

TOXICITY REVIEW OF METALS EMISSIONS FROM COAL-FIRED POWER PLANTS



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List of Abbreviations and Acronyms

ACS	American Cancer Society
ADD	Average Daily Dose
ADHD	Attention Deficit Hyperactivity Disorder
AML	Abandoned Mine Lands
ASCC	Alaska Systems Coordinating Council
BIT	Bituminous
CAA	Clean Air Act
CAMR	Clean Air Mercury Rule
CAP	Criteria Air Pollutants
CFPP	Coal-Fired Power Plants
EGU	Electric Generating Unit
EIA	Energy Information Administration
EPA	Environmental Protection Agency
ESP	Electrostatic Precipitators
FRCC	Florida Reliability Coordinating Council
HAP	Hazardous Air Pollutant
HICC	Hawaiian Islands Coordinating Council
ICR	Information Collection Request
LIG	Lignite
MACT	Maximum Achievable Control Technology
MATS	Mercury and Air Toxics Standards
MRO	Midwest Reliability Organization
NEI	National Emissions Inventory
NERC	North American Electric Reliability Corporation
NESHAP	National Emission Standards for Hazardous Air Pollutants
NHANES	National Health and Nutrition Examination Survey
NPPC	Northeast Power Coordinating Council
NSPS	New Source Performance Standards
OMB	Office of Management and Budget
PAH	Polynuclear Aromatic Hydrocarbons
PC	Pulverized Coal
PM	Particulate Matter
RC	Refined Coal
RFC	Reliability First Corporation
RTR	Risk and Technology Review
SERC	Southeastern Electric Reliability Corporation
SPP	Southwest Power Pool
SUB	Sub-bituminous
TOSHI	Target Organ Specific Hazard Index

USGS	U.S. Geological Survey
VOC	Volatile Organic Compound
TRE	Texas Regional Entity
WECC	Western Electricity Coordinating Council

List of Units of Measurement

PM ₁	Particulate Matter 1 µm
PM _{2.5}	Particulate Matter 2.5 µm
PM ₁₀	Particulate Matter 10 µm
MWe	Megawatts Electric
tpy	tons per year
µm	micrometer

List of Metals Reviewed and Chemical Element Symbol

As	Arsenic
Ba	Barium
Be	Beryllium
Bi	Bismuth
Cd	Cadmium
Cl	Chloride
Co	Cobalt
Cr	Chromium
Cs	Cesium
Cu	Copper
Ga	Gallium
Ge	Germanium
Hg	Mercury
Li	Lithium
Mn	Manganese
Mo	Molybdenum
Nb	Niobium
Ni	Nickel
Pb	Lead
Rb	Rubidium
Sb	Antimony
Sc	Scandium
Se	Selenium
Sr	Strontium
Th	Thorium
Tl	Thallium
U	Uranium
V	Vanadium
Y	Yttrium
Zn	Zinc

Executive Summary

This report is a summary of recent research findings regarding coal-fired power plant non-mercury metal emissions for the purpose of informing current U.S. Environmental Protection Agency (EPA) rulemakings for coal-fired power plant hazardous air pollutant regulations. This report provides a context for understanding the potential for non-mercury metals in coal-fired power plant emissions to have human health outcomes. It is anticipated that this information will be used in ongoing and future rulemakings, to support EPA regulation of coal-fired power plants' emissions of hazardous air pollutants and to help justify strengthened, protective limits on coal-fired power plants' emissions of non-mercury metals. Although the extent of exposures to these metals from the combustion of coal in power plants remain uncertain, and while the health effects of these metal mixtures in humans continue to be investigated, there is sufficient evidence from studies on exposures to emissions from mine wastes and other similar sources to conclude that these metals have serious and wide-ranging human health impacts, individually and in mixtures. Accordingly, reductions in emissions of these metals under the current MATS rule have produced substantial unquantified health benefits and greatly curtailed harmful metals that Congress specifically targeted in section 112, supporting EPA's decision to regulate these emissions under section 112 of the Clean Air Act. Furthermore, additional reductions of coal-fired power plants' emissions of these toxic metals would yield still unquantified benefits that would support strengthening the rule.

Direct particulate emissions from coal combustion have metal content that mirrors the metal profile found in the source coal. The specific metal content in source coal varies by geographical region and type of coal used. Although many metals found in coal have well-established toxicity profiles, regulation has previously focused on the benefits of reducing mercury as the combustion product for regulation. In addition to mercury, however, coal combustion is a source for airborne emissions of arsenic, lead, chromium, nickel, cadmium, and a host of other metals including uranium that have harmful health impacts.

This report is structured to provide the following information to address data gaps in the 2011 EPA Mercury and Air Toxics Standards (MATS) rule on hazardous air pollutant (HAP) emissions from coal- and oil-fired power plants:

- A summary of the 2011 MATS rule and data gaps. (Section 1)
- A description of the different types of coal and associated metal profiles. (Section 2)
- Consideration of multiple exposure pathways. While airborne emissions are the primary focus for exposure from power-plant emissions, data confirm that these airborne emissions also settle to impact soil and surface water, and move through the environment into food chains, leading to the possibility of ingestion exposures, especially soil ingestion in the case of young children. (Section 3)
- Consideration of both single and multiple metal exposure outcomes. Since a review of metal-emissions toxicity in 2011, there have been additional studies of the behavior of metals in the environment and of the resultant toxicity associated with exposures to both individual metals and metal mixtures. (Section 3)
- Consideration of recent findings on metal inhalation exposure health outcomes from sources that are different but comparable to coal combustion. There is little existing data

on health outcomes from exposure to metals from coal-fired power plant emissions. Particularly relevant is research focused on community exposures to abandoned mine waste which represents a similar composition of metals as found in coal, and that can be aerosolized and move through similar exposure pathways. Also relevant are studies of soils in areas with high naturally-occurring levels of arsenic which, like mine tailings, can be aerosolized. (Section 3)

- Consideration of recent studies related to progressively lower-dose toxicity and the toxicity of complex metal mixtures, which are important to consider in regulating these metals as power plant emissions. (Section 3)
- Summary and conclusions. (Section 4)

1. Introduction and Summary of Federal Supporting Docket¹

1.1 Introduction and Focus of this Report

This report summarizes recent research findings regarding coal-fired power plant non-mercury metal emissions for the purpose of informing current U.S. Environmental Protection Agency (EPA) rulemakings for coal-fired power plant hazardous air pollutant emissions in order to provide a context for understanding the potential for non-mercury metals in coal-fired power plant emissions to have human health outcomes. The primary focus of this report is on metal contaminants emitted from coal-fired power plants, and it therefore excludes the HAP classes of acid gases, dioxins and furans, polynuclear aromatic hydrocarbons (PAH), and volatile organic compounds (VOC) for which substantial toxicity documentation exists. The focus on non-mercury metals/metalloids, and radioisotopes allows us to consider specific data gaps in the existing federal supporting docket. By referencing information from a U.S. Geological Survey (USGS) report (Affolter et al. 2011) and building on a previous environmental health report on HAP emissions from coal-fired power plants (Spengler et al. 2011), we will identify metals that are not considered in recent risk assessments of EGU emissions conducted by the EPA. In subsequent sections, we will discuss the health implications of emissions of HAP metals from coal-fired power plants that are therefore ignored in the EPA's 2018 residual risk assessment. It is noted that in 2020, EPA issued an Appropriateness Rescission Rule that reversed the 2000 determination that it was appropriate to regulate coal- and oil-fired electric generating units under section 112 of the Clean Air Act (a determination that EPA reaffirmed in 2012 and 2016), but left the MATS rule in place concluding that the 9-in-1 million risk of cancer to the most exposed individual found in the risk review would preclude delisting the source category under section 112 (85 Fed. Reg. at 31,312).

1.2 Federal Supporting Docket

¹The EPA established two rulemaking dockets to support promulgating the MATS rule: Docket ID. No. EPA-HQ-OAR-2011-0044 (NSPS action) and Docket ID No. EPA-HQ-OAR-2009-0234 (NESHAP action). Previous dockets were incorporated into the final MATS docket, including the Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units—Final Report to Congress (February 1998); and the Mercury Study Report to Congress (December 1997). All documents in the dockets can be searched for with the docket ID number at www.regulations.gov. The MATS docket can be accessed at www.regulations.gov/document/EPA-HQ-OAR-2009-0234-20450.

The following sections offer an overview of the federal supporting docket for the U.S. EPA’s 2012 MATS rule on HAP emissions from coal- and oil-fired “electric generating units” (EGUs), as defined in section 112 of the Clean Air Act (CAA), which requires EPA to establish national emission standards for hazardous air pollutants (NESHAP). EGUs (or, more precisely, electric utility steam generating units) comprise a source category of coal- and oil-fired power plants with “a fossil fuel-fired combustion unit of more than 25 megawatts electric (MWe) that serves a generator that produces electricity for sale” (USEPA, 2018).

In recent reviews for the MATS rulemaking process, the EPA assessed 322 EGU facilities in the United States that emit approximately 5,100 tons of HAP per year (USEPA, 2018). The number of EGUs assessed, however, only accounts for about half of the total power plants covered by these standards in 2018. In addition to the federal docket, the EPA has a MATS webpage that describes the science and technology of “Cleaner Power Plants,” in which they estimate that there were 1,400 coal- and oil-fired EGUs at 600 power plants covered by these standards when the rule was promulgated. In an infographic, the EPA identifies power plants as the dominant emitters of mercury (50 percent), acid gases (77 percent), and many toxic metals (22-62 percent) in the United States (see Figure 1).² Of these 600 power plants in the EGU source category when EPA developed the MATS rule, over 440 were coal-fired (Spengler et al. 2011). Coal combustion accounted for about 45% of the electricity produced in the U.S. circa 2010 (Spengler et al. 2011). In 2020, coal accounted for only 19 percent of electricity generated in the United States, as coal electric generation declined by about 40 percent with 289 coal-fired power plants closing. However, 241 remain in operation.³

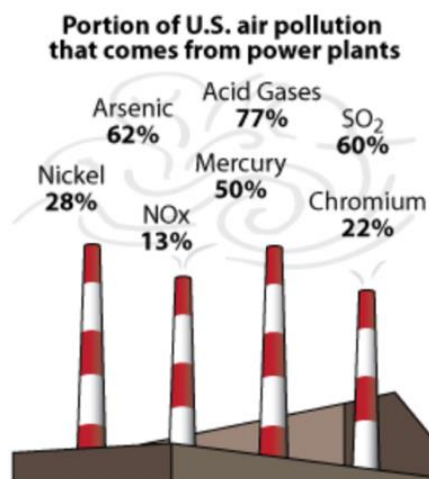


Figure 1. Screenshot of EPA Infographic: “Portion of U.S. air pollution that comes from power plants.” See <https://www.epa.gov/mats/cleaner-power-plants>; accessed September 29, 2021.

1.3 Data Gaps in the MATS Rulemaking Docket

There are six primary data gaps that we identified in the MATS rulemaking docket. These data gaps have all been addressed to some extent in toxicological literature published after 2011. Indeed, there currently exist relevant studies of metal exposure and toxicity assessments among populations living near abandoned mine lands (AMLs) and mill tailings sites, which can offer a deeper understanding of multiple pathways of exposure to metal mixtures and their environmental health implications for federal risk assessments and regulation of coal-fired power plants. The data gaps include:

² <https://www.epa.gov/mats/cleaner-power-plants>; accessed September 29, 2021.

³ <https://www.eia.gov/energyexplained/electricity/electricity-in-the-us.php>; accessed September 30, 2021.

1. **EPA only accounted for a small proportion of metals that are known to occur in various forms of particulates from coal-fired power plants in the MATS analysis.**

A comparison of the extracted data from the EPA’s summary of emissions with a USGS detailed report on coal-fired power plant emissions (Affolter et al. 2011) shows that only slightly more than one third of the metals that are known to occur in various forms of particulate matter emitted from coal-fired power plants are accounted for in EPA’s analysis in the MATS rulemaking (Table 1). Specifically, the EPA risk assessment accounts for 11 metals while the USGS report identified 30 different metals in coal-fired power plant (CFPP) emissions (Affolter et al. 2011). The decision to focus on 11 metals was made without consideration of more recent evidence on health effects of metal mixtures (discussed in Section 3). While much of the newer data presented is based on metal mixtures from mine waste sources, the similarities in metal profiles, respirable particulates, and multiple environmental exposure pathways between the mining and coal combustion sources make inclusion of these data relevant and important. One of the consequences of the EPA taking a narrow focus on a handful of metals is that consideration has not been given to potential multiple metal interactions and impacts on health that may occur with exposure to the full spectrum of metals present in the coal combustion process. Similarly, the EPA’s 1998 HAP study for Congress prioritized 14 of 67 HAPs including 6 metals: arsenic, beryllium, cadmium, lead, manganese, and mercury. The remaining 53 HAPs were not evaluated beyond the screening assessment.

Table 1. Metal Emissions (Not) Identified in the MATS Docket.

Metal Emissions Identified in the MATS Docket	Metal Emissions Not Identified in the MATS Docket
As, Be, Cd, Cr, Co, Hg, Mn, Ni, Pb, Sb, Se	Ba, Bi, Cl, Cr, Cs, Cu, Ga, Ge, Li, Mo, Nb, Rb, Sc, Sr, Th, Tl, U, V, Y, Zn

2. **There is a lack of consideration of size and chemical composition of particulates in relation to their toxicity** (see subsection 1.4.4 of the Regulatory Impact Analysis).
3. **The EPA’s cancer and non-cancer risk assessments do not seriously consider the full range of metal emissions known to occur in coal combustion and their various possible interactions in producing specific health outcomes.** Given recent research in the toxicology of metal mixtures from mining, EPA should consider new evidence of non-cancer health risks, which will be discussed in the following sections. EPA does attempt to combine risks across multiple carcinogens and non-cancer hazards, albeit using dated mixtures guidelines (USEPA 1986; USEPA 2000). Because the docket does not adequately address the interacting health effects of various forms of metal mixtures, it ignores a paradigm shift in environmental health science that goes beyond single-pollutant biomedical models (Breton and Farzan 2021; Keil et al. 2021; Tanner et al. 2020).
4. **Consideration is not given to the radioisotopes of radium, thorium, and uranium, which are radioactive and chemically toxic, and have long been known to occur in particulate emissions from certain coal-fired power plants (Affolter et al. 2011; Lee et al. 1975; Spengler et al. 2011).** Although radioisotopes are considered in the 1998 HAP

report to Congress, they are left out of the EPA 2018 Residual Risk Assessment (see Table 2). Additionally, the 1998 report to Congress considers the radiation risks of radioisotopes of uranium, thorium, and potassium but not their chemical toxicity as heavy metals.

5. **In addition to compounds of nickel and cobalt emitted from oil-fired power plants, compounds of arsenic emitted from coal- and oil-fired power plants are deemed risk “drivers” by the EPA, yet EPA does not investigate the risks of specific compounds of arsenic.** According to the 2018 Residual Risk Assessment : “EPA’s Cancer Guidelines express a preference for the use of reliable, compound-specific, biologically-based risk models when feasible; however, such models are rarely available” (USEPA, 2018).
6. **The docket lacks full consideration of compounding environmental health risks and vulnerabilities due to multiple, overlapping sources of toxic particulates among other emissions, and therefore does not account for environmental health disparities and injustices (Tessum 2021).**

In the non-Hg case study (Strum et al. 2011), the authors note that they do not cover all facilities in the category, and their assessment does not include potential impacts from different EGU facilities that overlap one another, or other possible sources of emissions. Their case studies look at facilities in isolation. For this reason, Strum and colleagues conclude that “the maximum risk estimates from the case studies may be underestimating actual maximum risks” (USEPA, 2011). This implies that there is an absence of information on the risks of compounding environmental health impacts from multiple sources.

The clear implication of these data gaps, individually and in combination—when considered in light of the findings discussed in the present report—is that there are significant risks of toxic metal emissions from coal-fired power plants (together with emissions from other sources) that EPA has not adequately considered in its rulemakings on this subject. Conversely, there are substantial benefits from reducing emissions of these metals that the MATS rule is currently producing, and that a strengthened rule would amplify, but that go unacknowledged in the record. Although our understanding of the severity and range of human health impacts that these metals inflict is evolving, as discussed below, it is clear that they harm human health through similar exposure pathways as result from coal combustion in power plants. Moreover, Congress included these metals on the list of hazardous air pollutants that EPA is required to reduce, through regulation, to the maximum degree achievable. The current approach—to assume that these metals do not have significant health effects until those effects are established in the scientific literature—contravenes the Clean Air Act’s precautionary directives and disregards findings that these metals cause various health harms in a range of settings, as discussed below.

1.4 Overview of the Existing Federal Docket

A general overview of four documents in EPA’s MATS docket is provided to identify gaps in the data on emission sources through comparative analysis with existing literature in the environmental health sciences on source, exposure, and health effects from metals emissions of CFPPs. These include:

1. **Residual Risk Assessment** for the Coal- and Oil-Fired EGU Source Category in Support of the 2019 Risk and Technology Review Proposed Rule (December 2018)

2. **The MATS Rule:** 40 CFR Parts 60 and 63 National Emission Standards for Hazardous Air Pollutants from Coal- and Oil-Fired Electric Utility Steam Generating Units and Standards of Performance for Fossil-Fuel-Fired Electric Utility, Industrial-Commercial-Institutional, and Small Industrial-Commercial-Institutional Steam Generating Units; Final Rule (Federal Register, Vol. 77, No. 32, February 16, 2012)
3. **Memorandum: Non-Hg Case Study** Chronic Inhalation Risk Assessment for the Utility Maximum Achievable Control Technology (MACT) Appropriate and Necessary Analysis (Strum et al. 2011)
4. **Regulatory Impact Analysis** for the Final Mercury and Air Toxics Standards (USEPA, 2011).

Each of these four documents will be discussed in the following subsections. These subsections highlight the specific metals emissions of concern in the MATS docket, as well as potential toxic metals emissions not included, and subsequent limitations and data gaps in the 2018 risk assessment, which EPA subsequently relied upon to decline to strengthen MATS. The 1998 HAP report to Congress is omitted here because it does not change the fact that EPA did not consider the significance of recent toxicological research on the health effects of metal mixtures known to occur in both mining processes and particulate matter from coal combustion. It is noted that the trace metals examined in the 1998 HAP Emissions Report to Congress for coal-fired units include arsenic, beryllium, cadmium, lead, manganese, and mercury.

1.4.1 Residual Risk Assessment for MATS

There is a multi-stage regulatory process for addressing the emissions of HAPs from EGUs. The EPA completed the first stage in 2012, by confirming that it was appropriate and necessary to regulate HAP emissions from EGUs and promulgating the MATS which are the technology-based NESHAP for the coal- and oil-fired EGU category. Unless EPA decides to conduct a review of the standards considering developments in control techniques, the second stage of the regulatory process is the residual risk review, in which the EPA assesses the health and environmental risks that remain after establishing the standards. Under CAA section 112(d)(6) and 112(f), the EPA is required to review and revise, if necessary, the MACT standards based on a residual risk and technology review (RTR).⁴ The 2018 Residual Risk Assessment for the MATS rule resulted in a 990-page document detailing the results of the risk review. At the core of the document is the 62 pages that contain the executive summary, introduction, methods, risk results for the coal- and oil-fired EGU source category, and general discussion of uncertainties in the risk assessment. The remainder of the document includes references, 11 appendices, several tables, and other supplementary data.

Most relevant to this review of potential data gaps in the docket is Table 3.1-1 (EPA 2018, 38-39), which summarizes emissions, as well as Appendix 1: Emissions Inventory Support Document (68-91). To create Table 2, we extracted the metal emissions in tons per year (tpy) from the EPA's summary of emissions from 322 facilities in the EGU source category for the RTR. To put this Table into context, the EPA estimates the total HAP emissions from coal- and oil-fired EGUs at

⁴ See <https://www.epa.gov/mats/final-revised-supplemental-finding-and-results-residual-risk-and-technology-review>, accessed September 20, 2021.

approximately 5,100 tpy while the metals reported in Table 2 account for 332 tpy. The metals emitted in the largest quantities are individual compounds of selenium, manganese, nickel, and chromium. EPA also calculated the emissions of individual compounds of lead, arsenic, mercury, and cadmium. The HAP risk “drivers” identified by EPA account for 90% of the cancer risk and include nickel and cobalt compounds from oil fuel sources, and arsenic compounds from oil- and coal-fired units (USEPA, 2018).

The following data gaps are noted:

- Table 2 highlights that only 11 metals were considered in the EPA review, leading to two data gaps. First, as noted above, although the review addresses the risks associated with each individual metal of concern, it does not adequately consider risks from exposure to metal mixtures of the 11 metals. The EPA review also does not consider additional metals that are known to occur in coal combustion.
- Consideration was given to the risks of radioisotopes of uranium-238, thorium-232, and potassium-40 in the EPA HAPs report to Congress (1998), but the chemical toxicity of uranium, thorium, and potassium as heavy metals was not considered; and there is no consideration of radium, aside from its intermediate decay product of radon-222.
- In the 2018 Residual Risk Review (USEPA, 2018), EPA followed dated mixture guidelines (USEPA 1986; USEPA 2000) by “aggregating effects of different substances in specific and limited ways” using a target organ specific hazard index (TOSHI). The review of recent toxicological literature presented in this paper indicates several ways to improve the guidelines for metal mixture research and reassess the risks from exposure to metal particulates emitted by coal combustion.

Table 2. Metal emissions from 322 coal- and oil-fired power plants in the EGU source category in tons per year (tpy), extracted from Table 3.1-1: Summary of Emissions from Coal- and Oil-Fired EGU source category and Dose-Response Values Used in the Residual Risk Assessment (USEPA, 2018). Metals emissions were selected from a list of total emissions including non-metals and associated risk assessments.

Specific Metal Compound	Emissions (tpy)
Selenium (Se)	68
Manganese (Mn)	46
Nickel (Ni)	39
Chromium (Cr) (III & VI)	38(III)/5(VI)
Lead (Pb)	6
Cobalt (Co)	6
Arsenic (As)	5
Antimony (Sb)	4
Mercury (Hg)	3
Cadmium (Cd)	0.8
Beryllium (Be)	0.4

1.4.2 The MATS Rule

The decision as to whether it is “appropriate and necessary” to regulate coal- and oil-fired power plants has a long history, leading up to the MATS rule of 2012. That history dates back to December 2000, when EPA determined it was “appropriate and necessary” to regulate coal- and oil-fired EGUs under CAA section 112 and listed the industry for regulation under section 112(d). This is referred to as “the A&N Finding.” In 2005, EPA published a final rule that purported to reverse the A&N Finding, attempted to remove coal- and oil-fired power plants from regulation under CAA section 112, and established the Clean Air Mercury Rule (CAMR) under CAA section 111. However, in 2008, in the case of *New Jersey v. EPA*, the U.S. Court of Appeals for the D.C. Circuit ruled that the attempted delisting of the source category was precluded by CAA section 112(c) and vacated the 2005 rule.

The timeline of recent historical events begins in 2011, when the EPA reaffirmed and updated the record underlying the initial A&N Finding of 2000, and finalized the MATS rule in 2012. The MATS rule was again contested and in 2015 was sent back to the Agency to include cost considerations to justify the A&N Finding. The finding was reaffirmed by EPA in 2016, taking into account several metrics of the cost of regulating coal- and oil-fired EGUs under section 112. In 2020, EPA again purported to rescind the appropriateness determination, but it retained the MATS.⁵ This timeline is summarized below:

- **On May 3, 2011**, EPA proposed NESHAPs for coal- and oil-fired EGUs, and standards of performance for fossil fuel-fired electric utility steam generating units under the authority of CAA sections 111 and 112 (76 FR 24976).
- **On February 16, 2012**, the final MATS and the Utility New Source Performance Standards (NSPS) rules were published in the Federal Register. This marked the first time that federal limits on emissions of HAPs from coal- and oil-fired power plants were imposed (Spengler et al. 2011). Under CAA section 112, EPA established NESHAPs for coal- and oil-fired EGUs which require the application of MACT. MACT standards are based on emissions levels currently achieved by the “best-controlled and lower-emitting sources in an industry.” At the time, EPA acknowledged that the MATS rule protects air quality and promotes public health by reducing emissions of HAPs listed in CAA section 112(b)(1).
- **The MATS rule was contested in 2015**, and a D.C. Circuit decision upholding the A&N Finding underlying MATS was appealed to the Supreme Court, which ruled in *Michigan v. EPA* that EPA neglected to “consider costs” when determining whether it is “appropriate and necessary” to regulate HAPs from coal- and oil-fired power plants.
- **In the 2016 Supplemental Finding**, the EPA affirmed that it was “appropriate and necessary” to regulate EGUs under section 112 of the CAA, considering costs.⁶

⁵ The fact sheet and the final report can be accessed here: <https://www.epa.gov/mats>; accessed September 22, 2021.

⁶ See <https://www.epa.gov/mats/regulatory-actions-final-mercury-and-air-toxics-standards-mats-power-plants>, accessed September 20, 2021.

- **In 2018**, the EPA conducted a Residual Risk and Technology Review, and proposed in 2019 to find both that it was not appropriate to regulate the industry considering costs, but also to retain the MATS rule.
- **May 22, 2020**, EPA purported to rescind the appropriateness determination, but retained the MATS emission standards. Based on the 2018 risk review, however, EPA decided not to strengthen MATS. The MATS rule continues to be in effect. Mercury and other HAP standards are applicable to coal- and oil-fired EGUs under section 112 of the CAA.

1.4.2.1 MATS Rule in Relation to Mercury and Non-Mercury Metals and Metalloids

A key driver behind EPA’s recurrent finding that it is “appropriate and necessary” to set HAP emission standards for coal-fired power plants is that they are “by far the largest anthropogenic source of Hg in the U.S.” An EPA estimate based on data from 2005 is that EGUs accounted for 50 percent of total domestic anthropogenic mercury emissions. Coal-fired power plants, in particular, accounted for 46% of mercury emissions in 2007 (USEPA, 2007). Mercury emissions from coal-fired power plants decreased in the U.S. by 85% from 92,000 pounds in 2006 to 14,000 pounds in 2016, when states began to implement MATS (USEPA, 2018). Coal-fired power plant emissions include 84 of the 187 HAPs (USEPA, 2007). According to Spengler and colleagues, coal-fired power plants produce total emissions of 386,000 tons of HAPs annually, or about 40% of all HAP releases from any point source, “more than any other point source category” (Spengler et al. 2011). Coal- and oil-fired EGUs are a major source of metal(loid) HAP emissions (Table 3). For arsenic, EGUs are responsible for 62% of total emissions, 39% of total cadmium emissions, 22% of total chromium emissions, 28% of total nickel emissions, and 83% of total selenium emissions (USEPA, 2012).⁷ In the 2018 residual risk assessment for the MATS rule, EPA concluded that mercury (Hg) emissions from EGUs pose a risk to 29 percent of modeled watersheds (USEPA, 2018) indicating the potential for environmental mobility and quantifiable risk of Hg emissions to public health via multiple exposure sources.

Table 3. EPA estimation of the percentage of metals emissions from U.S. EGUs out of total domestic anthropogenic emissions in 2005 (USEPA, 2012).

Metal	HAP	Percentage of Total Emissions
Mercury (Hg)	Mercury	50%
Arsenic (As)	Non-Mercury Metal	62%
Cadmium (Cd)	Non-Mercury Metal	39%
Chromium (Cr)	Non-Mercury Metal	22%
Nickel (Ni)	Non-Mercury Metal	28%
Selenium (Se)	Non-Mercury Metal	83%

⁷ U.S. Environmental Protection Agency-Science Advisory Board (U.S. EPA-SAB). 2011. Peer Review of EPA’s Draft National-Scale Mercury Risk Assessment. EPA-SAB-11-017. [http://yosemite.epa.gov/sab/sabproduct.nsf/BCA23C5B7917F5BF8525791A0072CCA1/\\$File/EPA-SAB-11-017-unsigned.pdf](http://yosemite.epa.gov/sab/sabproduct.nsf/BCA23C5B7917F5BF8525791A0072CCA1/$File/EPA-SAB-11-017-unsigned.pdf); accessed September 22, 2021.

1.4.3 The Non-Hg MATS Analysis

Information on non-Hg metal(oids) has been derived from analysis of data from the 2005 National Emissions Inventory (NEI) Data, and emissions data from the “Information Collection Effort for New and Existing Coal- and Oil-fired Electric Utility Steam Generating Units” (EPA Information Collection Request (ICR) No. 2362.01; Office of Management and Budget (OMB) Control Number 2060-0631). The results of the Non-Hg case studies of 16 power plants suggest that nickel, hexavalent chromium, and arsenic are cancer-drivers, and “the non-cancer risks are not significant” (Strum et al., 2011). The “low” non-cancer risks were driven by nickel, arsenic, and hydrogen chloride.

The cancer risk estimates from this assessment indicate that the EGU source category would not be eligible for delisting under section 112(c)(9)(B)(i) of the CAA, which specifies that a category may be delisted only when the Administrator determines, “that no source in the category (or group of sources in the case of area sources) emits such HAPs in quantities which may cause a lifetime risk of cancer greater than one in one million to the individual in the population who is most exposed to emissions of such pollutants from the source” (Strum et al., 2011). The cancer risk greater than one in one million to the most exposed individual remains the threshold for continuing to regulate HAP emissions from EGUs under the CAA.

1.4.4 Regulatory Impact Analysis for MATS

In the Regulatory Impact Analysis for MATS, EPA assumed that all fine particles, regardless of their chemical composition, are equally potent in causing premature mortality. This is a significant assumption because, according to EPA, “PM_{2.5} produced via transported precursors emitted from EGUs may differ significantly from direct PM_{2.5} released from diesel engines and other industrial sources, but no clear scientific grounds exist for supporting differential effects estimates by particle type” (USEPA, 2011). The assumption that there is not robust scientific evidence of the differential effects of PM ignores an established body of literature in the environmental health sciences, which will be discussed below in further detail. In addition to the limitations and data gaps identified above, there are also significant uncertainties regarding geographic specificity inherent in EPA’s source emissions data which include uncertainties about facility location and emission point parameters, or release point locations due to averages from facility locations rather than the location of each specific unit within the facility (USEPA, 2018).

1.5 Conclusion

This section identifies key data gaps that need to be filled to advance understanding of the importance of considering metals other than mercury in the regulation of coal-fired power plant emissions. These non-mercury metals occur in various organic and mineralogic compounds with variation in the makeup of the metal mixtures and particulate sizes with which the mixtures are associated. Once emitted into the airstream, airborne metal-particulates can settle out into soils and surface waters, creating a potential for multiple complex exposure pathways beyond direct inhalation, and allowing for exposures to occur far away from their sites of emission. The following sections offer a review of metals associated with different types of coal and the recent toxicological literature on metals and metal mixtures associated with human toxicity and a proxy analysis of health impacts from metal exposure from a better studied system (mine wastes) that can be used to estimate metal toxicity. Such information can better inform regulation of EGUs

and, in particular, improve understanding of the heightened and concentrated impacts of EGUs on fenceline communities.

2. Metals in Coal and Coal Combustion Products

In this section, a summary of published literature and reports documenting particulate matter (PM) and metals in coal-fired power plant emissions, including EPA's NEI data, is provided. This section highlights the following points and data gaps on metal emissions from burning coal and coal combustion products:

- As a result of the original 1998 RTC which focused on a subset of metals, the EPA's NEI collects data on only 11 HAP metals in emissions from coal-fired power plants. However, a total of 30 metals are detected in coal and combustion productions as reported in the USGS report by Affolter et al. (2011).
- There are substantial differences in PM_{2.5} and metal emissions from coal-fired power plants depending on the coal type burned.
- The levels of PM_{2.5} and metal emissions vary considerably based on the geographic location of the power plant and most affected regions have high levels of metal emissions for multiple metals.
- Partitioning of the metals and design of the power plants are important metrics in estimating the concentrations of metals in atmospheric emissions from coal-fired power plants.

2.1 Types of Coals

Coal contains high amounts of carbon and hydrocarbons for power generation; it is classified as a non-renewable source of energy as it takes over a million years to form from fossilized plants subjected to pressure and heat. Based on the heat generation capacity, ash content, and moisture, coal is classified into four types: anthracite, bituminous, sub-bituminous, and lignite (EHI 2011). Two types of coal, bituminous and subbituminous, account for up to 93% of the coal generated electricity in the U.S. (NRC 2010).

2.2 Particulate Matter Emissions from Coal-Fired Power Plants

An important determinant of the health effects of particulate emissions is particulate size. The smaller the particle, the more deeply it travels into the airways. Of greatest concern are particulates PM_{2.5} and smaller that can pass the systemic circulation system to reach other organs and pose a greater threat to human health (Gomes and Florida-James, 2014; Figure 2). This section discusses current knowledge of PM_{2.5} emissions from coal-fired power plants.

Emission factors have been used for a long time as a tool to develop emissions inventories at the national, regional, state, and local level for better management of air quality and development of new control strategies. These are representative values that relate the quantity of a pollutant released to the atmosphere (e.g., grams of PM) with an activity associated with the release of that pollutant (e.g., per kWh electricity generated). Out of the six criteria air pollutants (CAPs; PM, ozone, CO, SO₂, NO₂, lead), we focus here on PM_{2.5} emissions.

In a recent report, Ou and Cai (2020) developed a new approach to improve and apply a consistent methodology to estimate the power efficiencies and CAP (including particulate matter) emission factors for power generation from combustion of different fuel types. As shown in Table 4, on average, coal-fired power plant electricity generation accounted for 30% of the electricity generated in the United States in 2017. But the contribution of coal to electricity production varied substantially across the eight North American Electric Reliability Corporation (NERC) regions, ranging from 1.1 to 51%. Further, the associated PM_{2.5} released during burning of the coal differs up to 9-fold depending on the coal type (0.014 to 0.128 g/kWh).

Closer examination of Table 4 reveals that bituminous and sub bituminous coal are the major contributors to energy generation in the U.S. (as described in Section 2.1). Further, each of these coal sub-types exhibits varying levels of PM_{2.5} emitted per kWh of electricity generated. Sub bituminous coal contributes to relatively high emissions in the Western region (0.118 g/kWh). Lignite coal only accounts for 4.7% of the electricity generated at the national level, but it has the highest emission factor for PM_{2.5} at the national level (0.128 g/kWh). Finally, refined and lignite coals contribute higher levels of PM_{2.5} (0.128 and 0.105 g/kg/kWh respectively) in the Midwest region relative to these coal types in other regions (Table 4). This comparison of PM_{2.5} emission factors from 2017 data clearly illustrates the fact that emissions from coal-fired power plants vary based on the coal type used and geographic location of the power plant.

Table 4. National & North American Electric Reliability Corporation (NERC) regional weighted-average electricity emission factors for PM_{2.5} released from different types of coal (Ou and Cai 2020) ‡.

National/NERC	Coal (%) ^a	Coal subtype (%) ^b	PM _{2.5} (g/kWh) ^c
National	30.00	Subbituminous (38.8)	0.043
		Bituminous (35.9)	0.068
		Refined coal (19.6)	0.065
		Lignite (4.7)	0.092
Florida Reliability Coordinating Council	15.10	Bituminous (97.2)	0.109
Midwest Reliability Organization	51	Subbituminous (55.0)	0.038
		Lignite (13.4)	0.128
		Refined coal (31.6)	0.105
Northeast Power Coordinating Council	1.10	Bituminous (90.9)	0.015
Reliability First Corporation	38.10	Subbituminous (12.0)	0.039
		Bituminous (57.8)	0.063
		Refined coal (28.3)	0.077
Southeastern Electric Reliability Corporation	31.20	Subbituminous (30.5)	0.032
		Bituminous (39.8)	0.047
		Refined coal (29.4)	0.052
Southwest Power Pool	41.10	Subbituminous (88.4)	0.062
		Lignite (7.0)	0.014
Texas Regional Entity	29.50	Subbituminous (68.2)	0.061
		Lignite (31.7)	0.098
Western Electricity Coordinating Council	22.80	Subbituminous (32.2)	0.118
		Bituminous (61.9)	0.041
		Refined coal (5.5)	0.075

‡Data on fuel type and net energy generation were obtained from U.S. Energy Information Administration's (EIA's) form EIA-923 at the power plant level and the corresponding PM_{2.5} emissions were derived from the National Emissions Inventory (NEI) dataset released in 2017.

^aPercentages indicate the contribution of coal in the total electricity generation in the United States (national) & NERC regions.

^bPercentages indicate the contribution of different coal subtypes in the electricity generation at the national and NERC regional levels.

^cPM_{2.5} indicates grams of PM emitted from burning different coal types per kWh electricity generated.

2.3 Metals Emissions from Coal-Fired Power Plants

The EPA NEI reports metal emissions values from coal-fired power plants for 11 HAP metals (arsenic, beryllium, cadmium, chromium, cobalt, lead, mercury, manganese, nickel, antimony, and selenium). These data are available for the years 2008, 2011, 2014, and 2017. However, these 11 metals are only a small fraction of the total metals emitted from coal-fired power plants (as described in Section 1.3) and the composition of metals in different coal types varies. In 2011, Spengler et al. compared emissions of 8 out of 11 metals reported by EPA, including arsenic, beryllium, cadmium, chromium, lead, mercury, manganese, and nickel, across the three types of coal: bituminous, subbituminous, and lignite (EPA 2010). They reported that bituminous coal contains 2-3 times higher concentrations of six (arsenic, beryllium, cadmium, chromium, lead, and nickel) out of the eight metals than subbituminous coal—providing evidence for varying levels of metal emissions from different coal types.

An in-depth examination of emissions of the 11 metals related to coal type (reported across the 10 NERC regions) is shown in Table 5. Bituminous and subbituminous coal types account for the greatest metal emissions (as noted in Section 2.1 & 2.2). In addition, the percent of coal type used to generate electricity corresponds to the metal emissions (tons per year (tpy)). For example, Florida Reliability Coordinating Council (FRCC), RFC (Reliability First Corporation), and Southeastern Electric Reliability Corporation (SERC) regions that generate much of their electricity using bituminous coal had the highest emissions of arsenic, cadmium, cobalt, chromium (III & VI), mercury, manganese, nickel, lead, antimony, and selenium. Similarly, Midwest Reliability Organization (MRO), Southwest Power Pool (SPP), Texas Regional Entity (TRE) regions have the highest metal emissions from subbituminous coal-fired power plants. An exception is the Western Electricity Coordinating Council (WECC) region, where bituminous coal produces 62% of the electricity but metal emissions are higher from subbituminous coal. It is noted that though lignite and refined coals contribute higher PM_{2.5} emission factors in the Midwest region (Table 4), these coal types have lower metal emissions than other coal types (Table 5)—indicating the differences between PM_{2.5} emissions and metal emissions within a coal type.

An analysis of NEI 2017 coal-fired power plant emission data for 11 metals is shown in Figure 2. The combined total emissions of these 11 metals is greater in coal-fired power plants located in the Eastern United States. However, there are more localized areas in the west that also have high total amounts of these metal emissions including: Arizona; Montana; North Dakota; Washington; and Wyoming.

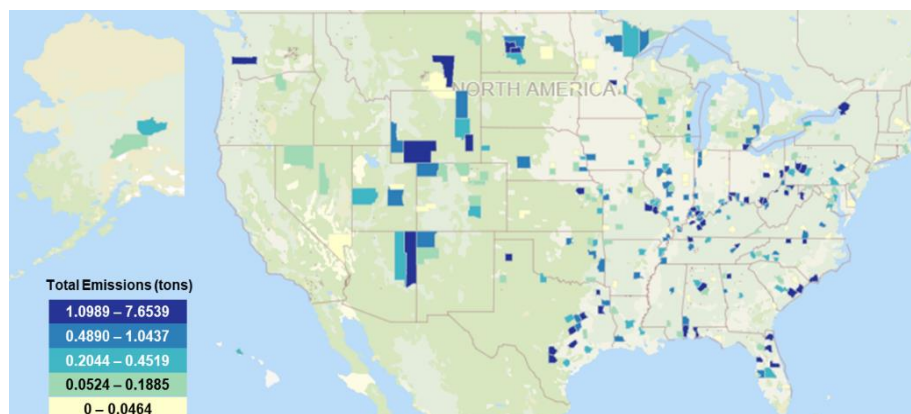


Figure 2: Total combined emissions (short tons/year) of 11 metals across the United States are represented. Metals include As, Be, Cd, Cr (III), Cr (VI), Co, Hg, Mn, Ni, Pb, Sb, and Se. Data were retrieved from EPA's NEI 2017 and the map was generated using NEI's interactive online tool.

Table 5. Metals emissions (tpy) released from different types of coal in the North American Electric Reliability Corporation (NERC) regions. Data on coal type was obtained from U.S. Energy Information Administration's (EIA's) form EIA-923 (2017) at the power plant level and the corresponding metal emissions were derived from the EPA National Emissions Inventory (NEI) 2017 dataset.

NERC ^a	State	Coal Type ^b	Metal Emissions (tons/year)											
			As	Be	Cd	Co	Cr (III)	Cr (VI)	Hg	Mn	Ni	Pb	Sb	Se
ASCC	AK	LIG	14	1	0.01	5	24	3	22	45	37	11	6	9
FRCC	FL	BIT	617	26	91	141	5340	728	170	7188	2165	429	220	14944
HICC	HI	BIT, SUB	61	4	0.1	23	104	14	9	198	164	47	27	121
MRO	IA, MN, MT, ND, NE, SD, WI	LIG	33	1	5	7	288	39	314	394	114	22	12	159
		SUB	363	33	63	196	2391	326	387	2519	1774	569	131	1974
		BIT, RC, SUB	76	5	14	27	647	88	55	914	321	70	29	918
		BIT, SUB	10	3	5	16	73	10	3	140	116	33	6	53
		LIG, SUB	20	1	1	4	116	16	16	64	54	24	6	232
		LIG, RC, SUB	99	11	20	52	597	81	347	1216	477	119	31	374
		LIG, RC	71	3	11	15	620	85	198	849	246	48	26	342
		RC, SUB	17	2	4	12	112	15	24	167	104	57	6	90
NPCC	CT, MA, NH	BIT	19	2	1	0.3	43	6	3	53	35	6	0.4	34
		SUB	5	0.3	0.3	1	40	6	3	8	16	5	1	8
RFC	DE, IL, IN, KY, MD, MI, MO, NJ, NY, OH, PA, VA, WV	BIT	964	48	137	259	7818	1066	717	9909	3815	1666	333	19536
		BIT, RC	682	30	101	158	5718	780	209	7904	2443	493	250	15973
		BIT, RC, SUB	455	18	74	84	2975	406	269	4612	1316	433	170	11546
		BIT, SUB	685	36	91	196	4938	673	467	5821	2359	594	211	10963
		RC, SUB	674	46	43	197	5844	797	325	1159	2322	669	143	1134
		SUB	179	23	20	78	1393	190	144	605	774	256	43	339
SERC	AL, AR, GA, KY, LA, MS, NC, SC, TN, VA	BIT	794	43	130	280	4907	700	503	17263	2618	933	272	12400
		BIT, RC	200	12	35	67	1701	232	72	2398	827	178	75	4508
		BIT, SUB	439	35	78	194	3360	458	283	3317	1949	530	151	5247
		RC	192	11	0.2	72	328	45	83	627	519	148	86	120
		SUB	271	22	23	101	2324	317	205	668	1067	307	64	496
SPP	IL, KS, LA, OK	BIT	33	1	5	7	283	39	73	388	113	22	12	808
		BIT, SUB	21	5	7	32	147	20	20	280	232	66	12	146
		LIG	6	2	3	10	45	6	37	86	71	20	4	65
		SUB	147	12	14	57	1260	172	162	388	596	171	35	270
TRE	TX	LIG	202	11	41	130	1503	205	361	2723	645	237	103	3940
		SUB	411	26	56	149	2344	320	392	2867	1460	366	159	2942
		LIG, RC	75	9	10	54	245	33	224	468	387	110	36	256
		LIG, SUB	471	37	72	308	3783	516	695	5513	3993	556	180	13539
WECC	AZ, CO, MT, NM, NV, OR, UT, WA, WY	BIT	90	22	40	136	622	85	48	1189	985	281	50	532
		BIT, SUB	43	9	15	55	250	34	67	478	396	120	23	285
		RC, SUB	102	4	15	22	888	121	59	1215	353	68	37	1281
		SUB	984	79	179	399	4951	675	785	6643	3777	1591	371	9102

^aASCC= Alaska Systems Coordinating Council, FRCC= Florida Reliability Coordinating Council, HICC= Hawaiian Islands Coordinating Council, MRO= Midwest Reliability Organization, NPCC= Northeast Power Coordinating Council, RFC= Reliability First Corporation, SERC= Southeastern Electric Reliability Corporation, SPP= Southwest Power Pool, TRE= Texas Regional Entity, WECC= Western Electricity Coordinating Council

^bBIT=Bituminous, LIG=Lignite, RC=Refined Coal, SUB= Sub-bituminous

Examination of individual metals shows that states with higher emissions for one metal generally also have higher emissions for the other metals (Table 6). For example, Kentucky, Texas, and West Virginia are the states with higher emissions for all 11 metals. Also, the states of Missouri, Florida, Ohio, Pennsylvania, and Indiana have higher emissions for 8-10 metals (Table 6).

Table 6: Metals emissions from coal-fired power plants across the top 10 states with the highest metal emissions. Emissions are listed in total emissions in the year 2017 (EPA NEI 2017). The 10 states are listed in increasing order of metal emissions.

Metals	Emissions from Top 10 States	Emissions Range across top 10 states (tons/year)	Emissions Range across all states (tons/year)
As	MT, IN, IL, KY, OH, FL, PA, WV, MO, TX	0.23-0.69	0.0002-0.69
Be	FL, IN, MN, WY, WV, IL, PA, KY, MO, TX	0.01-0.05	0.00003-0.05273
Cd	IN, AL, NC, PA, OH, MT, FL, KY, WV, TX	0.03-0.11	0.00001-0.11201
Cr (III)	TN, WY, PA, IN, KY, OH, FL, WV, MO, TX	1.5-4.4	0.0011-4.404
Cr (VI)	TN, WY, PA, IN, KY, OH, FL, WV, MO, TX	0.20-0.60	0.0002-0.6006
Co	FL, IA, MN, WY, PA, WV, IL, MO, KY, TX	0.07-0.39	0.0001-0.3932
Hg	MI, KY, WY, IN, WV, IL, OH, MO, ND, TX	0.14-0.92	0.00003-0.91583
Mn	NY, IN, PA, KY, OH, FL, AL, WV, NC, TX	2.5-6.6	0.0022-6.595
Ni	IL, WY, OH, PA, IN, FL, KY, WV, MO, TX	0.7-3.8	0.002-3.80
Pb	WV, IL, PA, KY, MT, OH, MO, IN, TX, MN	0.3-0.8	0.0005-0.81
Sb	IL, IN, MT, MO, OH, FL, KY, PA, WV, TX	0.1-0.3	0.0001-0.2900
Se	TN, TR ¹ , MT, IN, KY, PA, OH, FL, WV, TX	2.4-10.8	0.0004-10.79

¹TR= Tribal Region

At the county level (Table 7), it was noted that emission levels are often localized within larger regions. For example, while the state of Texas has the highest average levels of arsenic emissions (Table 6), Franklin County in Missouri contributes to the highest localized arsenic emissions (0.198 tons/year, Table 7). Examination of the county level data highlight that high localized metal emissions can occur in a state that is not in the top in terms of highest average metal emissions. Examples of these regions include AR-Jefferson (chromium (III & VI)), AK-Fairbanks North Star (arsenic, lead), CO-Moffat (beryllium), GA-Monroe (mercury), MS-Choctaw (beryllium, antimony), TX (mercury, nickel), UT-Emery (beryllium, cadmium, cobalt, nickel), VA-Hopewell City (mercury), and WA-Lewis (chromium (III & VI)). Another notable observation is that parts of the Navajo Nation in AZ, NM, and UT have reported high emissions for all 11 metals (Table 7). Taken together, these data illustrate that metal emissions vary substantially based on the localized geographic region which, in turn, can be expected to lead to localized health impacts. This in turn suggests the need to evaluate health impacts that may result from unequal distribution of metal emissions.

Table 7: Metals emissions from coal-fired power plants across the top 20 counties with the highest metal emissions. Emissions are listed in total emissions (short tons) in the year 2017 (EPA NEI 2017). The 20 counties are listed in increasing order of metal emissions.

Metals	Tons/year	Top 20 Counties with highest metal emissions from coal-fired power plants
As	0.066-0.198	AL-Mobile, WY-Sweetwater, MO-Jefferson, IN-Gibson, OH-Jefferson, TX-Potter, TX-Fayette, TX-Atascosa, TX-Rusk, PA-Armstrong, TX-Bexar, MS-Choctaw, PA-Beaver, AK-Fairbanks North Star, TX-Titus, FL-Putnam, TR-Navajo Nation, Arizona, New Mexico & Utah, OH-Gallia, WV-Harrison, MO-Franklin
Be	0.004-0.014	TX-Potter, WY-Sweetwater, OH-Gallia, MN-Itasca, WY-Campbell, MO-Jefferson, WV-Monongalia, TX-Freestone, CO-Moffat, KY-Carroll, IA-Pottawattamie, UT-Emery, ND-Mercer, MS-Choctaw, WV-Harrison, TR-Navajo Nation, Arizona, New Mexico & Utah, MT-Rosebud, TX-Titus, TX-Bexar, MO-Franklin
Cd	0.010-0.045	WV-Monongalia, UT-Emery, IN-Gibson, OH-Jefferson, NC-Person, TX-Fayette, AL-Mobile, TX-Atascosa, TX-Rusk, PA-Armstrong, MO-Franklin, TX-Titus, NC-Haywood, FL-Putnam, OH-Gallia, PA-Beaver, TX-Bexar, TR-Navajo Nation, Arizona, New Mexico & Utah, WV-Harrison, MT-Rosebud
Cr (III)	0.444-1.721	WA-Lewis, WV-Pleasants, AL-Jefferson, WV-Grant, AR-Jefferson, WY-Lincoln, WY-Sweetwater, MO-Jefferson, IN-Gibson, OH-Jefferson, TX-Fayette, TX-Atascosa, TX-Rusk, PA-Armstrong, FL-Putnam, TR-Navajo Nation, Arizona, New Mexico & Utah, TX-Titus, OH-Gallia, WV-Harrison, MO-Franklin
Cr (VI)	0.061-0.235	WA-Lewis, WV-Pleasants, AL-Jefferson, WV-Grant, AR-Jefferson, WY-Lincoln, WY-Sweetwater, MO-Jefferson, IN-Gibson, OH-Jefferson, TX-Fayette, TX-Atascosa, TX-Rusk, PA-Armstrong, FL-Putnam, TR-Navajo Nation, Arizona, New Mexico & Utah, TX-Titus, OH-Gallia, WV-Harrison, MO-Franklin
Co	0.010-0.045	WV-Monongalia, UT-Emery, IN-Gibson, OH-Jefferson, NC-Person, TX-Fayette, AL-Mobile, TX-Atascosa, TX-Rusk, PA-Armstrong, MO-Franklin, TX-Titus, NC-Haywood, FL-Putnam, OH-Gallia, PA-Beaver, TX-Bexar, TR-Navajo Nation, Arizona, New Mexico & Utah, WV-Harrison, MT-Rosebud
Hg	0.049-0.173	WY-Sweetwater, GA-Monroe, VA-Hopewell city, TX-Atascosa, TX-Harrison, TX-Limestone, MT-Rosebud, TX-Fayette, AL-Jefferson, TX-Titus, TR-Navajo Nation, Arizona, New Mexico & Utah, MO-Franklin, TX-Rusk, OH-Gallia, ND-Oliver, TX-Robertson, TX-Milam, TX-Freestone, ND-McLean, ND-Mercer
Mn	0.622-3.416	AL-Jefferson, WV-Grant, TX-Freestone, PA-Beaver, IN-Gibson, OH-Jefferson, TX-Fayette, TX-Atascosa, TX-Rusk, TX-Titus, PA-Armstrong, FL-Putnam, MT-Rosebud, TR-Navajo Nation, Arizona, New Mexico & Utah, OH-Gallia, WV-Harrison, NC-Person, NY-Jefferson, AL-Mobile, NC-Brunswick
Ni	0.239-1.059	ND-Mercer, OH-Jefferson, IA-Pottawattamie, TX-Fayette, UT-Emery, TX-Atascosa, WV-Monongalia, TX-Rusk, MS-Choctaw, PA-Armstrong, IN-Warrick, MT-Rosebud, FL-Putnam, OH-Gallia, TX-Bexar, TR-Navajo Nation, Arizona, New Mexico & Utah, WV-Harrison, TX-Freestone, MO-Franklin, TX-Titus
Pb	0.072-0.498	IL-Massac, FL-Putnam, MS-Choctaw, OH-Gallia, MN-Kandiyohi, AK-Fairbanks North Star, NC-Person, WV-Harrison, NC-Brunswick, TR-Navajo Nation, Arizona, New Mexico & Utah, NY-Jefferson, PA-Beaver, TX-Bexar, TX-Titus, MO-Franklin, OH-Tuscarawas, MN-St. Louis, MT-Rosebud, MN-Itasca, IN-Warrick
Sb	0.022-0.092	NC-Person, PA-Schuylkill, TX-Potter, IN-Gibson, OH-Jefferson, TX-Milam, TX-Fayette, TX-Atascosa, TX-Rusk, PA-Armstrong, PA-Beaver, FL-Putnam, TX-Titus, MO-Franklin, OH-Gallia, MS-Choctaw, TR-Navajo Nation, Arizona, New Mexico & Utah, TX-Bexar, WV-Harrison, MT-Rosebud
Se	1.015-3.653	MI-Monroe, FL-Citrus, FL-Orange, KY-Ohio, FL-Escambia, KY-Boone, WV-Pleasants, WV-Grant, TX-Milam, IN-Gibson, OH-Jefferson, PA-Armstrong, TR-Navajo Nation, Arizona, New Mexico & Utah, TX-Titus, PA-Beaver, FL-Putnam, OH-Gallia, TX-Freestone, MT-Rosebud, WV-Harrison

2.4 The Metal Content in PM_{2.5} Emissions

The previous two sections separately discuss PM_{2.5} emissions (Table 4) and the metal content of total emissions from coal-fired power plants (Table 5) in relation to overall coal usage and coal type in different regions of the United States. These data reflect that metal concentrations vary in different coal types as do PM_{2.5} emissions. However, it is important to point out that the relationship between particulate size and metal content is still not well understood. It is known that PM_{2.5} emissions from coal-fired power plants contain arsenic, cadmium, lead, and other HAP metals (Gonzalez-Maddux et al., 2014). Further, as already stated, it is known that the metal content in emissions varies substantially based on the geographic region, the type of coal burned, and the proportion of electricity generation contributed by coal. However, little information currently exists regarding the specific PM_{2.5}-associated metal content in these emissions.

Closely related to this is that the quantity of trace metals emitted from coal-fired power plants will depend on the power plant design and operation. For example, the design and choice of the air pollution capturing devices (e.g., electrostatic precipitators or fabric filter baghouses) will impact trapping of particles prior to emission (Affolter et al., 2011). Similarly, operating parameters such as flue gas temperatures will govern vapor formation in the furnace and partitioning of metals into particulates (Swanson et al., 2013; Senior et al, 2019).

2.5 Conclusion

In summary, this section highlights that the PM_{2.5} and, separately, metal emissions resulting from coal-fired power plants vary substantially based on the geographic region, the type of coal burned, and the design of the power plant. The unequal distribution of metal emissions in each state/county suggests that the associated health impacts will be similarly unequally distributed. Specifically, it is known that many metals concentrate in the PM_{2.5} and smaller fractions with coal combustion (Pacyna 1987; Yi et al. 2008). Further, these smallest fractions are those least likely to be removed by the power plant design and choice of the air pollution capturing devices (Affolter et al. 2011). Thus, the specific metal mixtures and concentrations can be expected to vary both with the type of coal burned and the geographic region as noted above in Tables 4-7. A key finding is that the unequal distribution of metal emissions suggests that there is a second data gap and signals the need to assess whether this unequal distribution results in localized health effects of concentrated emissions on fenceline communities.

3. Estimating Metal Toxicity in Coal-Fired Power Plant Emissions

This section discusses the complexities of understanding toxicity of metals in coal-fired power plant emissions and, within the constraints of these complexities, summarizes present knowledge and data gaps in this area including the following:

- One data gap is that little data are available that directly measure the toxicity of many of the metal HAPs in coal-fired power plant emissions. Here we propose that mine waste data is a surrogate that can be used to fill such data gaps. Further, we provide information on exposure routes and toxicity related to both cancer and noncancer risks from mine waste that can be used to estimate risk from metals HAPs emitted by coal-fired power plants.

- Metal mobility must be considered in evaluating toxicity. Metals from coal-fired power plant emissions occur in various organic and mineralogic compounds with varying particle sizes. Once entrained in the airstream, these airborne metals settle to impact soil and water sources, creating a potential for multiple complex exposure pathways beyond direct inhalation, and allowing for exposures to occur far distal to site of emission. There are data gaps related to both understanding the stability of metals within different media and in the ability to predict the likelihood of downstream exposures.
- One complexity in understanding metal toxicity is that the relationships between exposure and response for single toxicants varies among metals. Relationships can vary from linear responses where increasing exposure leads to increasing toxicity, to U-shaped dose response curves where the relationship has a ‘sweet-spot’ where no toxicity is observed even though toxicity was seen with both lower and higher doses.
- For metal mixtures, the toxicology is also complicated. Many metals act through similar mechanisms, or are cleared from the body in similar ways. In such cases, one may anticipate an additive response should multiple metal exposures occur. However, the potential for antagonisms between metals acting on similar target receptors or organs also exists, as does synergy in responses. As the number of metals in a mixture increases, the ability to discern these differences in response becomes more complex as the number of metals and possible interactions increases.

3.1 Relevance of Mine Waste Data as a Surrogate for EGUs’ Metal HAP toxicity

We propose that mine waste data can be used as a surrogate to understand metal toxicity in coal-fired power plant emissions. Table 8 identifies the metals that have been found in coal-fired power plant emissions (Affolter et al., 2011), but for which the previous MATS docket and the Spengler et al. (2011) white paper does not provide toxicity information. In the intervening 10 years, Superfund Research Centers at the University of Arizona and the University of New Mexico have conducted extensive work on community exposures to mine waste: datasets that can provide reasonable surrogates for evaluating the range of toxicity anticipated from community exposures to the non-Hg metals in coal-fired power plant emissions. Table 8 (Column 5) shows the overlap between the metals in mine waste mixtures that have been associated with significant toxicity and metals found in coal-fired power plant emissions. The metals studied in mine wastes represent a broader range of metals of concern than those studied in either the MATS or Spengler references. Several additional parallel characteristics between the two source types also support the appropriateness of this assumption of surrogacy:

1. Many of the metals of concern in coal-fired power plant emissions are co-located geologically. Because these metals are co-located geologically, similar combinations of metal mixtures can be expected to be found in both coal and mining waste sources.
2. Both sources (coal-fired power plants and mine tailings) have high potential for airborne emissions.
3. Both sources have been shown to concentrate toxic metals in the finer particle size fractions,
4. Clusters of nano-size particles have been observed in both source materials. Such particles have increased environmental mobility, especially in the air pathway and through soil deposition and plant uptake.

Table 8. Metals Emissions Identified in MATS Docket (USEPA, 2012); Affolter et al. (2011); and Spengler et al. (2011).

Documented Emissions (Affolter et al., 2011)	Metals Considered in the MATS Docket (2012)	Metals in Coal-Fired Power Plant Emissions by Spengler et al. (2011)	Metals for Which Significant New Data are Available*	Metals studied in Mine Waste Exposure Studies. (Figure 3)**
As	X	X		x
Ba			Ba	x
Be		X		x
Bi			Bi	
Cd	X	X		x
Cl			Cl	
Co	X			x
Cr	X	X		
Cs			Cs	x
Cu			Cu	x
Ga			Ga	
Ge			Ge	
Hg	X	X		x
Li			Li	
Mn	X	X		
Mo			Mo	x
Nb			Nb	
Ni	X	X		
Pb	X	X		x
Ra			Ra	
Rb			Rb	
Sb	X			x
Sc			Sc	
Se	X	X		
Sr			Sr	x
Th			Th	
Tl			Tl	x
U		X	U	x
V			V	x
Y			Y	
Zn			Zn	
*Mixtures not evaluated in any referenced sources				
** Not an inclusive list, but representative of mixtures discussed				

In addition to similarities in emission characteristics and mixtures, a final point supporting mine-waste exposures in consideration of EGU metals emissions is derived from the appropriateness of target populations. In the Western US both electrical generation plants such as those in the 4-Corners region, and the abandoned mine origins of metal mixtures, while not necessarily co-located, frequently exist in proximity to Indigenous communities. Many of the population study results reported in the metal mixture studies summarized here are derived from studies with

Indigenous populations, and therefore can be considered a representative example of populations exposed to coal-fired power-plant emissions.

3.2. Health Outcomes of Metal Exposures

The remaining discussion of toxicity focuses on two primary classes of emissions: those for which no data are currently incorporated in the previous docket (column 4 of Table 8), including individual metals and mixtures of metals; and those that are included in previous EPA assessments but for which more recent research demonstrates more sensitive toxicity endpoints (i.e. lower dose effects) than those previously considered in either population or lab-based studies. The latter category includes newer data on arsenic, uranium, and metal mixtures. While the mine waste studies are the primary focus, selected additional studies on health outcomes associated with exposures from other sources different from but comparable to coal combustion emissions are included with their relevance noted. While the majority of data referenced have been published in peer-reviewed publications, we also include some newer studies that are currently undergoing peer-review through incorporation of representative figures.

While cancer is frequently used as the benchmark in regulatory assessments, a significant number of other health outcomes, or systemic dysfunction that can increase likelihood of a variety of chronic diseases including cancer, are also debilitating to individuals' lives. In the synthesis of recent studies on individual metals (and mixtures) discussed in the remaining sections, both cancer, non-cancer, and systemic changes associated with exposures to metals will be discussed. Data from population studies will be incorporated, and, where possible, subsequent analyses validating observed relationships in animal models and model cell or cell-free systems will be added. A comparison of known toxicity from mine tailings metal exposure with metal levels that have been measured from coal-fired power plants is presented in Table 9. The comparable levels of metals (arsenic, lead, cadmium, and uranium) in the mine tailings studies with those generated from power plants suggests the possibility for both cancer and noncancer outcomes from power plant emissions, which the MATS rule has significantly reduced but that continue to pose a threat to human health at current levels.

Here we provide some general remarks on exposure and response for single toxicants and for toxicants in mixtures and in the following sections we provide some specific examples for each. For single toxicants, those relationships can vary from linear responses where increasing exposure leads to increasing toxicity, to U-shaped dose response curves where the relationship has a 'sweet-spot' where no toxicity is observed even though toxicity was seen with both lower and higher doses. A clear example of this would be seen for essential metals where a deficiency, or low-dose, would result in toxicity, the increased intake to achieve a nutrient sufficient dose result in a healthy organism, but higher- than- sufficient doses can result in toxicity again, generally of a different effect than observed with deficiency. When we examine metals, such U-shaped responses would be anticipated for metals such as selenium, copper, and zinc which are both toxic, and essential nutrients.

When we look at interactions in metal mixtures, the toxicology can again get complicated. Many metals act through similar mechanisms, or are cleared from the body in similar ways. In such cases, one may anticipate an additive response should multiple metal exposures occur. However, the

Table 9: Comparison of metal exposures from mine tailings with known toxicity and metal exposures that may occur from coal burning. Metals have been highlighted with different shades for easy comparisons.

	Health impacts associated with metal exposure from mining waste				Metals found in coal and coal fly ash		
Metals	Study	Ppm	Exposure Source	Health Impacts	ppm	Medium/ Source	Study
As	Wong et al. 2019	0.0075-0.75 (cells)	Sodium Arsenite Solution	Zn-deficient cells co-exposed with Arsenic showed an increase in oxidative stress and inflammatory markers. In mice, induced acute proinflammatory response was further enhanced by exposure to arsenic.	19-59	Fly ash	Affolter et al. 2011
		0.05-0.5 (mice)					
	Qin et al. 2012	0.15	Sodium Arsenite Solution	Arsenic promotes survival of cells with unrepaired DNA lesions, and this may be the mechanism underlying arsenic co-carcinogenic action.	0.86-6.65	Soil	Müller et al. 2020
	García-Rico et al. 2020	10.5 - 23.1	Playground Soil	Arsenic concentration in playground dust was positively associated with serum matrix metalloproteinase-9 (a marker for chronic respiratory disease, lung inflammation, cardiovascular disease and cancer) levels in children.	1.22-62.41	PM	
As	Fry et al. 2020	0-168	Soil	Modeling demonstrated carcinogenic risk from dust ingestion, followed by inhalation, in both adults and children. Dermal contact was determined to also pose carcinogenic risk for children.	5.8 - 34	Illinois Basin Coal	Zierold et al. 2020
Pb		0-855			28-50	Fly ash	Affolter et al. 2011
					7.7 - 24	Illinois Basin Coal	Zierold et al. 2020
Cd	Manjón et al. 2020	0.32-0.45	Garden Soil	Exposure of Cd via ingestion of carrots showed high carcinogenic risk for 2 to < 3-year-olds. Cumulatively, Cd exposure from soil, dust, water, and vegetables showed higher carcinogenic risk for 1 to < 2-year-olds and 2 to 3 year-olds.	1	Fly ash	Affolter et al. 2011
		0.15-0.35	Playground Soil		0.14 - 1.3	Illinois Basin Coal	Zierold et al. 2020
U	El Hayek et al. 2021	2.4-106	PM ₁₀ and PM ₁	Uranium in the solid particulate form is more toxic than soluble form and it causes cell cytotoxicity and DNA damage.	8-9	Fly ash	Affolter et al. 2011
					1.3 - 3.3	Illinois Basin Coal	Zierold et al. 2020

potential for antagonisms between metals acting on similar target receptors or organs also exists, as does synergy in responses. As the number of metals in a mixture increases, the ability to discern these differences in response becomes more complex as the number of metals and possible interactions increases. These difficulties for many decades led researchers to examine one metal at a time, although recognizing that the likelihood of exposures to single metals in environmental settings is extremely small. In spite of this recognition of complexity, researchers in the past 10 years have focused more effort on development of methods to address the complexity and produce more appropriate understanding of the toxicity anticipated in real-world environmental exposures (<https://www.niehs.nih.gov/news/events/pastmtg/2015/statistical/index.cfm>). Although many analytical methods to overcome the barriers to mixture analyses are now in use, generalization of results is complicated by the method used. Cluster methods that pull out patterns of mixtures within a population exposure that are associated with specific responses are dependent on those particular mixtures being replicated in other situations for prediction of specific responses. Approaches that look at a hypothesis driven-subset of a complex mixture may miss subtle differences in responses that occur when other metals, either antagonist or synergistic, are present in addition. Effects of single metals at relatively low concentrations may also influence the outcome of specific mixtures without being a major component of the environmentally relevant mixture being assessed. Likewise, a metal that is little studied, and often ignored as part of a mixture exposure as a result, may be a major determinant in the outcome. In spite of these limitations, mixture analyses present a much more realistic prediction of real-world exposures than single metal analyses can accommodate. Due to the complexity of metals in coal and in oil, and the geographical differences in anticipated emissions, determinations of real-world anticipated impacts will encompass mixture exposures that are complex and variable, requiring thoughtful consideration of the mixtures data, where available, to ensure these effects are captured.

3.2.1 Exposure to Single Metals - Arsenic

Arsenic and uranium are two single metals for which additional toxicity information has become available through Superfund Center and other work in recent years. These will each be discussed separately.

Arsenic is a complex toxicant. A number of studies have been published since 2011 that provide more specificity of the impact of arsenic exposure on human health. In summary, arsenic is a Class I carcinogen (IARC 2012) known to induce oxidative stress and increase DNA damage through inhibition of DNA repair (Ding et al. 2008; Ding et al. 2009; Qin et al. 2012). Arsenic also has a number of noncancer effects in humans including effects on respiratory system development and function, dermal effects (e.g., hyperkeratosis), gastrointestinal effects, anemia, peripheral neuropathy, and liver or kidney damage (Kravchenko and Lysterly 2018). Examples of recent studies are summarized below. Taken together, these suggest that health outcomes may occur from arsenic exposures that are below currently regulated levels and that these outcomes may disproportionately affect vulnerable populations, especially children, due to multiple exposure routes that result from living in proximity to aerosol sources (coal-fired power plants/mine tailings), arsenic exceeding maximum contaminant levels in drinking water, and outdoor activities that disproportionately bring children in contact with soils as discussed below.

- A recent community-based analysis used the EPA's health risk assessment model to assess the risk of exposure to arsenic from a coal-fired power plant via different routes of exposure (Müller et al., 2021). The carcinogenic risk of arsenic exposure was estimated at

approximately 10 km from the coal-fired power plant. Results show that when air and soil exposure pathways were considered together in areas with elevated concentrations of arsenic in the air, there was a carcinogenic risk in most of the evaluated areas. Inhalation was the major pathway that contributed to the average daily dose of arsenic however, this study emphasizes that the carcinogenic effect of arsenic is enhanced when multiple routes of exposure exist. In this case close proximity to the coal-fired power plant produces metal-containing air emissions that can either be inhaled or deposited onto soils. Metals deposited onto soils may result in subsequent exposures through re-aerosolization (inhalation) or ingestion. Thus, inhaled particulates came from both air and soil that was aerosolized following metal deposition. There was also exposure via oral (ingestion) and dermal exposure pathways, in this case associated primarily with soils (Müller et al., 2021). Children, who are known to have higher soil contact, would be particularly susceptible to additional exposures through ingestion.

- A study of children in five schools in Mexico showed a positive association between elevated arsenic (an average of 16.9 mg/kg among the schools) in playground dust and a biomarker for arsenic exposure in urine and blood samples that is associated with chronic respiratory disease, lung inflammation, cardiovascular disease and cancer (García-Rico et al., 2020).
- A study of children and adults living near a copper smelter in Namibia that processes ores with high arsenic content showed that arsenic, lead and copper contamination was spatially associated with proximity to smelter operations. Modeling of carcinogenic risk showed that dust ingestion was the most important pathway, followed by inhalation, for both adults and children. Dermal contact to trace elements in dust was also determined to pose a carcinogenic risk for children, but not adults. Source apportionment using Pb isotopes confirmed metal contributions were from industrial emissions. Although soil concentrations were below EPA soil guidelines, dust wipe values were elevated (1012 ug/m² for arsenic) and similar to those at other international smelter locations (Fry et al., 2020).
- Two recent studies utilized a mouse model to demonstrate that inhalation exposure to real-world mine tailings dust containing a mixture of arsenic and other metals affects lung development (Witten et al., 2019). This study is the first to systematically collect and characterize real-world mine tailings dust in a murine inhalation exposure model (Witten et al., 2019). The most significant changes in lung structure and function were observed in male mice when exposure was chronic; occurring continuously in utero, after birth and into adulthood. Changes included increased airway hyper-reactivity, increased expression of epithelial to mesenchymal (EMT) transition protein markers and increased expression of cytokines related to eosinophils (white blood cells), changes which could predispose for diminished lung function and/or lung diseases over time. This was followed by a second study by the same group that used simulated dust spiked with arsenic to isolate effects of arsenic. Results again showed significant increases in pulmonary baseline resistance, airway hyper-reactivity, and airway collagen and smooth muscle expression and, further, that the responses were dependent on the level of calcium arsenate in the simulated dust, providing evidence that arsenic-laden dust can induce precursors of chronic lung damage (Chau et al., 2021).
- A recent cell culture study showed that low-level arsenic exposures (50 to 500 ppb) cause changes in oxidative stress associated genes. Wong et al. (2019) conclude that arsenic

exposure to zinc-deficient cells caused a decrease in intracellular zinc levels (Zn is an essential micronutrient) and increased oxidative stress and inflammation response— these responses are similar to effects of zinc deficiency. These results suggest that the effects of nutritional status and arsenic exposures may be additive in susceptible populations and that both need to be considered in assessing exposure.

- A 2012 cell culture study showed that low concentrations of arsenic (175 ppb as arsenite) reduced cell programmed destruction of DNA damaged cells and that the survival of these cells may be an important mechanism that supports co-carcinogenic outcomes (Qin et al., 2012). Trivalent arsenite or As(III) as arsenous acid, H_3AsO_3 , predominates in releases to air from industrial processes (USEPA, 2019), making this observation relevant in consideration of risk.
- Monomethylarsonic acid (MMA) and dimethylarsonic acid (DMA) are metabolites of inorganic arsenic produced by animals, plants and bacteria. Dimethyl and trimethyl forms of arsenic can volatilize into air (HSDB 2009). A recent study compared the effect of arsenic (as arsenite) vs. MMA exposure through drinking water on disruption of the production of red blood cells (erythropoiesis) (Medina et al., 2021). Arsenite induced impairment at 37.5 ppb, but MMA induced impairment at 7.5 ppb (below the drinking water standard for arsenic). Metabolites in the ppb range would be relevant exposure assumptions based on the ppm concentrations of arsenic in fly ash reported in Table 9.

3.2.2 Exposure to Single Metals - Uranium and Vanadium

Burning of the three major fossil fuels—coal, oil and natural gas—releases varying quantities of the naturally occurring radionuclides of the uranium-238 and thorium-232 series which are entrained in combustion gases, emitted to the environment, and available for aeolian transport and inhalation exposures to humans or other receptors (EPA 1998 Utility HAPS RTC). As early as 1954, it has been recognized that coal-burning, through transfer into the air, could increase human inhalation exposure to uranium and associated radionuclides (Anderson et al., 1954). While much of the work on uranium toxicity has been done in the context of mixture exposures, fundamental studies indicating the toxicity of uranium alone in model systems have been done to understand its contribution to mixture effects.

- Immunotoxicity. Oral uranium is poorly absorbed from the gut, as evidenced in minimal deposition in lymphoid tissues following 60-day drinking water exposures of mice to 5 or 50 ppm uranium as uranyl acetate to mirror the range of concentrations found in unregulated water sources often used for drinking water on Navajo Nation (Bolt et al., 2018). However, the lack of absorption results in high concentrations of ingested uranium to the epithelial immune cells lining the small intestine (Medina et al., 2020). Uranium exposures produced widespread suppression of CD4- intraepithelial lymphocytes, providing a critical framework for understanding population immunotoxicity observed in populations with chronic ingestion of poorly absorbed uranium. The gastrointestinal tract immune cells are known to play a vital role in systemic immune health.
- DNA damage and retention. Uranium (as uranyl acetate) in cell cultures produces dose-dependent DNA strand breaks at 10 to 100 μM concentrations, even below concentrations that affect cell viability (100 μM). The most sensitive toxicity endpoint identified for uranium is the inhibition of the DNA repair protein poly (ADP-ribose) polymerase with as low as 10 μM concentrations of exposure to uranyl acetate for 24 hours (Cooper et al., 2016). That environmentally relevant exposures to uranium can induce this toxicity is

demonstrated by observation of increased DNA damage associated with elevated blood uranium in human populations (Popp et al., 2000; Lourenco et al., 2013). In contrast to arsenic which inhibits DNA repair through specific competition at zinc-finger sites that control the activation of repair proteins, uranium appears to be non-specific for zinc-finger binding, and can inhibit DNA repair enzymes through multiple mechanisms, making uranium a more significant toxicant in inhibition of DNA repair. In related studies, uranium was shown to impair the function of a range of DNA repair proteins linked to Xeroderma Pigmentosum - Complementation group A and aprataxin (Cooper et al., 2016). Retention of damaged DNA can lead to cell death, or underlie mechanisms leading to development of chronic disease associated with the physiologic systems in which the damaged cells function.

- Cardiovascular hypertensive response. Aspiration of either 0.22 μmol of uranium or vanadium in solution resulted in increased vascular contraction and decreased vascular relaxation in response to normal neurotransmitter stimuli for these effects, a response consistent with the hypertensive responses resulting from inhalation of mine dust in mice (Zychowski et al., 2018), and the observation of increased likelihood of hypertension in populations exposed to mine waste (Hund et al., 2015).

3.2.3 Metal Mixtures

Actual exposure to toxic metals usually occurs in mixtures rather than single metals. Research on single metal exposures offers a basis for understanding the basic toxic effects of metals, but there has been a general acknowledgement in the field of environmental epidemiology, toxicology, and environmental science in general that it is time to go beyond a single-pollutant model (Breton and Farzan 2021; Keil et al. 2021; Tanner et al. 2020). Whereas the previous section offered insight into the health implications of exposures to individual metals, this section considers an emerging area of environmental health research on the implications of mixed-metals toxicity, an area that remains an identified data gap in the regulatory docket.

Exposures to complex mixture sources as would be anticipated from coal-fired power plant emissions reflect simultaneous exposures to multiple metals, complicating the determination of toxicity. While many metals share mechanisms of action within the body, patterns of synergistic, antagonistic, and additive responses are all possible. The understanding of mixture toxicity contains significant uncertainty that is dependent on the questions asked and the methods used in analyses. For example, as noted previously, when toxicity is assessed relative to a specific population the results will be informative for populations with similar exposure sources, but possibly not generalizable to mixture exposures that differ both in specific composition of metals and relative balance of concentrations for each. Conversely, if methods that assess the effects of single metals within a mixture are used, the analysis will provide insight into single metal effects that may be altered in different mixtures.

3.2.4 A Metal Mixture Example – Uranium and Other Metals on the Navajo Nation

As an example, to illustrate both the complexity and importance of considering health impacts of exposures to metals mixtures, the case of uranium mines on Navajo Nation can be considered. Biomonitoring of urine confirmed exposures of this population to multiple metals in mixtures at concentrations that exceed those observed in the U.S. population as a whole through the National

Health and Nutrition Examination Survey (NHANES). Urine uranium concentration distribution medians in pregnant women on Navajo Nation were 2.5 to 2.8 times the comparable NHANES data for pregnant women nationally (Hoover et al., 2020). Pregnant women in the Navajo Birth Cohort also had urine concentrations for manganese, cadmium, and lead that exceeded the NHANES 50th and 95th percentiles during pregnancy, sometimes as much as 2.5~3 fold as is the case for uranium. More than 500 abandoned uranium mines are located on Navajo Nation, but the data reported in the Hoover et al. report are inclusive of participants across Navajo Nation, including mined and unmined regions with as many as 25% of the participants showing no evidence of exposure.

Biomonitoring of urine samples during pregnancy also revealed that, even though there are consistencies in the source of contamination, within populations individual activities can result in exposures to a wide range of metal mixtures, and distinctly different patterns of exposures. Figure 5 shows six different patterns that were observed in pregnant women on Navajo Nation when exposures were clustered against 24 different metals analyzed. The clusters were identified against an outcome of pre-term birth, which will be discussed below in Section 3.3.2 describing observed effects of exposures to metal mixtures.

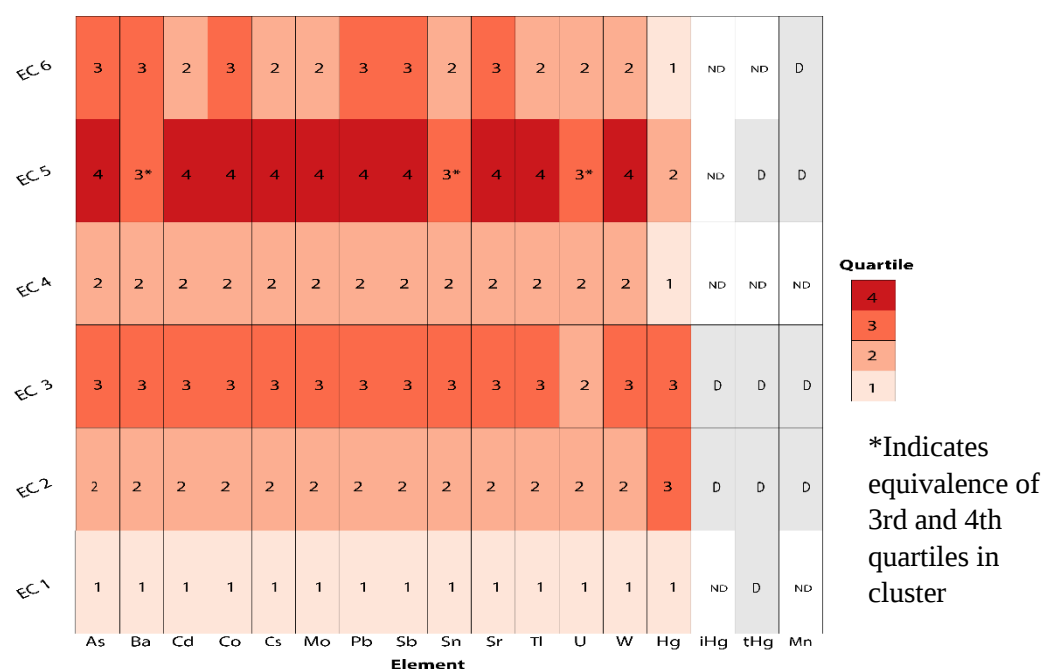


Figure 3. Clusters describing patterns of exposures observed within a population of pregnant women with respect to the 17 analytes on the x-axis. The numbers and shading correspond to the quartile of the range of exposures for each metal/ metalloid. In this population, >20% of the population (Cluster 1) had no evidence of exposure to any of the measured metals. While more than 40% (Clusters 5 and 6) showed the highest quartiles of exposure to nearly all metals. These results confirm that metals often move together in the environment setting the stage for complex interactions in responses.

Finally, exposures were found in some children at birth to exceed the 95th percentile of NHANES adults for uranium (infant and early childhood comparisons are not available in NHANES), with

concentrations increasing through the first 5 years of life for both uranium and arsenic (Figure 6). Remaining metals are currently being analyzed.

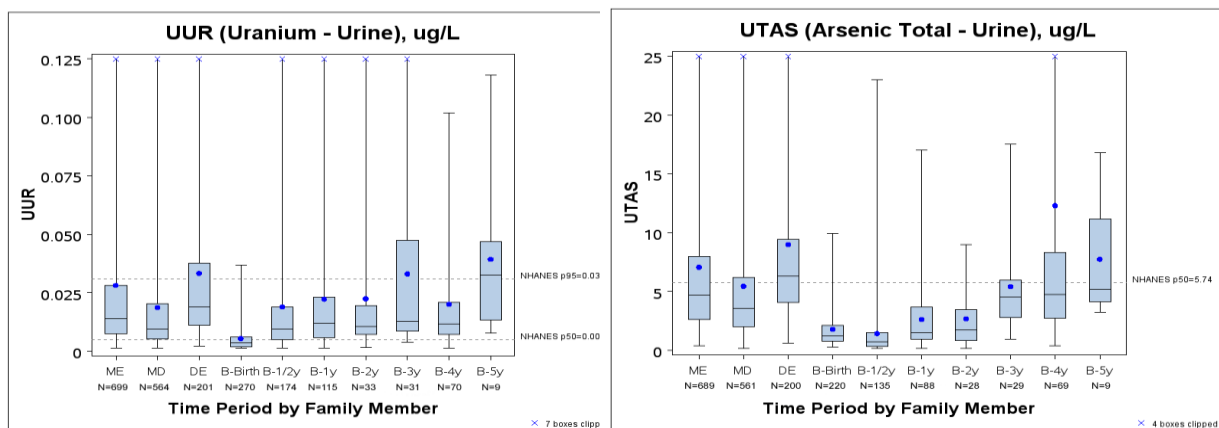


Figure 4. Biomonitoring results for Mothers at Enrollment (ME, Mothers at Delivery (MD), Dad's at enrollment (DE), and children annually through age 5 confirming increasing exposures to uranium and arsenic as children age. Note comparison reference lines to NHANES 50th and 95th %iles for U.S. adults shown in dotted lines.

3.2.5 Cancer and Noncancer Effects of Metals Mixtures

The studies reported in the following sections have examined mixtures toxicity at multiple levels to try to ascertain consistencies in effects, as well as to drill down into a mechanistic level that can inform potential interventions by breaking the cycle of toxicity. By definition, the effects reported from population studies reflect environmentally relevant exposures, and no effort to extrapolate the specific exposure doses has been made, although the biomonitoring concentrations resulting are available for mixture components. The studies were developed and conducted in partnership with exposed communities and include health care providers, community members, and researchers in efforts to understand the contributions of exposure to existing health profiles within communities on Navajo Nation exposed to metal mixtures associated with abandoned uranium mine waste.

3.2.5.1 Cardiovascular Toxicity

Population studies informed by survey responses and medical record confirmation of health profiles indicated that the community exposures to mine waste as defined by the proximity of the home to multiple mine sites and behaviors that increased contact with waste increased the likelihood of cardiovascular disease (hypertension), and the likelihood of multiple chronic diseases when considering cardiovascular disease, kidney disease, and diabetes as a group. While recognized risk factors for cardiovascular disease in the population were also significant predictors, including BMI and the family history of disease, an additional 25% increase in disease likelihood resulted from exposures (Hund et al., 2015).

Serum from those in the study, when incubated with human vascular tissue, showed increased response of mRNA for cytokines that are associated with atherosclerosis in other work (Harmon et al., 2017). Model inhalation exposures of mice to solubilized dust from mine waste also resulted

in increased contraction of vessels in response to a contractile stimulus (serotonin), and decreased relaxation in response to a relaxation stimulus (acetylcholine) relative to both background particulate matter and vehicle controls. This response was again consistent with hypertension in response to exposure to the waste mixtures, and could be simulated with exposure to purified vanadium or uranium alone (Zychowski et al., 2018).

These data support that multiple components of the metal mixtures in mine waste, as well as the mixtures themselves, are consistent in increasing the likelihood of hypertension at environmentally relevant concentrations.

3.2.5.2 Immune Dysfunction and Autoimmunity

Elevated autoimmunity has been associated with living in proximity to multiple abandoned mine sites on Navajo Nation and behaviors increasing contact with waste. The pattern of autoantibodies is consistent with those seen in environmentally induced autoimmunity. Drinking water content of uranium, even at low concentrations, was significantly associated with autoantibody production in both men and women, although arsenic was negatively associated with autoantibody production in women (Erdei et al., 2019). These findings are striking in that men and women showed equivalent levels of autoantibody increases, even though antibody prevalence is generally higher in women. The observation of perturbation of intestinal immunoregulatory cells provides a potential mechanism for uranium-induced immunotoxicity within these mixtures (Medina et al., 2020).

3.2.5.3 Preterm Birth

Exposures to the two highest of the metal mixture clusters (Clusters 5 and 6 in Figure 3) are associated with a 2.7-3.2 fold respective increase in the relative risk of preterm birth (manuscript submitted and in review).

3.2.5.4 Oxidative Stress

Results examining oxidative stress associated with mixture-exposures in pregnant women underscore the complexity of interpretation of mixtures data. Oxidative stress is a systemic change that has been associated mechanistically with the occurrence of multiple chronic diseases including diabetes, hypertension, and kidney disease – all at high prevalence in the Navajo population represented in these data. When looking at the contributions of three specific metal components in the mixture, uranium, arsenic, and zinc, in a hypothesis-driven study, only arsenic was shown to increase oxidative stress markers (Dashner-Titus et al., 2018). However, a subsequent analysis utilizing a two-step approach to identify significant contributors to oxidative stress from the broader set of metals in the exposure mixture found cesium and the arsenic metabolite dimethylarsinic acid to positively contribute to oxidative stress, while barium and thallium had a negative effect. Significant interactions of zinc with cesium and cobalt, and arsenic (III) and thallium were also found (Luo et al., 2019). Existing literature has far less often focused on mixture components cesium, barium, and thallium, underscoring the need for consideration of mixtures due to the lack of existing data on many of the components.

The analysis of the complete metal set in relation to oxidative stress underscores both the complexity of the interactions of metals in mixtures, and the fallacy of assuming that a lack of data indicates a lack of effect. Approaches that analyze mixtures by targeting specific components of

mixtures based on focused hypotheses can miss toxicity of less studied components, and perpetuate our lack of understanding of the contribution of these metals in environmental mixtures to toxicity in exposed populations.

3.2.5.5 Retention of DNA Damage

As documented in the individual metal toxicity Sections 3.3.1 and 3.3.2, inhibition of DNA repair enzymes by both arsenic and uranium is the most sensitive toxicity endpoint observed for these metals in laboratory as well as population-based studies. Subsequent retention of damaged DNA can be an underlying mechanism contributing to development of chronic diseases including cancer and immune dysfunction at these environmentally relevant and observed exposure levels. This toxicity occurs through competitive binding at a binding site for zinc that controls the function of repair enzymes. Zinc deficiency can exacerbate this effect, while increased systemic zinc, an essential nutrient, can outcompete the metals including uranium and arsenic and restore function of the repair proteins (Cooper et al., 2016; Ding et al., 2017). Zinc is an essential nutrient, but also a metal found in emissions of coal-fired power plants for which the MATS docket lacks toxicity data. While high-dose exposures to zinc can be toxic, restoration of sufficient systemic zinc in those with deficiency could serve to counteract toxicity of other metals as well. Zinc is one of the metals for which a U-shaped dose-response curve such as discussed in the introduction to this section would be anticipated.

3.3 Environmental Mobility of Non-Hg Metals and Resulting Exposure Pathways

Metals from coal-fired power plant emissions occur in various organic and mineralogic compounds with varying particle sizes. Once entrained in the airstream, these airborne metals eventually settle and deposit to impact soil and water sources. This creates the potential for multiple complex exposure pathways beyond direct inhalation, and allows for exposures to occur far distal to the site of emission. Literature on metal mobility varies as to the stability of metal chemical and mineralogic forms within different media, and the likelihood of downstream exposure pathways occurring as a result of initial releases to the environment.

Here we use research on abandoned mine wastes as a surrogate. Modeling studies, validated by existing national environmental data sets, have shown that wind-blown mobility and redistribution of abandoned mine waste results in non-uniform deposition of unstabilized and uncontained mine wastes over large regional areas (Stovern et al., 2016; Lin et al., 2020). The concentration of toxic metals in the finer size particle fractions and identification of crystalline nano-size structures reinforce the potential for toxic metals in wastes to be readily lofted in air currents preferentially, compared to crustal aluminum and iron which are more dominant in the larger size fraction (Arvasarala et al., 2017). Studies working with Indigenous communities within air and water sheds of uranium and gold mining operations have identified the occurrence of uranium, arsenic, vanadium, and other metals in soils, sediments, and waters affected by the legacy of abandoned mines. Mobility from soil to water is impacted by pH within environmentally relevant ranges observed in field sites. Specifically, mobility is enhanced for arsenic and uranium in acidic conditions. On the other hand, increasing calcium and carbonate (bicarbonate) at circumneutral pH has been shown to limit the mobility of uranium in water (Blake et al., 2015; Arvasarala et al., 2017; DeVore et al., 2019; Gonzalez-Estrella et al., 2020). A recent study on uranium toxicity demonstrated that uranium binding to organic carbon, a form consistent with metals in combustion

emissions, increases both the bioavailability and cellular toxicity of fine airborne particles of uranium (El Hayek et al., 2021). Finally, recent research shows that bioavailability for plant uptake is enhanced in soils and waters with increasing calcium concentrations, as is the transport within the plants from the roots up into the shoots, increasing the likelihood of occurrence in the food chain (Rodriguez-Freire et al., 2021; El Hayek et al., 2018; Hayek et al., 2019; DeVore et al., 2021).

The above findings, related to mine wastes, have significant implications for understanding the mobility of coal-fired power plant emissions considering similarities in composition, particle size fractions and potential for complex environmental conditions and media. It is expected that environmental mobility (and toxicity) will be modulated by these characteristics with important ones including particle size, aqueous and geochemical environment, pH, and carbon content. As just one example related specifically to coal, one would expect alterations in pH to result from release of coal-associated sulfur dioxide (SO₂) during coal burning. As such, variability in composition of the source coal, e.g. with respect to the sulfur content, would predict associated variability in the chemical form of particles emitted during combustion. In this case, release of SO₂ may be accompanied by a decrease in pH that could enhance mobility of metals from soil to water as noted above, while binding to carbon could increase bioavailability as well, emphasizing the importance of capturing worst case scenarios in the regulatory framework.

In summary, the complex processes affecting the mobility and resulting toxicity of metal mixtures within the range of environmentally relevant parameters support the importance of considering multiple exposure pathways, including inhalation and ingestion in determining potential risks. Ingestion pathways themselves are complex including through water, direct ingestion of soil (important for children), and ingestion of foods that have taken up deposited metals. Such foods may be plant-based resulting from direct uptake of metals into the plant or may be animal-based resulting from foraging of plants that have taken up metals.

4.0 Summary and Conclusions

This report provides context for understanding the potential for metals other than mercury that are present in coal-fired power plant emissions to have adverse human health outcomes. This report is not meant to be comprehensive but rather builds on research that was generated prior to the 2011 MATS rule to address key points of interest. It is anticipated this information will be used in ongoing and future rulemakings, to support EPA regulation of coal-fired power plants' emissions of hazardous air pollutants and to help justify strengthened, protective limits on coal-fired power plants' emissions of non-mercury metals.

Briefly, this report provides evidence that more consideration should be given to metals other than mercury in the regulation of coal-fired power plant emissions. There are up to 30 different metals in such emissions that can contribute to both cancer and noncancer health risks. There is evidence that it is not sufficient to consider the toxicity of these constituent metals individually. Research suggests that when present as mixtures, these metals may have cumulative or even synergistic contributions to toxicity. Research also suggests that it is not enough to solely consider inhalation as an exposure route but that cumulative exposure occurs as a result of inhalation, ingestion of dusts and soil (especially by young children), and ingestion of water and homegrown produce that

may have been impacted by emissions. Finally, current regulation does not take into full consideration vulnerable or disadvantaged populations who are more likely to live in proximity to coal-fired power plants and other sources of toxic metal emissions such as mine wastes.

To reiterate, there are significant risks from toxic metal emissions from coal-fired power plants (together with emissions from other sources) that EPA has not adequately considered in its rulemakings on this subject. Correspondingly, there are substantial benefits from reducing emissions of these metals that the MATS rule is currently producing, and that a strengthened rule would amplify, but that go unacknowledged in the record. Our understanding of the severity and range of human health impacts that these metals inflict is evolving and the recent work on exposures to metal mixtures in mine waste reviewed here has substantially increased our understanding of these exposure-toxicity relationships. It is clear that such wastes harm human health through similar exposure pathways that result from coal combustion in power plants. Moreover, Congress included these metals on the list of hazardous air pollutants that EPA is required to reduce, through regulation, to the maximum degree achievable. The current approach—to assume that these metals do not have significant health effects until those effects are established in the scientific literature—contravenes the Clean Air Act’s precautionary directives and disregards findings that these metals cause various health harms in a range of settings.

Although the extent of exposures to these metals from the combustion of coal in power plants remains uncertain, and while their health effects in humans are still being investigated, there is sufficient evidence from studies on exposures to emissions from mine wastes and other similar sources to conclude that these metals have serious and wide-ranging human health impacts, individually and in mixtures. Accordingly, reductions in emissions of these metals under the current MATS rule have produced substantial unquantified health benefits and greatly curtailed harmful metals that Congress specifically targeted in Clean Air Act section 112, supporting EPA’s decision to regulate these emissions under section 112. Furthermore, additional reductions of coal-fired power plants’ emissions of these toxic metals would yield still more unquantified benefits that would support strengthening the rule.

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