

To: David Ailor, COETF
From: Susan Barnes and Ian Donaldson, Trinity Consultants
Date: May 3, 2024
RE: Upper Prediction Limit Calculations with Intra-Mine Variability Factor

On August 16, 2023, the U.S. Environmental Protection Agency (EPA) proposed revisions to the National Emission Standards for Hazardous Air Pollutants (NESHAP) for the Coke Ovens: Pushing, Quenching, and Battery Stacks (PQBS) source category, 40 CFR 63 Subpart CCCCC, and the NESHAP for the Coke Oven Batteries source category, 40 CFR 63 Subpart L. As part of the proposed Subpart CCCCC rule, EPA proposed standards for several previously unregulated hazardous air pollutants (HAPs), including new limits for mercury.

The Coke Oven Environmental Task Force (COETF), which represents all four companies that operate byproduct recovery coke plants in the U.S., previously commented to EPA that the proposed limits are not achievable and do not account for variability of raw materials, including mercury (Hg), fluorine, and chlorine.¹ Each coke plant has unique specifications for cokemaking, including metallurgical coal sources and properties. Thus, the potential variation of mercury, fluorine, and chlorine in coal blends introduced to the batteries over the course of normal operations is high, indicating the need for consideration of raw material (metallurgical coal) variability in establishing Hg, hydrogen fluoride (HF), and hydrochloric acid (HCl) emission limitations. The COETF has requested Trinity's assistance in determining how to address this raw material variability in calculating emission limitations for the industry, following EPA's established methodologies in other rulemakings.

In several other rulemakings, EPA has properly acknowledged that an intra-quarry variability (IQV) factor should be used to set MACT limits that reflect the effects of raw material input variability.² In these rulemakings, EPA has applied an IQV factor to EPA's computed upper prediction limit (UPL) to reflect the MACT floor emission rates.³

In this memorandum, Trinity is recommending how EPA should apply its well-established methodology for addressing mercury, fluorine, and chlorine content variability in raw materials utilized by byproduct recovery coke plants, to set MACT floor emission limits for Battery Stacks and Pushing Emission Control Devices

¹ See Comments of the COETF on the Proposed Rule National Emission Standards for Hazardous Air Pollutants for Coke Ovens: Pushing, Quenching, and Battery Stacks, and Coke Oven Batteries; Residual Risk and Technology Review, and Periodic Technology Review (EPA-HQ-OAR-2002-0085, EPA-HQ-OAR-2003-0051), October 2, 2023; "Coke Ovens RTR Proposed Rule: White Paper on Proposed Standards" submitted by the COETF to EPA on December 29, 2023; and "Compliance Schedule Concerns 020924.pdf" submitted by the COETF to EPA on February 9, 2024.

² See the final Portland Cement NESHAP, 40 CFR 63 Subpart LLL; final Brick and Structural Clay Products Manufacturing NESHAP, 40 CFR 63 Subpart JJJJJ; and Lime NESHAP, 40 CFR 63 Subpart AAAAA as proposed on February 9, 2024.

³ In this memorandum, we have revised the term "intra-quarry variability" (IQV) factor to "intra-mine variability" (IMV) factor. This simply acknowledges that the key raw materials used in byproduct recovery coke plants (blends of metallurgical coals) are sourced from deposits more commonly referred to as "mines," and not from "quarries." Nevertheless, the technical concept remains identical in that the requisite raw materials (metallurgical coals) from coal mines contain inherently varying and naturally occurring concentrations of Hg and other HAP-related constituents outside the control of the coke plant operator and which are used in existing MACT floor sources on a regular basis.

(PECs) at those plants. The variability in concentrations of HAP-related constituents in emissions from these sources is directly proportional to the concentration of these constituents that naturally occur in blends of key raw materials (i.e., metallurgical coals) used in the byproduct recovery cokemaking process. Coke producers must ensure that the coke they produce achieves the quality specifications dictated by their customers. Coke quality is determined by the coking process utilized and the coals charged to their batteries to meet battery and customer specifications. This means that they must limit the sources of coals they supply to their batteries based on the coke quality specifications imposed on them by the process and their customers, and not by the concentrations of HAP-related constituents in the raw material they utilize (metallurgical coal).

MACT Floors

The existing sources that EPA chooses as the basis for the emission limits are referred to as the “best performing sources” (aka, “MACT floor sources”). For source categories with fewer than 30 sources, the MACT floor is typically the best-performing five emission units. In the proposed rule, EPA has identified the MACT floors for each pollutant and emission unit category. In several cases, this includes the AKS-Middletown-OH facility, which is no longer operating (and, hence, which Trinity has removed from the MACT floor sources). In addition, EES Coke Battery, LLC (EES) recently submitted additional stack testing data to EPA. Trinity has incorporated these data into the analysis.

Upper Prediction Limit

Once the best performing sources are established, EPA uses a UPL calculation to establish an emission standard. The UPL represents the value below which one can expect the average of a future specified number of observations to fall, within a certain predicted confidence interval. For example, if the MACT floor consists of a data set of 1-hour runs from stack testing results for a specific type of source, a UPL can be used to predict the maximum average of any future three 1-hour runs for an emission unit in that specific source group (assuming the data set is part of the same population of data) within a specified confidence interval. The UPL takes into account the average emissions and variability in emissions within sources in the MACT floor. MACT emission limits are typically set using a 99% confidence interval.

EPA calculated the emission limits in this proposed rule using the UPL approach.⁴ However, the UPL calculation itself does not capture additional variability from naturally occurring elements (e.g., mercury, chlorine, fluorine) in the key raw materials utilized (i.e., metallurgical coals). Below we discuss the nature and methodology Trinity used, similar to that used by EPA in prior rulemakings, and how this methodology should be incorporated into a final MACT limit to account for the variability for trace amounts of naturally occurring mercury, fluorine, and chlorine in metallurgical coals utilized in byproduct recovery coke plants.

Intra-Mine Variability Factor

While the UPL calculation accounts for variability in the test run data collected (i.e., stack measurement variability, or run-to-run variability), it does not account for variability caused by differences in raw materials at a source, or between sources. EPA has accounted for variability in the mercury content in raw materials in establishing mercury standards in the cement, lime, and brick industries. Like these industries, the mercury content in key raw materials (i.e., metallurgical coals) being utilized in byproduct recovery coke plants has a direct impact on the mercury emissions from Battery Stacks and PECs. In addition, HCl and HF emissions from these emissions sources are directly related to the chlorine and fluorine contents,

⁴ Refer to equations from *Use of the Upper Prediction Limit for Calculating MACT Floors*, Tina Ndoh, EPA Office of Air Quality Planning and Standards, July 30, 2014.

respectively, in these raw materials (i.e., metallurgical coals). Therefore, incorporating this variability into the standard UPL calculation for mercury, HCl, and HF will better predict future emissions from these emission sources, helping to ensure that the calculated MACT Floor is “achieved in practice.”

In prior rulemakings, EPA accounted for an IQV factor with a “pooled” variability factor (s^2). By incorporating the IQV, the UPL equation should predict the highest average of a specified number of future runs with the defined confidence level for a source in the specified emission category utilizing raw materials with naturally varying mercury content. EPA has calculated the IQV, as follows:

$$IQV = (RSD \times Mean)^2$$

Where:

<i>IQV</i>	=	Intra-Quarry Variability factor
<i>RSD</i>	=	Relative Standard Deviation (i.e., standard deviation divided by arithmetic mean) of raw material mercury content data
<i>Mean</i>	=	Arithmetic mean of emissions data in the MACT floor

In this analysis, Trinity has used this same formula for calculating an Intra-Mine Variability (IMV) factor for normally distributed data sets. For those MACT floor data sets for the proposed rule that are lognormally distributed, Trinity has revised this IQV formula and calculated an IMV factor for the byproduct recovery coke industry, as follows:⁵

$$IMV = (RSD \times Logmean)^2$$

Where:

<i>IMV</i>	=	Intra-Mine Variability factor
<i>RSD</i>	=	Relative Standard Deviation (i.e., standard deviation divided by arithmetic mean) of coal mercury content data
<i>Logmean</i>	=	Arithmetic mean of log-transformed emissions data in the MACT floor

Mercury

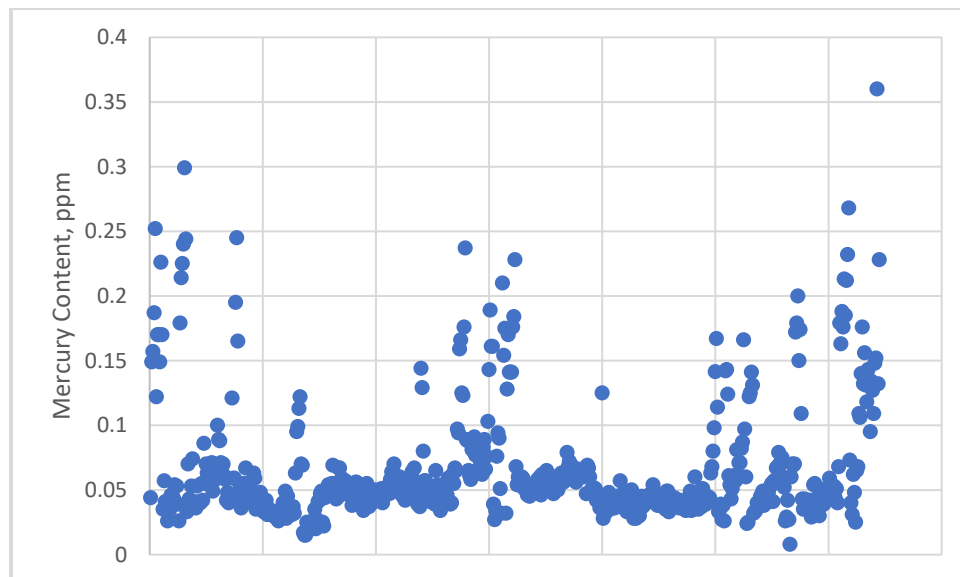
In this analysis, Trinity has evaluated the mercury content of raw materials (metallurgical coals) from three byproduct recovery coke plants: Monessen, Clairton, and Burns Harbor. These plants were chosen because they are in both the Battery Stack and PEC MACT floors and because metallurgical coal mercury content data are available.⁶ The data set consists of 646 coal samples analyzed for mercury from 2015 and 2023, where each sample was associated with a specific mine (data from 65 coal mines included in data set). This is a very robust data set, with significantly more data than were used in establishing MACT limits via an IQV for other industries. The brick industry, for example, provided 167 samples nationwide, with the final IQV

⁵ In byproduct recovery coke plants, the critical parameters of coal play a pivotal role in determining coke quality, yield, and performance. Proximate and petrographic analyses, coking properties, ash fusion temperature, size, grindability, and thermal properties are among the key factors that must be carefully evaluated and controlled. By understanding and optimizing these parameters, these plants can produce coke that will enhance the efficiency, sustainability, and competitiveness of steelmaking processes, as well as high-quality coke for diverse industrial applications.

⁶ The Warren plant uses the same coal sources as Monessen and these data are also representative for that plant. This plant is not in the MACT Floor for mercury, although would be subject to the proposed emission limits.

being based on seven samples from four plants.⁷ Mercury content of metallurgical coals used in this analysis ranges from 0.008 to 0.360 parts per million (ppm), as shown in Figure 1. This wide range clearly indicates the need for consideration of coal mercury variability in establishing mercury emission limitations.

Figure 1. Mercury Content of Metallurgical Coals



Due to differences in the available precedents for setting the relative standard deviation (RSD) upon which an IQV is set, two different methods are provided in this memorandum: (1) evaluating the RSD of all available data, and (2) evaluating the RSD of the average mercury content of each coal mine. These are discussed in more detail below. Calculations supporting this analysis are provided in the attached spreadsheet.

Option 1: IMV Factor Based on RSD of All Available Data from MACT Floor Sources

The most recent proposed version of the Lime MACT⁸ set an IQV using the mercury content of individual samples for two plants in the MACT floor. To follow this precedent, under option 1, Trinity has calculated the RSD as follows:

$$RSD = \frac{SD}{Mean}$$

Where:

<i>RSD</i>	=	Relative standard deviation
<i>SD</i>	=	Standard deviation of 646 mercury content values
<i>Mean</i>	=	Arithmetic mean of 646 mercury content values

The RSD calculated in this manner is 0.70.

⁷ EPA-HQ-OAR-2013-0291-0660, Appendix E.

⁸ EPA-HQ-OAR-2017-0015-0172.

Option 2: IMV Factor Based on RSD of Coal Mine Averages from MACT Floor Sources

Mercury limits in the Brick MACT were established based on the RSD between plant average mercury content.⁹ Following this methodology for byproduct recovery coke plants is best achieved by calculating the RSD as follows:

$$RSD = \frac{SD_{AVG}}{Mean_{AVG}}$$

Where:

RSD = Relative standard deviation
SD_{AVG} = Standard deviation of average mercury content of 65 mines
Mean_{AVG} = Arithmetic mean of average mercury content of 65 mines

The RSD calculated in this manner is 0.72.

Proposed Emission Limits

Table 1 below shows the resulting UPL-based mercury limitations after updating the MACT floor and adding an IMV as described above.

Table 1. Proposed Mercury Limits

Emission Unit	Emission Limitation Using Option 1 RSD (lb/ton coke)	Emission Limitation Using Option 2 RSD (lb/ton coke)
Existing Battery Stacks	1.3E-4	1.3E-4
Existing PEC	3.6E-6	3.7E-6

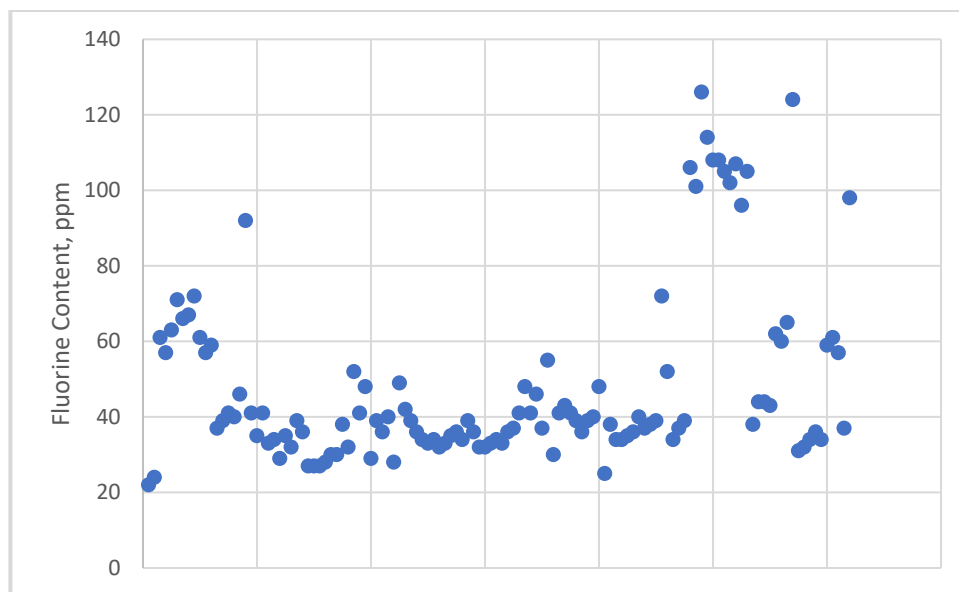
Acid Gases

In this analysis, Trinity has evaluated the fluorine and chlorine content of raw materials (metallurgical coals) from three byproduct recovery coke plants: Monessen, Clairton, and Burns Harbor.¹⁰ These plants were chosen because they are in both the Battery Stack and PEC MACT floors and because metallurgical coal fluorine and chlorine content data are available. The data set consists of 124 coal samples analyzed for fluorine and chlorine content, where each sample was associated with a specific coal mine (data from 33 coal mines included in data set). This is a very robust data set, with significantly more data than were used in establishing MACT limits via an IQV for other industries.

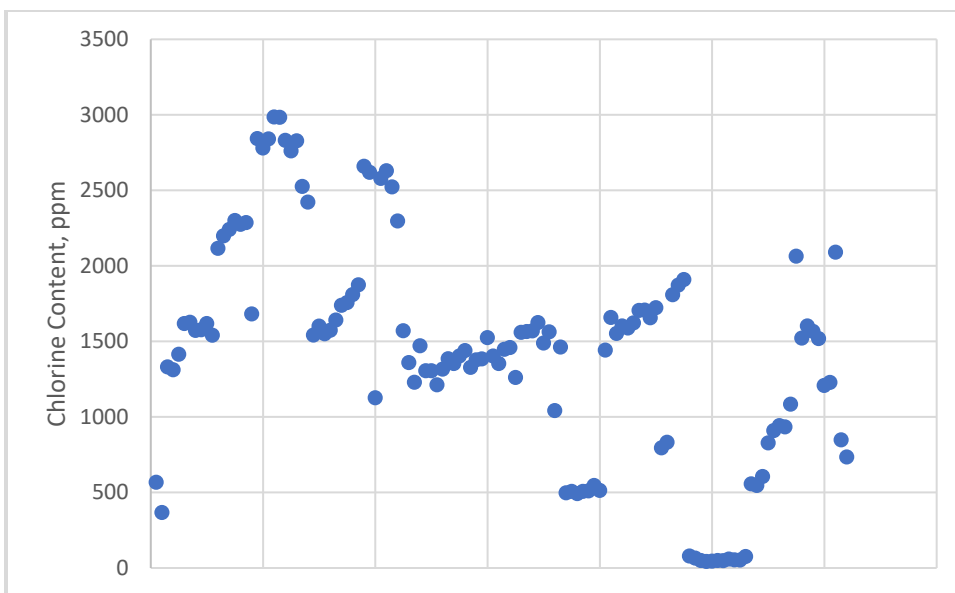
Fluorine content of metallurgical coals used in this analysis ranges from 22 to 126 ppm as shown in Figure 2.

⁹ EPA-HQ-OAR-2013-0291-0660, Appendix E.

¹⁰ The Warren plant uses the same coal sources as Monessen and these data are also representative for that plant. This plant is not in the MACT Floor for acid gases, although it would be subject to the proposed emission limits.

Figure 2. Fluorine Content of Metallurgical Coals

Chlorine content of metallurgical coals used in this analysis ranges from 42 to 2,985 ppm as shown in Figure 3.

Figure 3. Chlorine Content of Metallurgical Coals

As shown in Figures 2 and 3, data for both fluorine and chlorine show high variability. In setting an acid gas emission limit, which is the sum of HF and HCl, this variability must be properly addressed in order to ensure that the MACT floor is representative of raw material variability and that emission limits are achievable.

As with mercury, Trinity calculated the RSD of the fluorine and chlorine content using two different methodologies, as shown in Table 2.

Table 2. Fluorine and Chlorine Concentration Relative Standard Deviation

	Fluorine	Chlorine
Option 1 – RSD of All Available Data	0.49	0.51
Option 2 – RSD of Mine Averages	0.45	0.46

Proposed Emission Limits

Using the RSDs from Table 2 and the updated MACT floors, Trinity calculated UPLs separately for HF and HCl and then summed them to determine an acid gas emission limit. Tables 3 and 4 below show the results of this analysis for battery stacks and PECs, respectively.

Table 3. Proposed Acid Gas Limits for Battery Stacks

	IMV Based on Option 1 RSD (lb/ton coke)	IMV Based on Option 2 RSD (lb/ton coke)
UPL for HF	1.36E-3	1.35E-3
UPL for HCL	1.50E-1	1.45E-1
Proposed Acid Gas Limit	1.6E-1	1.5E-1

Table 4. Proposed Acid Gas Limits for PECs

	IMV Based on Option 1 RSD (lb/ton coke)	IMV Based on Option 2 RSD (lb/ton coke)
UPL for HF	6.01E-4	5.94E-4
UPL for HCL	1.42E-2	1.33E-2
Proposed Acid Gas Limit	1.5E-2	1.4E-2

Hydrogen Cyanide

Trinity did not calculate an IMV factor for hydrogen cyanide (HCN). However, the EES data did include stack testing data for HCN. Accordingly, Table 5 provides updated UPL calculations to reflect the update to the MACT floor.

Table 5. Proposed HCN Limits

Emission Unit	Emission Limitation (lb/ton coke)
Existing Combustion Stacks	3.9E-2
Existing PEC	3.2E-2

Conclusion

Mercury, HF, and HCl emissions from Battery Stacks and PECs at byproduct recovery coke plants are directly related to the mercury, fluorine, and chlorine content, respectively, of the key raw materials (metallurgical coals) required to produce quality coke. EPA's established methodology for incorporating an IQV into MACT standards for other industries should be applied to the proposed mercury limits for coke Battery Stacks and PECs.

Using the specific statistical precedents set in the final Brick and proposed Lime MACT rulemakings, Trinity has presented two options above for calculating an RSD which could be added to UPL calculations to account for intra-mine variability (IMV) in mercury, fluorine, and chlorine concentrations. Accounting for this variability is essential to ensure that the MACT floor coke plants can continuously comply with the established emission limits over time, because the mercury, fluorine, and chlorine content varies in the raw materials (metallurgical coals) those plants must use to produce quality coke.

EPA should adjust the MACT floor source category data to account for the fact that the AKS-Middletown-OH facility has ceased operations and to reflect newly available stack testing data for EES. EPA should then follow the precedent in the Brick MACT of choosing the higher RSD of the evaluated options and incorporating this RSD into an IMV factor to the UPL calculation to establish the final MACT floor limits for Battery Stacks and PECs.

A summary of the revised MACT floor emission limits for existing Battery Stacks and PECs calculated using an IMV is provided in Table 6 below.

Table 6. Summary of Revised Emission Limits for Existing Sources

Pollutant	Battery Stacks Emission Limit (lb/ton coke)	PEC Emission Limit (lb/ton coke)
Mercury	1.2E-4	3.7E-6
Acid Gases	1.6E-1	1.5E-2
Hydrogen Cyanide	3.9E-2	3.2E-2