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Evaluation of Air Toxic Emissions from Advanced and Conventional Coal-fired Power Plants

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Title III of the 1990 Clean Air Act Amendments (CAAA) mandated that the U.S. Environmental Protection Agency (EPA) evaluate emissions and health risks associated with 189 hazardous air pollutants (HAPs) emitted from the stacks of electric utility steam generating stations [1]. EPA is currently proceeding with the electric utility study and is scheduled to summarize its findings in a report to Congress in November 1995.

In anticipation of the CAAA, the Electric Power Research Institute (EPRI) initiated the Power Plant Integrated Systems: Chemical Emission Studies (PISCES) research program [2]. Parallel to EPRI's efforts, the Department of Energy (DOE) has conducted field measurements as part of three DOE studies: (1) the Clean Coal Technology program, (2) the Comprehensive Assessment of Air Toxic Emissions from Coal-Fired Power Plants program, and (3) an internal program at Morgantown Energy Technology Center (METC) to characterize advanced power systems [3, 4]. The combined EPRI and DOE efforts have sampled every significant configuration of conventional power systems (including fuel type, boiler configurations, particulate control technologies, flue gas desulfurization systems, and NO_x control technologies) as well as a number of advanced power systems. The advanced power systems include a circulating fluidized-bed combustor (CFBC), pressurized fluidized-bed combustor (PFBC), and an integrated gasification combined cycle (IGCC).

This paper evaluates the distribution of various trace metals, trace metal removal efficiencies, and trace substances emissions from these advanced and conventional power systems. The measured emissions from four different power systems are evaluated. This small sample population only allows a screening comparison of flue gas stack emissions from advanced power systems with a conventional pulverized-coal-fired plant. Because the data from the advanced power systems are limited, this paper is not meant to be a detailed comparison of air toxic emissions from the various power system designs. The objective of this paper is to provide the reader with an overview of how the various trace elements are distributed in these power systems (i.e. in what streams are the trace elements discharged) and to provide a first order estimate of expected HAPs emissions.

1. DESCRIPTION OF SITES TESTED

This paper evaluates the air toxics measurements at three advanced power systems and a "base case" conventional power plant. Table 1 provides an overview of the four power plants that were tested. A brief description is provided below:

Integrated Gasification Combined Cycle (IGCC) - DOE and EPRI conducted air toxic measurements at Destec Energy's Louisiana Gasification Technology, Inc.'s (LGTI's) 160 MW IGCC power plant in Plaquemine, Louisiana. The LGTI plant produces medium Btu synthesis gas (syngas) for consumption by two gas turbine power generating units. A low-sulfur, western sub-bituminous coal is gasified in Destec's oxygen-blown, two-staged, entrained-flow, slagging gasifier [5, 6]. Conventional low-temperature clean-up processes are used to remove contaminants from the syngas. For example, during testing, a venturi scrubber was used to control particulates and a Selectamine^{TM} scrubber to control sulfur species. The overall particulate removal efficiency is >99.8%. A process flow diagram illustrating the LGTI system is provided in Figure 1.

Several "state-of-the-art" IGCCs are being constructed as part of the DOE Clean Coal Technology program. Each of these IGCCs will be significantly different from LGTI in the design of the gasification process and the syngas clean-up system - as well as the coal being burned. Thus results from the LGTI plant are not expected to be completely representative of all IGCCs.

Pressurized Fluidized Bed Combustor (PFBC) - DOE conducted extensive air toxics testing at the Tidd PFBC Demonstration Plant located in Brilliant, Ohio. The Tidd plant is a 70 MW bubbling-bed PFBC that burns a Pittsburgh #8 eastern bituminous coal. Dolomite is added, along with the coal, into the bubbling bed for SO_2 control. The flue gas is treated by seven two-stage cyclones (~93% efficiency), prior to a ruggedized gas turbine. Final particulate removal is accomplished with an ESP. The overall particulate removal efficiency is >99.5%. This efficiency is calculated based on the coal analyses and the particulate emissions, and does not include the dolomite added into the PFBC. The Tidd plant is also conducting a hot gas clean-up demonstration. Treated flue gas from one of the primary cyclones is diverted to a ceramic barrier, advanced particle filter which employs silicon carbide candle filters [7, 8, 9]. The particulate removal efficiency for the candle filters is >99.5%. A process flow diagram illustrating the Tidd system is provided in Figure 2.

Circulating Fluidized Bed Combustor (CFBC) - EPRI Site 10 (illustrated in Figure 3) burns a sub-bituminous coal in a nominal 100 MW CFBC. Limestone is fed into the fluidized bed for SO_2 control. Particulate removal is accomplished by a fabric filter, with >99.9% particulate control efficiency [10]. Unfortunately, field results from Site 10 are limited in comparison with the other three field sites. Because of a forced outage of the boiler (due to a tube leak), only one day of sampling could be completed. Because of this unexpected outage, only one run was conducted at Site 10, instead of the triplicate measurements taken at the other field sites. In addition, the CFBC was the first site tested in the EPRI PISCES program. Sampling and analytical methods have evolved a great deal since this testing was conducted, thus, of the results obtained, many are below the method and/or analytical detection limit.

Conventional Pulverized-Coal-Fired (PC) Plant - EPRI Site 12 (Figure 4) was chosen as the base case conventional PC power plant. Site 12 burns a bituminous coal in a 690 MW wall-fired boiler and employs an ESP for particulate control and a wet limestone FGD system for SO_2 control [10]. The particulate removal efficiency of the ESP was 98%, and the overall ESP/FGD particulate removal efficiency was 99.8%. Site 12 is a relatively new unit (commenced operation in 1984) and includes an ESP and wet FGD which are the "conventional" pollution control technologies that may be required at new units.

2. FIELD RESULTS

Table 2 presents the target HAPs analytes for the EPRI and DOE field studies. This is a subset of the 189 HAPs listed in the CAAA and was selected based on expected presence in stack emissions from utility power plant and potential health risks. The target analytes are listed into two groups - the trace elements and the organics. The organics are created during the combustion or gasification processes. Because these compounds are generally present in the vapor phase at stack conditions, organics are not effectively controlled by a conventional particulate control device. The trace elements (such as arsenic,

chromium, mercury, and chloride) are present in the coal. The trace elements can be subdivided into 2 subgroups - depending upon their relative volatility.

1. Particulate phase metals - These metals partition primarily to the solid phase (at conventional particulate control temperatures - 300°F) and are effectively controlled by a conventional particulate control device. These include arsenic (As), cadmium (Cd), chromium (Cr), nickel (Ni), and lead (Pb).

2. Volatile inorganics - These include mercury (Hg), chlorine (Cl), and selenium (Se). This sub-group is relatively volatile at stack gas conditions and is not consistently controlled by a conventional particulate control device (at ~300°F).

Results from EPRI's PISCES and DOE field tests indicate that trace element emissions from conventional power plants can vary by 3 - 4 orders of magnitude. Measurements at the same plant burning similar coal can vary by an order of magnitude from one day to the next. This variability in emissions from plant-to-plant and from day-to-day must be considered when comparing results from different field tests. This variability is due to process conditions (such as the coal) as well as sampling and analytical variability [11].

Trace Elements

Figure 5 compares the mean emission factors for select trace elements for all four power systems. The emissions are generally less than 10 $\mu g/Nm^3$ - which is in the parts-per-billion (ppb) range. The trace metals emissions are generally low for all four power systems. Note the 95% confidence intervals about the means have also been included. For arsenic emissions from the PFBC plant, the confidence interval is relatively good, indicating that the three measurements are close together. However, for nickel emissions from the PC plant, the 95% confidence interval is significant and includes zero. The uncertainty in the mean value can be high and must be considered when comparing results among systems.

Also shown in Figure 5 are the particulate emissions. These are important to note since emissions of particulates and trace metals associated with the particulate phase can be related. There are other factors that may affect this relationship as well. For instance, the particulate emissions may include mist eliminator carry-over from the wet FGD systems or condensibles such as sulfuric acid mist. In the IGCC plant, the particulate loading in the syngas was lower than the particulate emissions from the turbine stack. This could be due to several reasons including ambient particulates in the combustion air to the turbine.

The four power systems burn different coals, with varying heating values (as well as sulfur and trace element analyses). In all cases, the trace element emissions have been normalized to the heating value of the coal. A normalization like this is useful when comparing emissions across various power plants of similar design but different coal heating values. However, the thermal efficiency of the advanced power systems (PFBC and IGCC) are expected to be significantly higher than a conventional PC plant. This must also be considered when comparing expected total emissions from the four power systems.

Figures 6 to 9 illustrate how select trace elements distribute in each of the four power systems. For each of the trace elements, these figures plot the percentage of the input streams (coal and sorbent) associated with each discharge. For example, in Figure 6 (IGCC), the fifth bar represents chromium. For the total coal input of chromium about 120% is discharged with the slag, about 5% is captured in the Selectamine™ scrubber, and <1% is discharged in the sweet water, and <5% in the turbine stack. The total for each bar often does not add up to 100%. This is a result of the difficulty sampling and analyzing for these trace species. Because of these difficulties, a material balance closure (mass amount of species in outlet streams compared to inlet streams) of 70 to 130% is the target goal.

IGCC - The high-temperature, high-pressure gasification process at LGTI produces a slag which captures most of the "major" elements (Fe, Al, Ca) as well as the particulate phase metals (As, Cr, Ni). Two of the volatile inorganics (Hg, Cl) are present in the slag at low levels (see Figure 6). This is consistent since these compounds being in the vapor phase at gasifier conditions. However, a substantial fraction of the selenium was present in the slag. Some removal of trace elements is observed in the venturi particulate scrubber and the Selectamine™ scrubber.

PFBC - At the Tidd demonstration plant, the majority of the particulate phase metals are captured in the bed ash and the cyclone ash (Figure 7). The ESP is a polishing device. Dolomite is calcined in the combustor and used for SO_2 removal. Selenium appeared to be neutralized and absorbed by the calcium sorbent. At the combustor temperatures, the mercury and the chlorides were not effectively captured. The Tidd Demonstration plant includes a slipstream to evaluate hot gas clean-up using silicon carbide candle filters (Advanced Particle Filter - APF). Because of limited measurements, it is difficult to illustrate (in Figure 7) how the trace elements distribute across the APF. As described in a previous paper, most of the trace elements passing through the APF are in the vapor phase. This is in contrast to the ESP, where trace elements penetration is associated with the particulate matter [7].

CFBC - As noted earlier, results from the CFBC are limited due to a forced outage and the fact that a large portion of the results were below the detection limits. The CFBC design is quite similar to a "conventional" PC plant in that it is not a pressurized system and the particulate control occurs at conventional temperatures. Even though the results are limited, they show that the majority of the particulate phase metals are captured in the fabric filter (Figure 8). Because of the calcined limestone, the fly ash is highly alkaline and capable of neutralizing the selenium and chlorides. These can then be effectively removed along with the particulates. Due to analytical difficulties, gas-phase mercury results are not available for the CFBC. The mercury measurements at the CFBC were void due to analytical difficulties.

Conventional PC Plant - The majority of the particulate phase metals in the conventional PC plant are captured in the ESP ash (Figure 9). The volatile inorganics are captured in the FGD system in a manner similar to the Selectamine^{TM} scrubber at the LGTI plant. It is important to note that mercury is not consistently captured at all FGD systems and the removal efficiency may be related to the form of mercury - whether it is oxidized (i.e. $HgCl_2$) or elemental Hg [10].

Particulate Phase Metals

The particulate phase metals are generally associated with the fly ash particulates (at conventional particulate control temperatures around 300°F) and are effectively controlled by conventional particulate control device (e.g. ESP, fabric filter). Figure 10 compares the removal efficiencies for select particulate phase metals for each of the four power systems described. The particulate removal efficiency is also shown. A more efficient particulate control device will tend to have better particulate phase metal removal efficiencies as well. The removal efficiencies are calculated based on the total emissions and the coal analyses.

Volatile Inorganics

The volatile inorganics are usually not captured by a conventional particulate control device. Under both the EPRI's and DOE's field studies, in a limited number of conventional PC plants, mercury was associated with the particulate phase, and thus captured by the ESP or fabric filter. The reason for this is not understood. Mercury removals across a wet FGD system have also been seen and are highly variable. The removal efficiency appears to be dependent on the form of mercury in the flue gas [10]. It is apparent from the studies completed to date, that mercury control is not completely understood. EPRI and DOE are sponsoring additional research in this area.

Selenium and HCl (from the chlorine) are also volatile compounds and would generally pass through an ESP or fabric filter - unless absorbed on the particulates. Selenium behaves similarly to sulfur in that it forms SeO_2 during combustion and can be neutralized and absorbed by an alkaline sorbent such as lime/limestone as well as by the alkalinity in the fly ash particles. Hydrochloric acid can also be neutralized by an alkaline particle, but the thermodynamics only become favorable at conventional particulate control temperatures (~300°F). Both selenium and HCl are effectively removed by a wet FGD system [10].

Figure 11 compares the removal efficiencies of the volatile inorganics for all four power systems. It is important to note that removal efficiencies are related to equipment design as well as site-specific factors such as the initial concentration in the fuel. For example, the CFBC and the IGCC burn low-sulfur sub-bituminous coals, thus sulfur removal efficiencies may be lower for than if the sites burned a higher sulfur coal. Both the conventional PC plant and the IGCC employ a wet sulfur removal system. The IGCC plant also employs a wet particulate scrubber. Both plants achieve good (>60%) removal of the volatile inorganics. The fluidized bed combustors employ a limestone bed for SO_2 removal. The alkaline particulates appear to effectively remove selenium and HCl in both the PFBC and the CFBC. Very little mercury removal was observed at the PFBC. The mercury measurements at the CFBC were not meaningful due to analytical difficulties.

Organic Compounds

Organics are generally produced during the combustion process, and efficient combustion may lead to very low levels of these compounds. The emissions of organics at each of the four power systems were generally low (at the parts-per-billion levels). Figure 12 plots the concentration of benzene and toluene as well as the benzo(a)pyrene [B(a)P] equivalents for all four power systems. The polynuclear aromatic hydrocarbons (PAHs) include a number of compounds. The B(a)P equivalent provides a method of summing all the various PAHs. B(a)P equivalents were calculated using the EPA protocols of multiplying the detected PAHs by the weighted equivalency factors.

3. SUMMARY

HAPs measurements for the advanced power systems are limited, thus it is not possible to develop any definitive comparisons in regards to emissions from the various power systems. For each advanced power systems, only one set of measurements was conducted, and this was limited to burning one coal. Each of the four power systems burned different coals. The advanced power systems are first generation designs and future plants may incorporate

more "state-of-the-art" designs which may likely reduce total expected emissions. EPRI Site 12 was chosen as the "base case" conventional plant, and by no means, does it represent all conventional power systems. When one considers that emissions from the EPRI PISCES and DOE tests (over 50 conventional power systems tested) varied several orders of magnitude, it is not possible to develop conclusions in regards to which power system has higher or lower emissions. It would not be appropriate to estimate emissions for all PC plants using the emissions from Site 12, and, similarly, it is not appropriate to estimate emissions from another IGCC, PFBC, or CFBC based on the measurements of one field site.

What can be concluded from these field measurements is that the air toxics emissions (including the trace elements and the organic compounds) from all four power systems were generally low and were in the parts-per-billion (ppb) levels. Removal efficiencies of the particulate phase metals (e.g. As, Cr, Ni) were consistently greater than 90%. Removals of the volatile inorganics (e.g. Hg, Se, and Cl) were more variable and, in general, less efficient.

4. ACKNOWLEDGMENTS

The authors would like to acknowledge the Radian Corporation who conducted the sampling and analytical measurements at the four power plants discussed in this paper. In addition, we would like to thank the various host utilities including American Electric Power, Destec Energy, and New York State Electric & Gas who co-sponsored and hosted the various field measurements.

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Table 1: Summary of Advanced Power Systems and Base Case Conventional Power System

Power Plant System	Coal Type	Coal % S	Nominal Size (MW)	Particulate Control	SO_2 Control
Integrated Gasification Combined Cycle	Sub	0.4	160[1]	Venturi scrubber	Selectamine scrubber
Pressurized Fluidized Bed Combustion[2]	Bit	3.4	70	Cyclone + ESP	Limestone injection
				Cyclone + Candle filters	
Circulating Fluidized Bed Combustion	Sub	0.5	100	Fabric Filter	Limestone injection
Pulverized Coal[3]	Bit	2.8	690	ESP	Wet FGD

[1] Net power production including both electricity and steam.

[2] A slipstream of treated flue gas from one of the primary cyclones is diverted to a ceramic barrier, advanced particle filter which employs silicon carbide candle filters.

[3] A pulverized coal-fired boiler with an ESP and wet FGD system was chosen as the base case conventional plant. The base case plant is EPRI Site 12 [10].

Table 2: Target HAPs Grouping — EPRI/DOE Field Studies

Particulate Phase Metals	Volatile Inorganics	Organics
Antimony	Mercury	Benzene
Arsenic	Selenium	Toluene
Beryllium	Chlorine/HCl	Formaldehyde
Cadmium		PAHs *#
Chromium	**Others**	Dioxins/Furans *
Cobalt	Mercury speciation *	
Lead	Radionuclides *	
Manganese		
Nickel		

* Measured at selected plants.

\# PAH - Polycyclic Aromatic Hydrocarbons

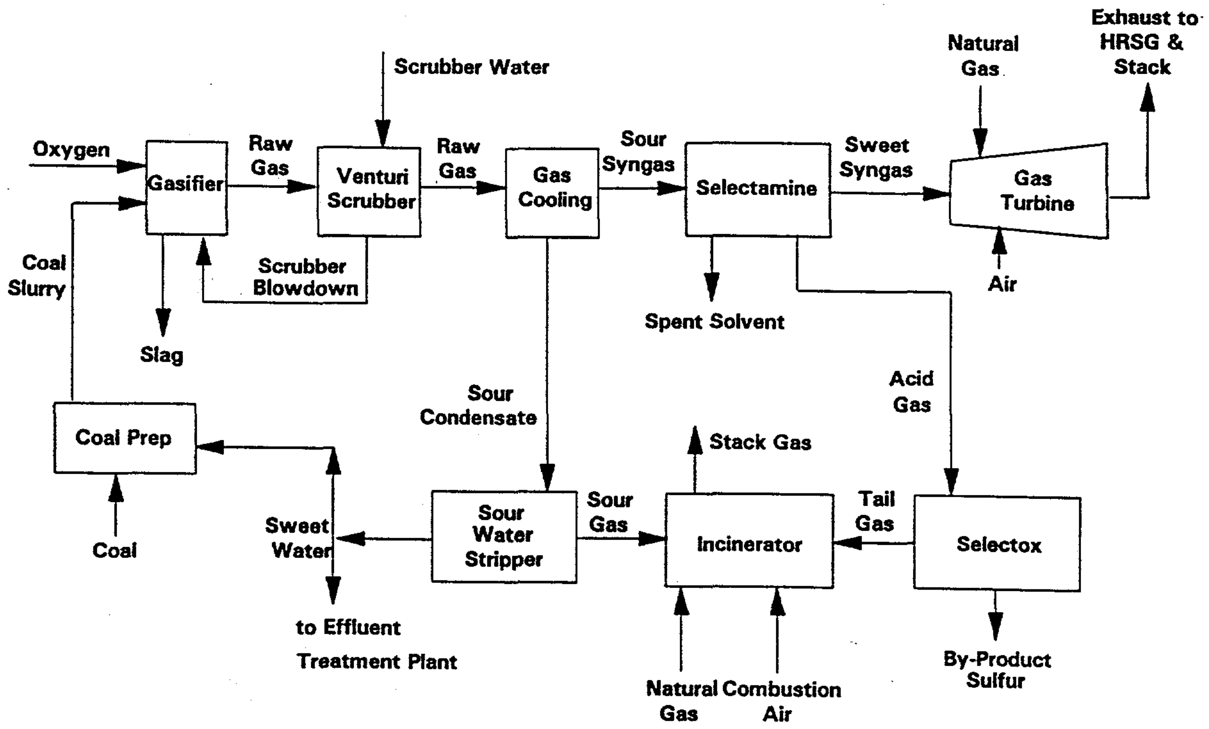

Figure 1 - IGCC Process Flow Schematic

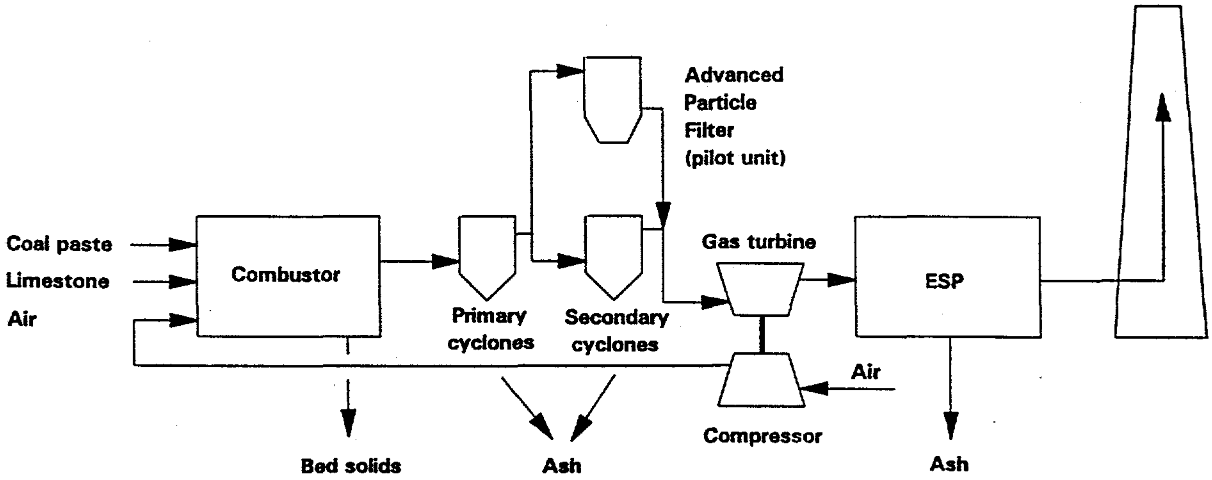

Figure 2 - PFBC Process Flow Schematic

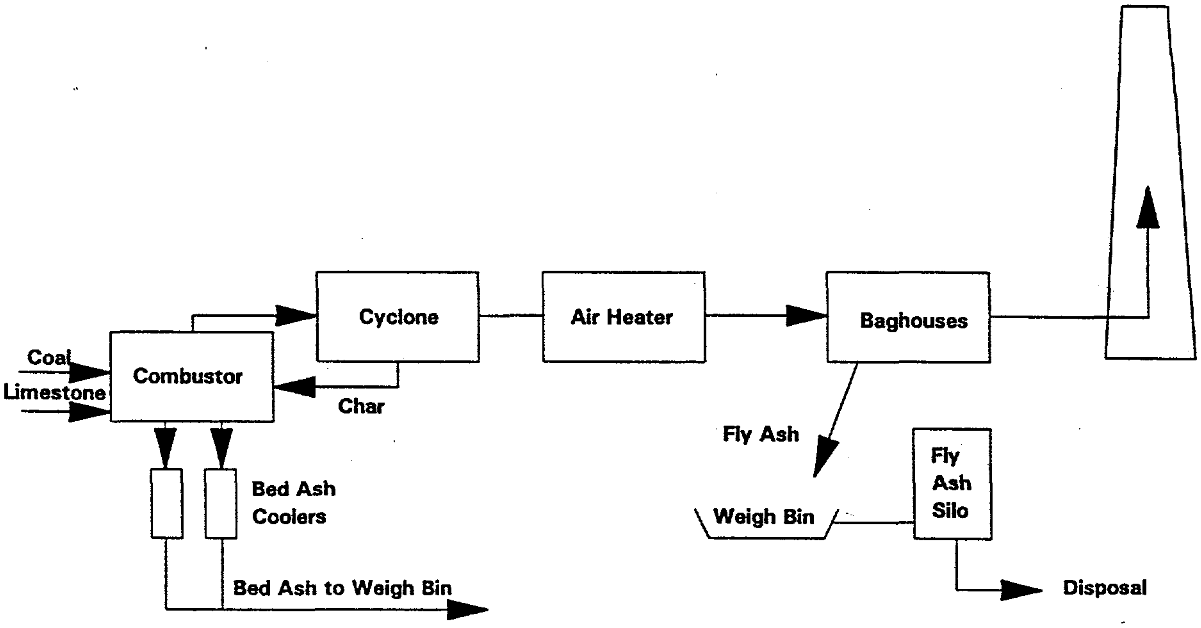

Figure 3 - CFBC Process Flow Schematic

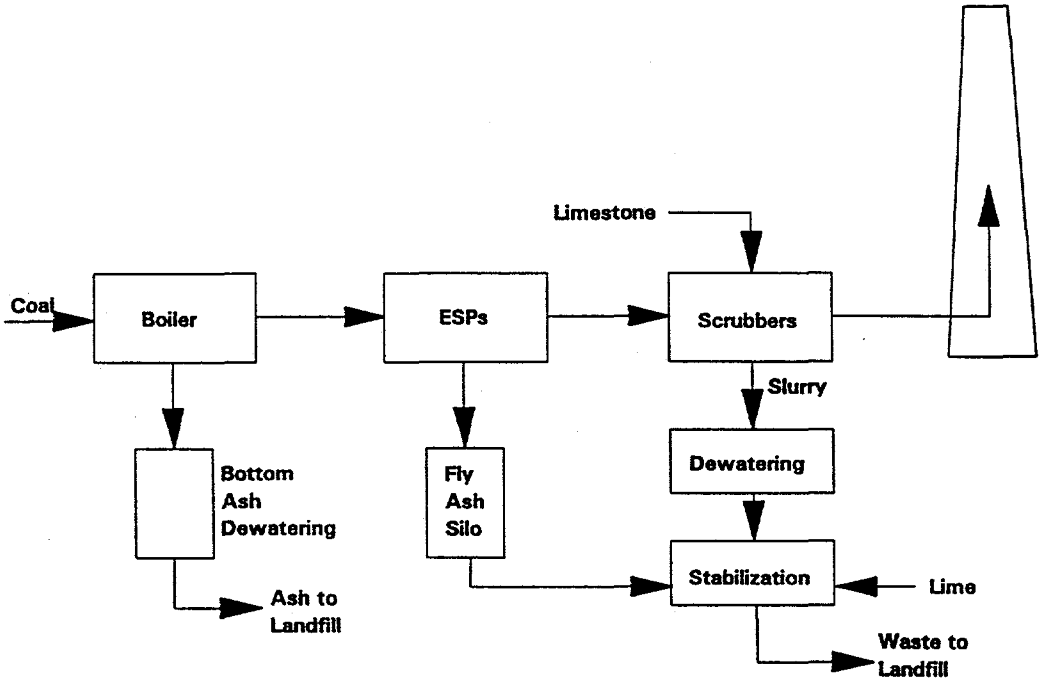

Figure 4 - Conventional PC Plant Process Flow Schematic

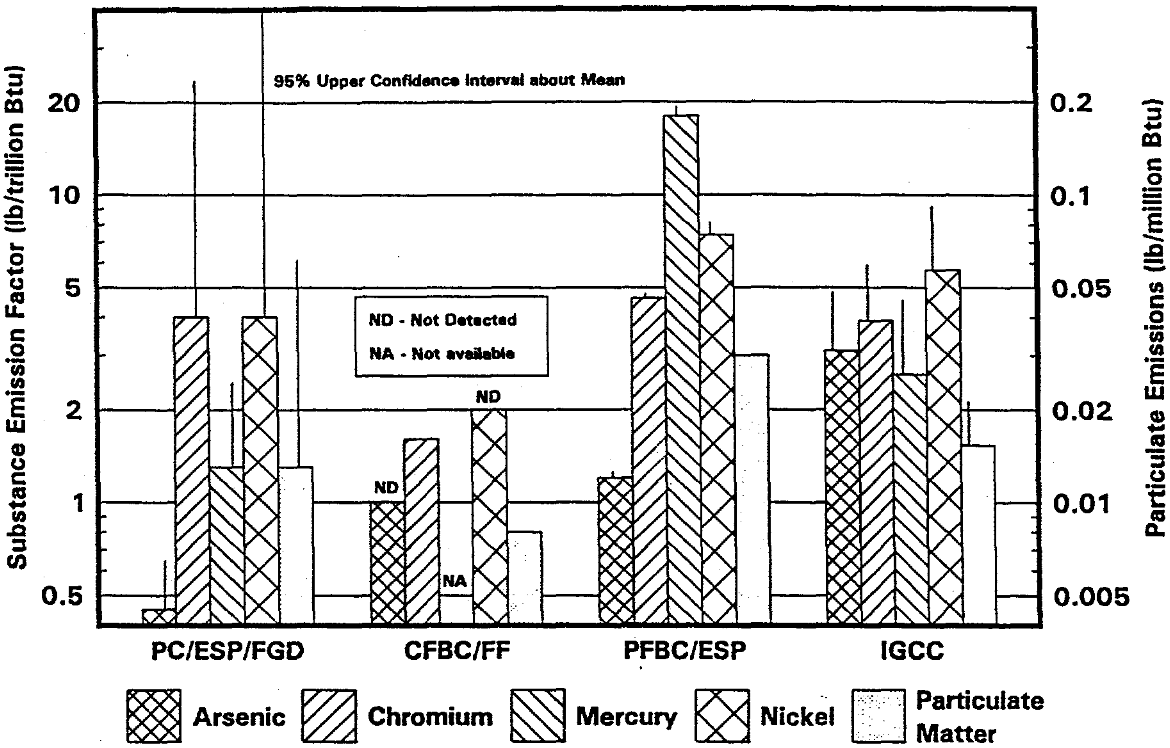

Figure 5 - Comparision of Trace Metal Emission Factors
PFBC particulate emissions are based on monthly measurements conducted by the utility.

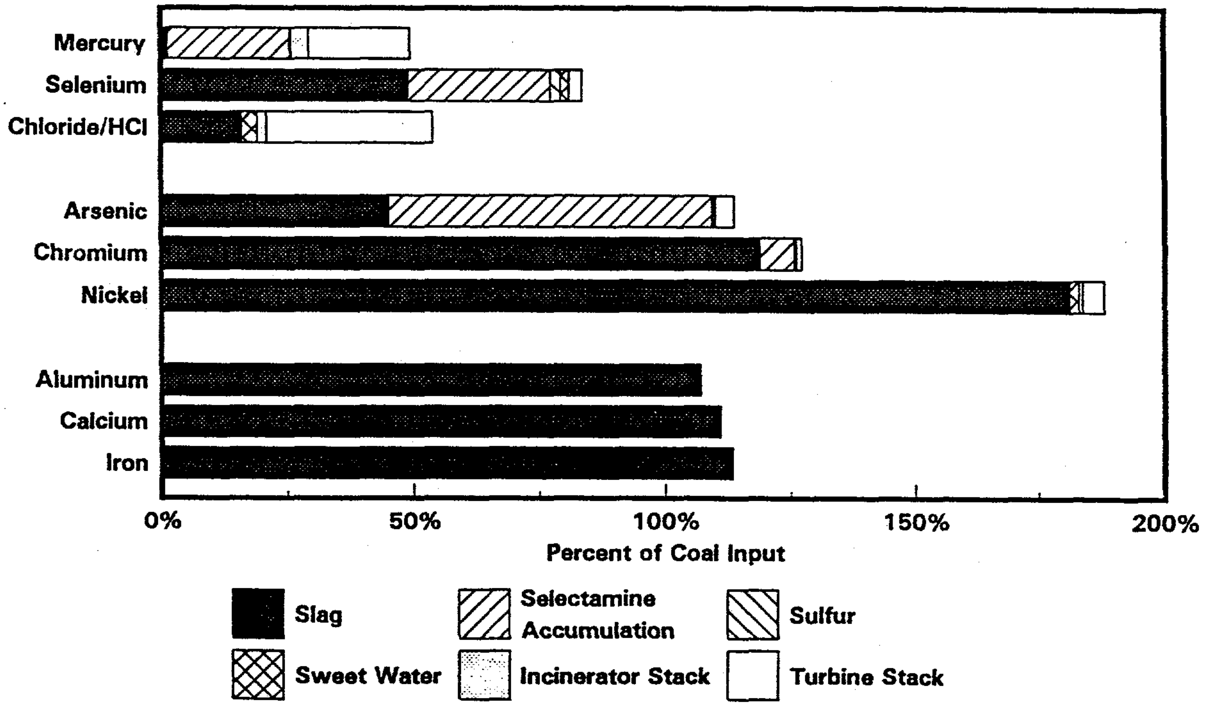

Figure 6 - Distribution of Trace Elements at the IGCC Plant

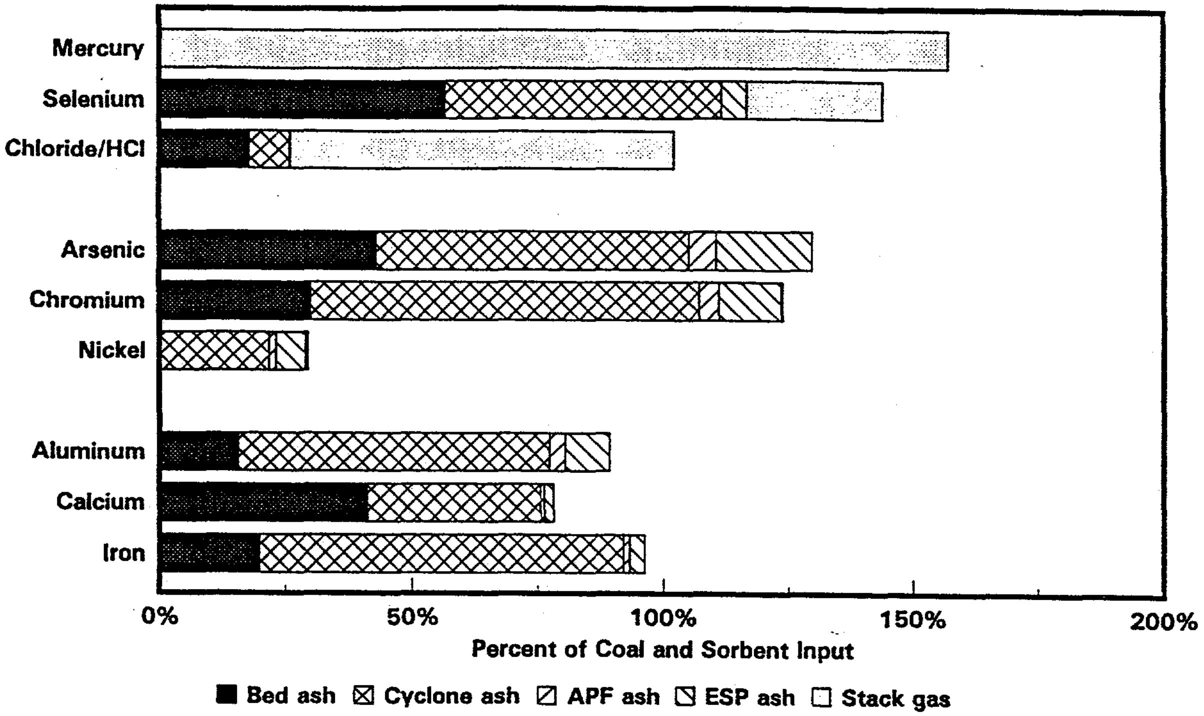

Figure 7 - Distribution of Trace Elements at the PFBC Plant

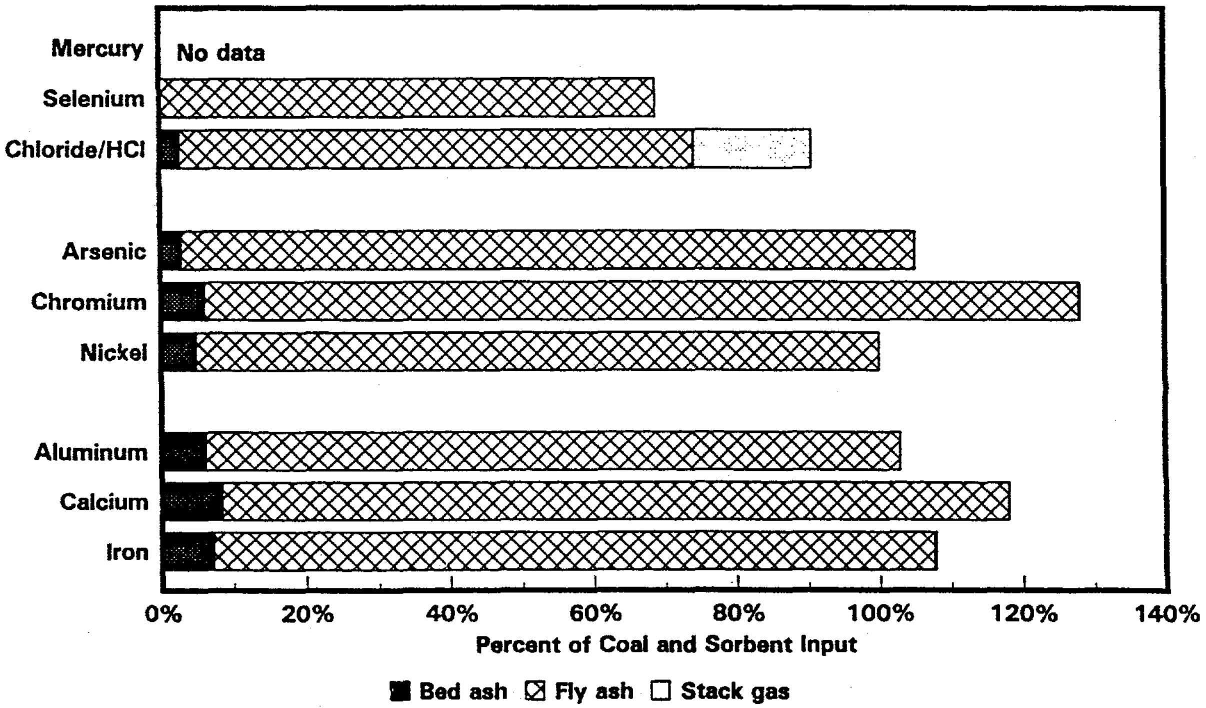

Figure 8 - Distribution of Trace Elements at the CFBC Plant

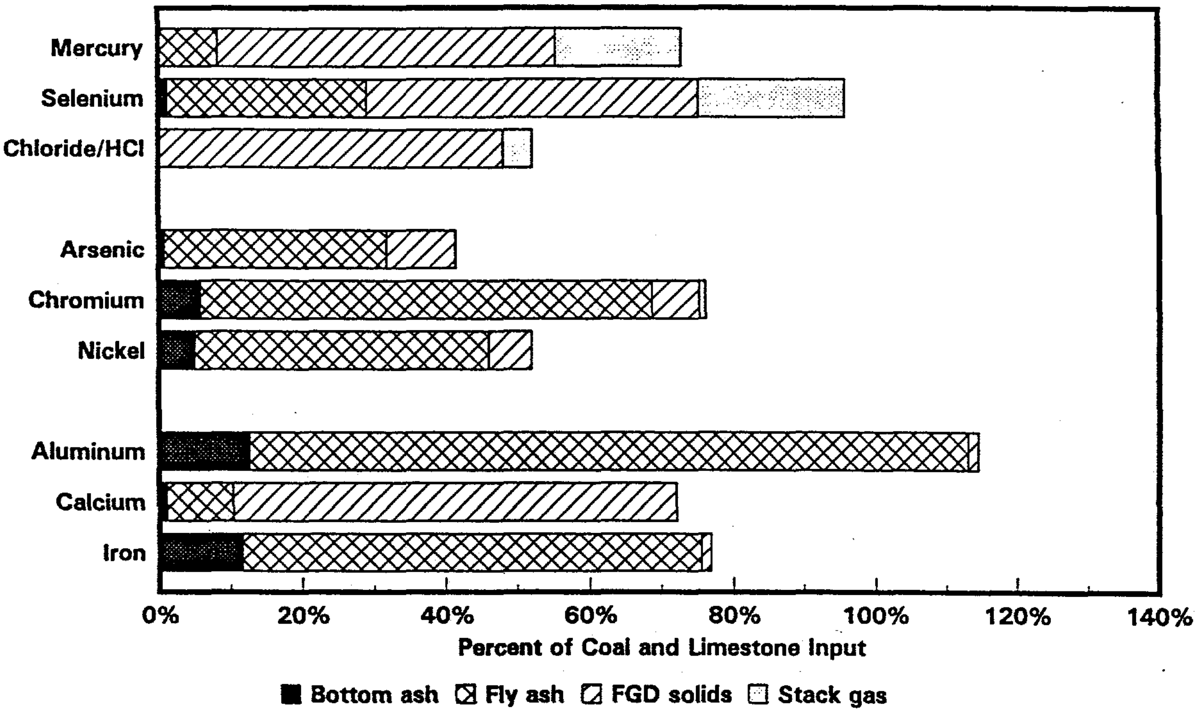

Figure 9 - Distribution of Trace Elements at the Conventional PC Plant

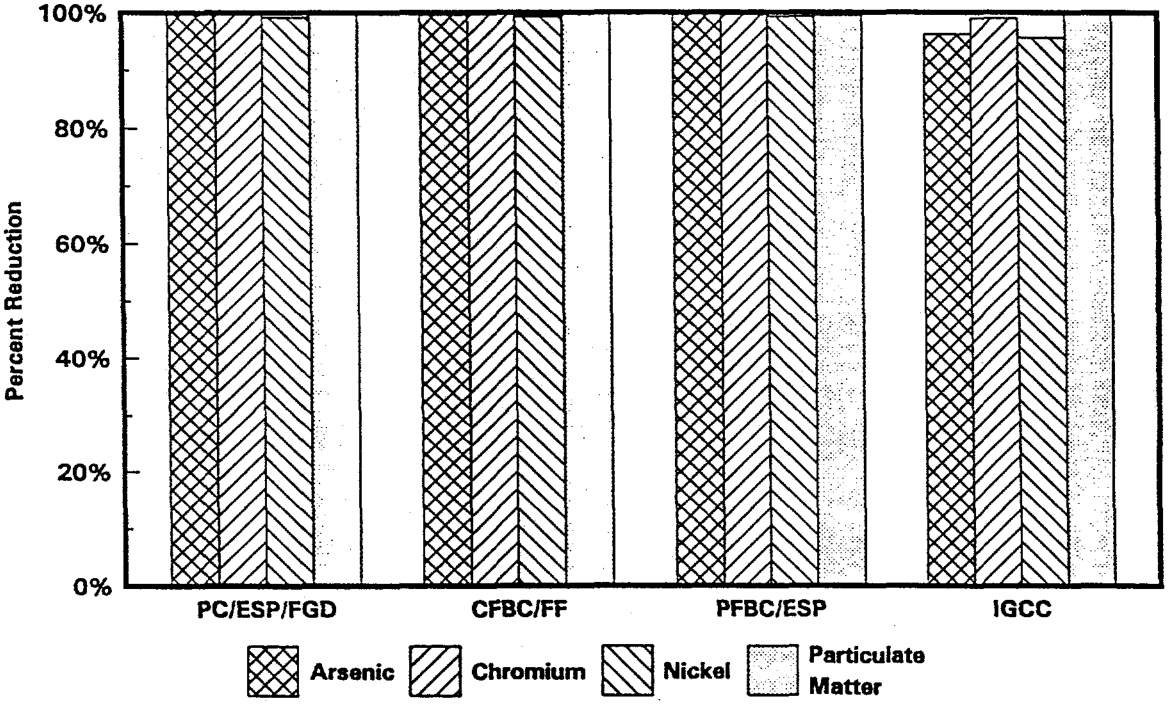

Figure 10 - Comparison of Particulate Phase Metal Removal Efficiencies
Removal efficiencies are based on the trace metal/particulate emissions and the coal analyses

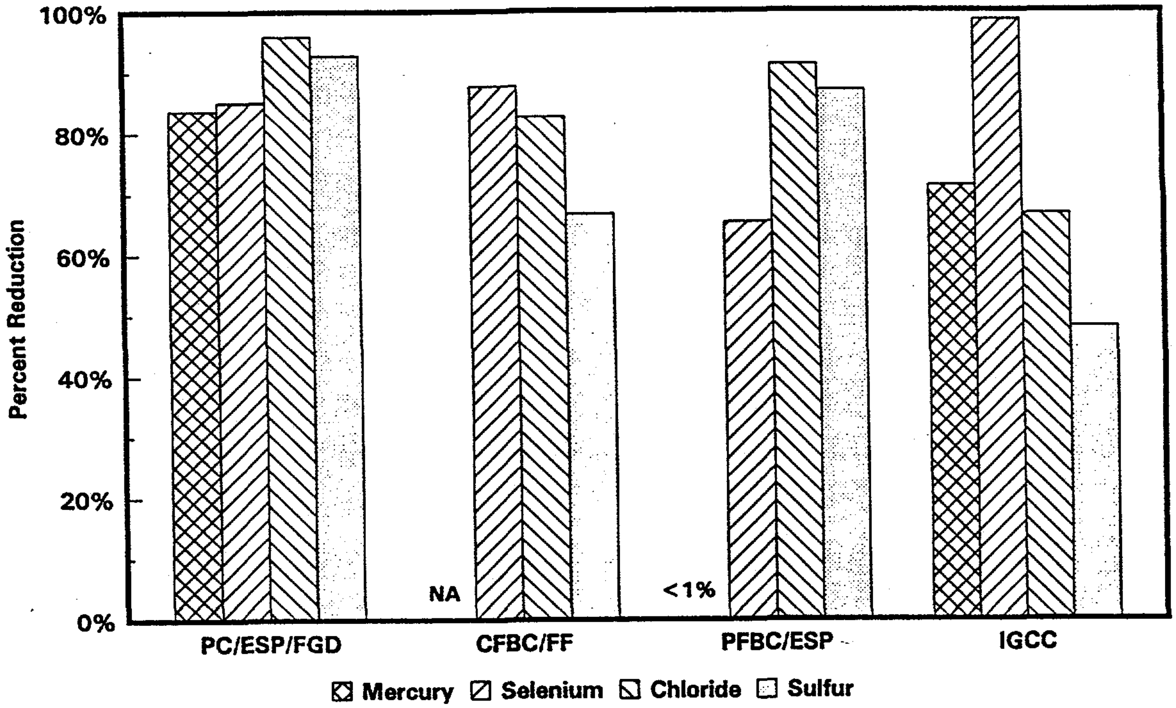

Figure 11 - Comparison of Volatile Inorganic Removal Efficiencies
Removal efficiencies are based on the trace metal/particulate emissions and the coal analyses

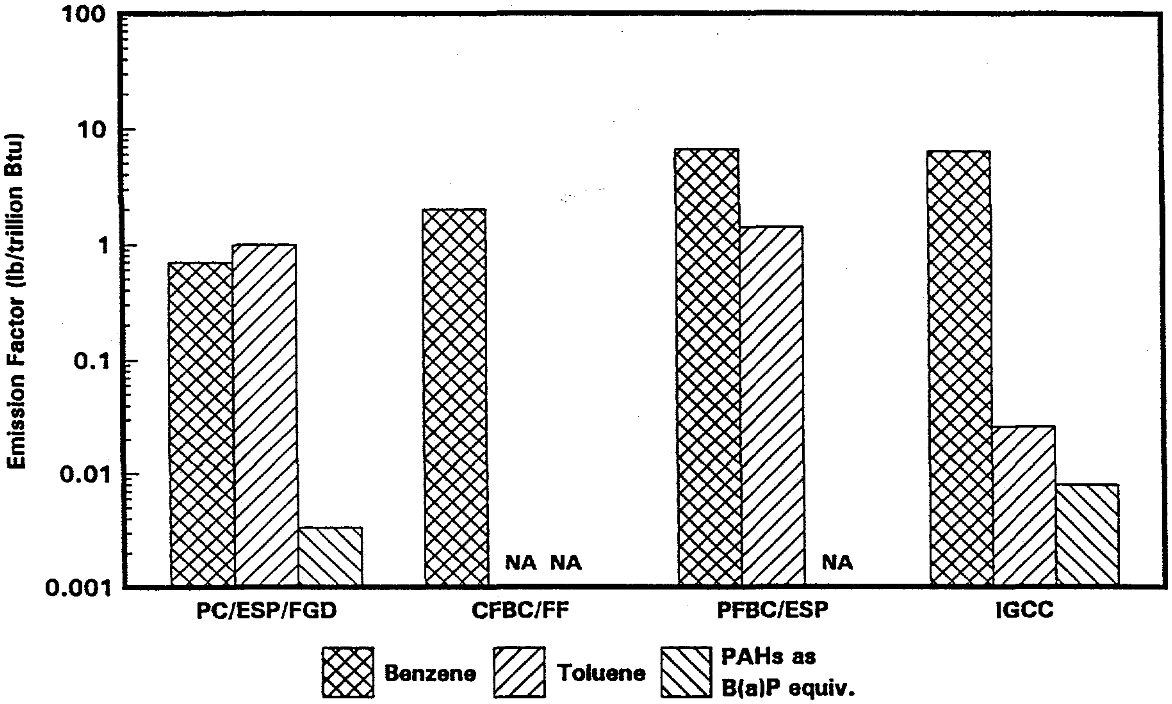

Figure 12 - Comparison of Organic Emissions