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PROJECTIONS OF AIR TOXIC EMISSIONS FROM COAL-FIRED UTILITY COMBUSTION:
Input for Hazardous Air Pollutant Regulators[a]

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RATIONALE

Significant changes are expected in the regulation of hazardous air pollutants (HAPs), changes caused primarily by Title III of the 1990 Clean Air Act Amendments (CAAA) and secondarily by Section 112(b) of the Clean Air Act (CAA), as amended in 1990.[1] The CAA requires the regulation of nearly 200 HAPs, although not all are considered significant in the generation of electricity.

The U.S. Environmental Protection Agency (EPA) is required by the 1990 CAAA to promulgate rules for all "major" sources of any of these HAPs. According to the HAPs section of the new Title III, any stationary source emitting 10 tons per year (TPY) of one HAP or 25 TPY of a combination of HAPs will be considered and designated a major source. In contrast to the original National Emission Standards for Hazardous Air Pollutants (NESHAP), which were designed to protect public health to "an ample margin of safety," the

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new Title III, in its first phase, will regulate by industrial category those sources emitting HAPs in excess of the 10/25-TPY threshold levels, regardless of health risks.

The trace elements normally associated with coal mineral matter and the various compounds formed during coal combustion have the potential to produce hazardous air toxic emissions from coal-fired electric utilities. Under Title III, the EPA is required to perform certain studies, prior to any regulation of electric utilities; these studies are currently underway. Also, the U.S. Department of Energy (DOE) maintains a vested interest in addressing those energy policy questions affecting electric utility generation, coal mining, and steel producing critical to this country's economic well-being, where balancing the costs to the producers and users of energy with the benefits of environmental protection to the workers and the general populace remains of significant concern.

SCOPE

Only a limited database exists for toxic emissions from coal-fired utility boilers, and there is a high degree of uncertainty concerning much of the data on some of the more volatile components contained in the generated flue gas. Hence, this paper addresses the key air toxic emissions likely to emanate from continued and expanded use of U.S. domestic coal. Specifically, as input for U.S. regulators, the author provides calculated values of the expected volumes of trace element emissions from a new, modern 500-MW, coal-fired plant,[b] relative to the CAAA 10/25-TPY stipulation. These values are calculated on the

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[b] Hypothetical, new, modern, stringently regulated, 500-MW, coal-fired plant, assuming particulate capture efficiency of 99.8%; compliance flue gas desulfurization (FGD), although unspecified as to type; and operation at 70% capacity factor.

basis of the coal quality characteristics, as sampled and analyzed carefully by geologists,[c] and projected emissivity factors described by the author in a technical report.[2] They are provided by region on the basis of the coal's typical elemental concentration of each HAP.

This paper does not project or calculate the expected volumes of real-world trace element emissions for existing coal-fired plants on the basis of actual coal burned and real-time emissivity factors, although such work merits and requires much careful study. Moreover, this report does not compile studies in which trace element measurements have actually been assembled under controlled experimental conditions or mass-balance studies.

Finally, significant organic compounds identified as HAPs, actually measured by various researchers, are identified herein for completeness.

ELEMENTS IDENTIFIED AS HAZARDOUS AIR POLLUTANTS

Most trace elements are associated primarily with the mineral portion of the coal -- aluminosilicates and silicates, carbonates, and sulfides are the major phases. And certain elements of environmental concern, such as arsenic, cadmium, mercury, lead, and selenium, are typically associated with pyrite and other sulfide minerals. A higher proportion of organically bound trace elements appears in low-rank coals (i.e., lignites and subbituminous coals) rather than bituminous coals. Ultimately, although both natural and anthropogenic sources emit trace elements (and their compounds) into the atmosphere, many result as airborne particulate matter and some as vapors.

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[c] Sampled coal is not beneficiated subsequent to mining or prior to analysis.

In the aforementioned work[2], the elements identified in the 1990 CAAA as HAPs are characterized and quantified by U.S. region (and state) by coal rank, including:

- Antimony (Sb),
- Arsenic (As),
- Beryllium (Be),
- Cadmium (Cd),
- Chlorine (Cl),
- Chromium (Cr),
- Cobalt (Co),
- Fluorine (F),
- Lead (Pb),
- Manganese (Mn),
- Mercury (Hg),
- Nickel (Ni),
- Phosphorus (P),
- Selenium (Se), and
- Radionuclides -- thorium (Th) and uranium (U).

A number of primary literature sources have been examined[3-11] regarding the presence and magnitude in the source coal of these precursor trace elements of potential air toxic emissions from the combustion of coals of different types. And although difficult to characterize and quantify, the HAPs derived from trace element emissions are somewhat straightforward because they vary primarily by fuel source.

PROJECTED COAL ELEMENT HAZARDOUS AIR POLLUTANT EMISSIONS

Rigorous derivation of standard combustion emission factors is difficult because of the inherent nature of partitioning behavior,[d] despite a variety of values having been reported by

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[d] Partitioning behavior indicates three basic types: Group 1 includes those trace elements of low volatility, even at high combustion temperatures, that have no tendency to concentrate in or on the fly ash and that tend to concentrate in the coarse residues or distribute themselves equally between the coarse residues (bottom ash) and the finer particulates (fly ash) -- Mn, Th, and, probably, P; Group 2 includes those trace elements that are vaporized but then recondense, preferentially concentrating in the fly ash fraction and exhibiting pronounced concentration in the smaller fly ash particles -- Sb, As, Cd, and Pb; Group 3 includes those trace elements that are also vaporized but remain mostly in the gas phase and may be emitted from the stack in a gaseous form -- Cl, F, and Hg; and several trace elements are considered intermediate -- Be, Co, Cr, Ni, and U (between Groups 1 and 2) and Se between Groups 2 and 3.

many researchers. Also, emission factors other than those actually measured may be only conjectural because partitioning behavior seems to depend on combustion system design, operating conditions, control devices, combustion temperature, amount of excess air, residence time, chemical form in the feed coal, and other variables.

Nevertheless, to guide the regulator and the policymaker in judiciously regulating and monitoring HAPs, a single emission factor is assumed for each HAP trace element in this report (Table 1). Although somewhat simplistic, this set of emissivity factors can be considered to represent the best conservative estimates available for emissions from a new, modern, environmentally regulated, pulverized-coal-fired (PCF) power plant. Therefore, these assumed, simplified emission factors permit the volume of trace element emissions from U.S. coals by region and rank to be calculated.

Appalachian Bituminous Coal

The U.S. Eastern Province includes large deposits of bituminous coal and some anthracite. It stretches from the northeastern portion of Pennsylvania southwestward to northern Alabama. The Eastern Province is about 900 miles long in a northeast to southwest direction, and almost 200 miles wide at its broadest point. The coal deposits of particular interest include the anthracite field in northeastern Pennsylvania and the bituminous coal of the Appalachian Region, which begins in Pennsylvania, runs through West Virginia into Kentucky, and includes parts of southeastern Ohio, central Tennessee, and western Virginia.

Data on U.S. Appalachian bituminous coals have been tabulated recently[2] by state for Pennsylvania, Ohio, West Virginia, Virginia, eastern Kentucky, and Tennessee, on the basis of source data by Swanson et al.,[3] Gluskoter et al.,[4] Zubovic et al.,[5] and Henderson et al.[6] and corroborated by White et al.[7]

Based on the typical concentrations of the HAP trace elements in bituminous coal of the Appalachian Region, total annual emissions may reach nearly 65 TPY, with probable Cl emissions of more than 22 TPY, F emissions of nearly 26 TPY, and Mn emissions of more than 13 TPY (Table 2). The other trace element HAPs do not appear to contribute significantly to emissions at their typical concentrations; each accounts for less than 0.82 TPY. However, As, Co, and Se may also need to be monitored because each of their emissions levels could exceed 10 TPY if higher, although atypical, concentrations of these elements were found in the actual feed coal burned.

Interior Bituminous Coal

Three regions make up the Interior Province -- the Northern Region in Michigan; the Eastern Region in Illinois, southwestern Indiana, and western Kentucky; and the Western Region, comprising parts of Iowa, Missouri, Nebraska, Kansas, Arkansas, and Oklahoma. The Eastern Region is the most important commercially. It occupies an area of roughly 50,000 square miles. Although Illinois has more coal resources than any other state, Kentucky leads the nation in coal production.

Data on U.S. Interior bituminous coals have been tabulated recently[2] by state for Indiana, Iowa, Missouri, Oklahoma, Arkansas, Illinois, and western Kentucky on the basis of source data by Swanson et al.,[3] Gluskoter et al.,[4] Zubovic et al.,[5,8] and Abernethy et al.[9] and corroborated by White et al.[7]

Based on the typical concentrations of the HAP trace elements in the Interior bituminous coal region, total annual emissions may exceed 91 TPY, with probable Cl emissions of more than 58 TPY and F emissions of nearly 26 TPY (Table 2). The other trace element HAPs do not appear to contribute significantly to emissions at their typical concentrations; each accounts for less than 0.80 TPY, except for Pb (1.19 TPY) and Mn (3.31 TPY). However, Pb, Mn, and Se may also need to be monitored because each of their emissions levels could exceed 10 TPY if higher, although atypical, concentrations of these elements were found in the actual feed coal burned.

Rocky Mountain Bituminous Coal

The Rocky Mountain Province contains the coal fields in the mountains running roughly north and south from New Mexico through Colorado, Utah, and Wyoming to Montana. Virtually every rank of coal can be found somewhere in the province. Rocky Mountain Province coals are higher in rank than Northern Plains coals of the same geological age because of increased thermal and tectonic stresses.[10]

Data on U.S. Rocky Mountain bituminous coals have been tabulated recently[2] by coal field and state for Hanna, Wyoming; North Park, Colorado; Uinta, Utah; and Black Mesa, Arizona on the basis of source data by Swanson et al.,[3] Gluskoter et al.,[4] and Zubovic et al.[5] and corroborated by White et al.[7]

Based on the typical concentrations of the HAP trace elements in the Rocky Mountain bituminous coal region, total annual emissions may reach 46 TPY, with probable Cl emissions of nearly 16 TPY and F emissions of nearly 28 TPY (Table 2). The other trace element HAPs do not appear to contribute significantly to emissions at their typical concentrations; each accounts for less than 0.31 TPY, except for Mn (0.95 TPY). However, Mn may also need to be monitored because its emission level could exceed 10 TPY if a higher, although atypical, concentration of this element were found in the actual coal burned.

Northern Plains Lignite and Subbituminous Coal

Just east of the Rocky Mountains lies the Northern Great Plains (Northern Plains) Province. It contains one of two major lignite deposits in the United States. The Northern Plains Province lies mostly in western North Dakota and eastern Montana, as well as in parts of northeastern Wyoming and northwestern South Dakota. The deposits of the province continue across the border into Canada. The Fort Union Region is the largest single deposit of lignite in the world. The province also contains the subbituminous fields of northern Wyoming and northern and eastern Montana.

In contrast to the Northern Plains subbituminous coals, the most important western coals are the low-sulfur Powder River Basin subbituminous coals. Because subbituminous coals have higher heating values and lower moisture contents than lignites, they are considered more suitable for long-distance transport. These Powder River Basin subbituminous coals are now consumed in 20 states, with shipments predominantly to the Midwest and Texas.[10] (No calculated values are presented here for the Powder River Basin subbituminous coals because of inconsistencies in the data reported by rank and state.)

Data on U.S. Northern Plains North Dakota lignites and Montana and Wyoming subbituminous coals have been tabulated recently[2] on the basis of source data by Swanson et al.[3] and Zubovic et al.[5] and corroborated by White et al.[7]

Based on the typical concentrations of the HAP trace elements in the Northern Plains lignite and subbituminous coal region, total annual emissions may exceed 46 TPY, with probable Cl emissions of nearly 22 TPY and F emissions of more than 22 TPY (Table 3). The other trace element HAPs do not appear to contribute significantly to emissions at their typical concentrations; each accounts for less than 0.35 TPY, except for Mn (1.67 TPY). However, As, Mn, and Hg may also need to be monitored because each of their emissions levels could exceed 10 TPY if higher, although atypical, concentrations of these elements were found in the actual feed coal burned.

Gulf Lignite

The Gulf Province is the other major lignite deposit in the United States. It stretches from Alabama and Louisiana into Texas; it runs in a wide belt southwestward through Texas to the Rio Grande.

Data on U.S. Alabama[e] Gulf lignites have been tabulated recently[2] on the basis of source data by Swanson et al.[3] and White et al.[11] and corroborated by White et al.[7]

Based on the typical concentrations of the HAP trace elements in the Gulf lignite region, total annual emissions may reach 367 TPY, with probable Cl emissions of 215 TPY, F emissions of 124 TPY, and Mn emissions of 16 TPY (Table 3).[f] The other trace element

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[e] No Texas data were available in the format required for the calculation.

[f] One must remember that the calorific value of Alabama lignite is about one-half that of North Dakota lignite (3,910 ± 1000 vs 7,170 ± 500 Btu/lb) and that the major constituent of lignite is water, on the order of 35-55%.

HAPs do not appear to contribute significantly to emissions at their typical concentrations; each accounts for less than 1.21 TPY, except for F (4.18 TPY) and Se (3.36 TPY).

Alaskan Subbituminous Coal

Data on U.S. Alaskan subbituminous coal have been tabulated[2] on the basis of source data by Swanson et al.[3] and corroborated by White et al.[7]

Based on the typical concentrations of the HAP trace elements in the Alaskan subbituminous coal region, total annual emissions may exceed 84 TPY, with probable Cl emissions of nearly 24 TPY and F emissions of nearly 57 TPY (Table 3). The other trace element HAPs do not appear to contribute significantly to emissions at their typical concentrations; each accounts for less than 2.06 TPY, the calculated value for Mn.

Summary -- Projected Hazardous Air Pollutant Emissions

On the basis of regional calculations involving the typical trace element concentrations of the U.S. coals and lignites studied, even a new, modern, 500-MWe, PCF, electricity-generating power plant would probably exceed the 25-TPY CAAA limit for HAPs. Thus, if electric utilities are included for regulation after the currently mandated EPA studies, plants in these regions may be subject to monitoring and regulation.

ORGANIC COMPOUNDS IDENTIFIED AS HAZARDOUS AIR POLLUTANTS

In contrast to trace element HAPs with their primary dependence on the source coal's composition, the emissions of organic HAPs depend far more heavily on boiler configuration and operating conditions. Hence, these are even more difficult to identify and quantify than the trace elements. Nevertheless, selected organic compounds identified as CAA HAPs were highlighted and characterized in the aforementioned report[2] as particularly important with respect to potential electric utility regulation. These organic compounds include:

- **Acetaldehyde,**
- **Benzene,**
- **Benzidine,**
- **Carbon tetrachloride,**
- **Chloroform,**
- **Dibenzofuran,**
- **Dioxin**
- **Formaldehyde,**
- **Hexachlorobenzene,**
- **Naphthalene,**
- **Phenol,**
- **Toluene,**
- **Trichloroethylene, and**
- **Polynuclear aromatic hydrocarbons (PAHs).**[8]

Various researchers[12-14] have detected and measured specific organic emissions from conventional PCF power plants. Of particular note and significant concentration[12] appear to be the individual organic compounds benzene, ethylbenzene, fluoranthene, phenanthrene, pyrene, toluene, and *o*-, *m*- and *p*-xylenes.[8] These nine compounds represent those of significant concentration as actually detected, not projected or hypothetically calculated.

FUTURE RESEARCH

Several research projects stand out deserving continued activities in characterization, measurement, model implementation, and additional study. Immediate actions include:

1. Incorporation of coal trace element data into existing air transport/dispersion models that forecast emissions.

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[8] The polynuclear aromatic hydrocarbons (PAHs), a subclass of the generic polycyclic organic matter (POM), include more than 100 compounds (e.g., anthracene and benzo(a)pyrene, which have been determined to be active carcinogens).

2. Projection of emissions from real, not hypothetical, coal-fired plants by using actual coal trace element data and knowledge of in-situ control technologies.
3. Identification and quantification of those organic compounds typically emitted from coal-fired plants, to focus regulators on a select group of significantly detrimental organics.

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TABLE 1 Assumed Simplified Emission Factors for HAP Trace Elements from Coal-Fired Power Plants

Element	% Emitted[a]
Sb	2.4
As	2.8
Be	2.0
Cd	2.7
Cl	10.0
Cr	1.0
Co	1.2
F	30.0
Pb	1.8
Mn	2.0
Hg	89.0
Ni	0.4
P	0.1
Se	14.4
Th	0.4
U	1.2

[a] On the basis of input fuel.

TABLE 2 Calculated Annual Trace Element Emissions from a New, Modern, 500-MWe Power Plant Firing U.S. Bituminous Coal (TPY)

Element	Appalachian	Interior	Rocky Mountain
Sb	0.03	0.05	0.01
As	0.81	0.70	0.07
Be	0.04	0.07	0.02
Cd	0.02	0.23	0.02
Cl	**22.3**	**58.2**	**15.9**
Cr	0.22	0.18	0.07
Co	0.09	0.10	0.03
F	**25.9**	**25.6**	**27.8**
Pb	0.30	1.19	0.13
Mn	**13.4**	3.31	0.95
Hg	0.23	0.15	0.07
Ni	0.06	0.14	0.02
P	0.40	0.44	0.25
Se	0.73	0.79	0.30
Th	0.02	0.02	0.02
U	0.02	0.05	0.02
Total	**64.57**	**91.22**	**45.68**

TABLE 3 Calculated Annual Trace Element Emissions from a New, Modern, 500-MWe Power Plant Firing U.S. Lignite or Subbituminous Coal (TPY)

Element	Northern Plains Lignite/Subbituminous	Gulf Lignite	Alaskan Subbituminous
Sb	0.02	0.07	0.09
As	0.14	0.56	0.10
Be	0.02	0.13	0.02
Cd	0.01	0.12	0.01
Cl	**21.5**	**215**	**23.7**
Cr	0.08	0.67	0.33
Co	0.04	0.28	0.09
F	**22.1**	**124**	**56.7**
Pb	0.16	1.20	0.17
Mn	1.67	16.0	2.06
Hg	0.13	0.53	0.09
Ni	0.02	0.27	0.08
P	0.34	4.18	0.27
Se	0.23	3.36	0.52
Th	0.02	0.11	0.04
U	0.02	0.13	0.03
Total	**46.50**	**366.61**	**84.30**

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