 fold. With these enrichments, it will be even more important that emission limits be developed for each individual urban HAP metal and be correspondingly monitored. Without this assurance, the EPA should not allow coal ash to be used in the manufacture of Portland cement.

9.3. Specific comments and recommended changes to PS 12A and Procedure 5

CES requests the EPA consider making the following specific changes to the performance specifications to reflect changes to total mercury monitoring and the elimination of contemporary-technology-specific requirements as well as others as noted below.

- Subsection 6.1.1 Data Recorder Scale: This should be replaced to simply require the operator to include the range defined by the zero and span values noted above. The EPA proposed requirement seems to be based on old technology and defining this range with a specific numeric requirement (two times the standard) serves no functional purpose and unnecessarily restricts development. Modern technology based on computers and computer automation should not be restricted by this old technology-based requirement. CES contends that this specification is unnecessary and should be eliminate
- Subsection 6.1.2 CE determination: Change “zero” to “low” and “upscale” to “high”. CES believes this will result in improved clarity.
- Section 8.0 PS Tests: CES recommends the EPA consider re-writing this section around clearly defined analytical objectives of establishing the CEMS initial accuracy, precision, linearity, stability and system integrity based on some of the same principals used in EPA Method 301.

- Subsection 8.1.3 Extraction point: This should be re-written to allow for possible in situ monitoring systems that do not require sample extraction.
- Section 3.0 Definitions: A new subsection should be added defining “zero” or the bottom of the range for valid results, and instructions as to what value to use when a result falls below this defined concentration. CES recommends that “zero” be replaced with detection limit as defined by the EPA in Title 40, Appendix B Part 136 as the minimum concentration of a substance that can be measured and reported with 99% confidence that the analyte concentration is greater than zero. If the concentration is less than this concentration, the reported value should be replaced with a concentration equal to the detection limit.

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