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Project Summary

Evaluation of Relative Environmental Hazards from a Coal Gasifier

S. K. Gangwal, J. G. Cleland, and R. S. Truesdale

During the past 4 years, a laboratory-scale coal gasification facility was developed and used to study the generation and environmental assessment of pollutants from coal gasification operations. Detailed chemical analyses of the four effluent streams, namely gas, aqueous condensate, tar, and ash, were performed for more than 30 runs in which a variety of coals ranging from lignite to bituminous were gasified.

Brief descriptions are given for the gasification reactor and the associated sampling and analysis system. Problems encountered with analysis and special techniques for analysis of complex samples are described. The relative environmental hazards of the various effluent streams are determined, using multimedia environmental goals (MEG) methodology. Toxicity and mutagenicity of the streams are assessed using bioassays.

More than 400 constituents are identified in the various effluent streams. Environmentally significant and nonsignificant constituents are ranked according to discharge severity. The nonsignificant constituents are candidates for elimination in future research. Finally, data from the laboratory gasifier are discussed in relation to those reported on large-scale processes.

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back.

Introduction

Under the sponsorship of the Fuel Process Branch of the U.S. Environmental Protection Agency's Industrial Environmental Research Laboratory, Research Triangle Park, N.C., Research Triangle Institute (RTI) conducted an experimental research program to study pollution problems associated with coal gasification. A laboratory-scale coal gasification reactor and sampling system was installed and more than 60 tests were conducted using a variety of U.S. coals. The gasifier was operated with coal, steam, and air at about 1000°C and 200 psig to yield a low-Btu gas as a primary product and aqueous condensate, tar, and ash (reactor residue) as by-products. Each of these four product and by-product streams was characterized in detail, using modern chemical analysis and bioassay techniques. More than 400 constituents were identified in the effluent streams and some 100 constituents were quantitated for more than 30 gasification tests.

Using the Environmental Protection Agency's multimedia environmental goals (MEG) methodology, the quantitated constituents were grouped into insignificant and significant constituents from an environmental (health) hazard point of view. The significant constituents were than ranked according to their severity of discharge. Ranges, 95 percent confidence intervals, arithmetic means, and geometric means for the stream concentrations and production factors (amount produced per unit amount of coal gasified) of the significant constituents were plotted as bar charts. Data for the stream production factors from the RTI gasifier were compared to those available in the literature for large-scale gasifiers. Also, data for the significant stream constituent concentrations and production factors were compared to those available in the literature for larger-scale gasifiers, coke plants, coal liquefaction units, combustors and incinerators, ambient background levels, and regulated levels.

Results and Discussion

The complete report which this publication summarizes contains data for significant stream constituents from more than 30 gasification tests. These results are presented as bar graphs (including minimum, maximum, 95 percent confidence interval, arithmetic mean, and geometric mean) of concentrations and production factors. Typical figures appearing in the report for concentration and production factors are shown here as Figures 1 and 2 for the product gas stream.

In Figure 1, concentrations of most significant product gas constituents are plotted versus their DMEGs. Here DMEG is a health-based stream concentration and is defined as a concentration of a pollutant in an undiluted effluent stream which will not adversely affect people exposed for short periods of time. DMEG is generally derived using existing toxicity and biological data; a complete explanation is available in the report. Here it is sufficient to say that the lower the DMEG value is for a certain pollutant, the more hazardous is the pollutant. In Figure 1, the 45° line (dotted) corresponds to concentration = DMEG. The concentration bars include the minimum, maximum, arithmetic mean, geometric mean, and 95 percent confidence interval for the product gas constituents of greatest significance. The significance of the 45° line is that it allows easy visual inspection of the relative degree of significance and relative hazard of the various constituents. The higher the bars or means are above the 45° line the greater are the hazards. Also constituents plotted on the left of Figure 1 have lower DMEGs and are consequently more hazardous at the same concentration levels.

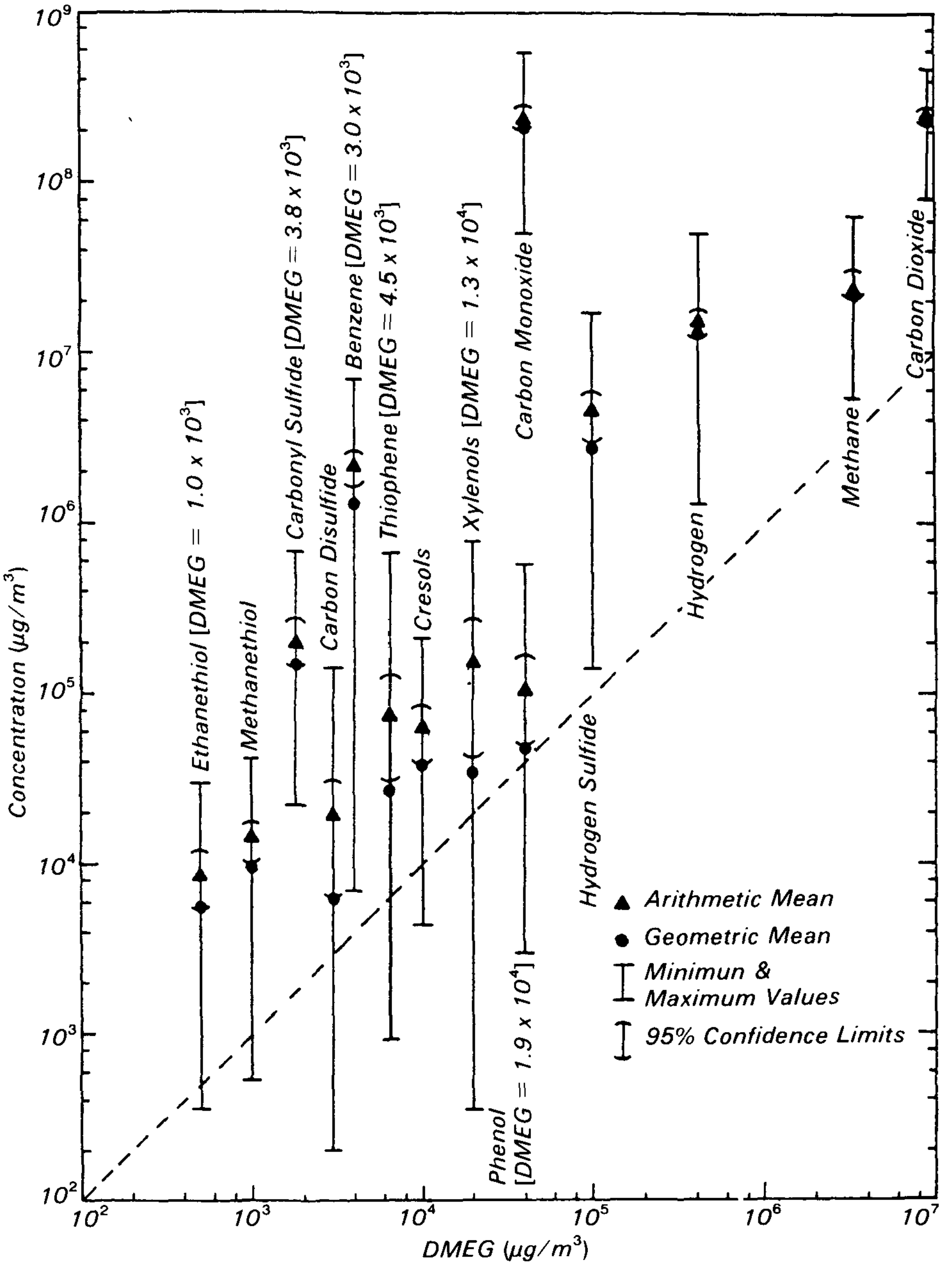

Figure 1. *Ranges of concentration for most significant product gas constituents.*

In Figure 2, the production factors (expressed as μg produced per g of coal gasified) of the significant product gas constituents are shown. Again minimum, maximum, arithmetic mean, geometric mean, and 95 percent confidence interval are shown on production factor bar graphs. The constituents in Figure 2 are listed in order of increasing discharge severity from left to right. Here discharge severity is defined as the arithmetic mean concentration divided by DMEG and represents a hazard factor associated with each constituent. Thus, CO (the product that one is trying to maximize) represents the highest product gas hazard. Figure 2 gives an immediate account of how much of a constituent to expect from the coal gasifier, i.e., how much of a by-product can be produced (e.g., benzene, toluene, and xylenes) or how much of a pollutant needs to be controlled (e.g., H_2S and carbonyl sulfide).

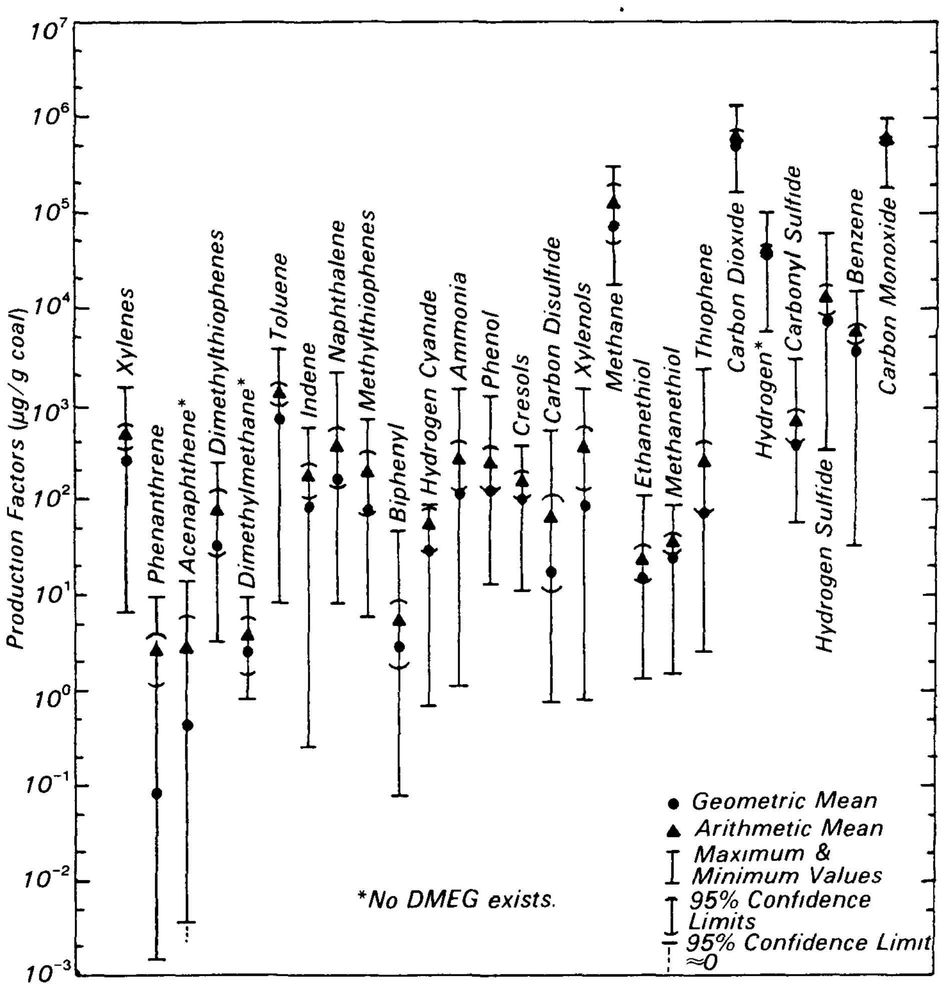

Figure 2. *Ranges of production factors for significant product gas constitutents.*

Figures similar to 1 and 2 are available in the complete report for the aqueous condensate, tar, and ash streams. The report also contains detailed comparisons of total stream production and significant pollutant production from the RTI gasifier to those from larger-scale gasifiers, coal liquefaction units, coke plants, ambient background levels, and regulated levels. Typical comparison tables are given here as Tables 1 through 3. In Table 1, the overall stream production from larger-scale gasifiers is compared to that from the RTI laboratory gasifier. The aqueous condensate production is greatly influenced by the gas cooling process imposed and end use. Thus, the Wellman-Galusha process produces no condensate because the entire hot gas is combusted. Thus, any moisture would end up in the combustion flue gas. The Chapman-Wilputte process uses an evaporator system, and no condensate is discharged as liquid. No tars are produced from the Wellman-Galusha and the Koppers-Totzek gasifiers because the former uses an anthracite coal and the latter is an oxygen-blown entrained-bed process. For the air-blown gasifiers, RTI gas production appears to be low, presumably because of a lower throughput in the laboratory reactor. Ash is influenced by coal type, but in general the RTI range compares favorably to that of other gasifiers. Tar production compares favorably to METC, GFETC, and Lurgi but not to Chapman-Wilputte.

In Table 2, the laboratory gasifier product gas is compared to those obtained from other gasifiers. The 95 percent confidence interval of concentration and production factors for the laboratory gasifier compares quite favorably to values for larger-scale gasifiers, especially low-Btu gasifiers. This

Table 1. *Comparison of Effluent Stream Production from Different Gasifiers*

	95% Confidence Range (This Study)	*Chapman Wilputte*	*Wellman Galusha (Anthracite Coal)*	*METC*	*GFETC (Oxygen blown)*	*Lurgi (Oxygen blown)*	*Koppers Totzek (Oxygen Blown)*
Gas m^3/kg coal	*2.4-3.0*	*2.9*	*5.4*	*2.8-6.0*	*0.98-1.1*	*1.2*	*2.0*
Aqueous Condensate L/kg coal	*0.49-1.1*	*0*	*0*	*1.5-4.9*	*0.2-0.33*	*0.82*	*0.22*
Tar g/kg coal	*15-23*	*100*	*0*	*11-47*	*16-23*	*21*	*0*
Ash kg/kg coal	*0.14-0.26*	*0.06*	*0.9*	*0.07-0.18*	*0.06-0.07*	*0.18*	*0.24*

lends credence to the RTI gasifier's ability to simulate large-scale processes with respect to both pollutant production and production of major gas constituents. The only gas constituent for which the RTI concentration appears low in comparison to other gasifiers is ammonia. This may be caused by low scrubber efficiency for ammonia capture. Fortunately, the majority of the ammonia condenses out as aqueous ammonia in the condensate and thus the low gas value introduces little error to overall ammonia production. Also adequate comparison with other gasifiers is difficult because of differences in condensation or quench temperatures and procedures. Tables similar to Table 2 are available in the report for aqueous condensate, tar, and ash streams.

Comparisons of pollutant levels in the RTI gasifier condensate with pollutant levels in the aqueous effluents from other coal conversion processes, ambient levels in U.S. waters, and regulated levels are given in Table 3. Comparisons with most stringent effluent standards for the U.S. and Canada (and with end-of-pipe effluent standards set by the State of Illinois) suggest that, in certain locations, aqueous effluents will have to be processed to reduce the levels of all the pollutants listed. Considering the variation in the chemical nature of these pollutants, a diverse approach may be necessary when deciding on wastewater treatment options; several techniques will have to be combined for proper treatment in areas, such as Illinois, with strong regulation of effluent composition.

Comparison of the levels of the pollutants (ammonia, sulfides, thiocyanates, phenol, and cyanides) in the RTI condensate with coke plant liquors suggests that these streams are very similar in gross chemical composition. Point source effluent limitations have been established by the EPA for aqueous effluents from by-product coking plants under the Clean Water Act. Cyanide, phenol, ammonia, and sulfide are regulated under these source-based effluent limitations. As a result, control techniques have been developed for these streams and are presently well-established technology. Because of the similarity between coke plant liquor and gasifier condensate, this technology from the coking industry can probably be easily applied to gasifier aqueous condensate.

Similarly, point source effluent limitations also have been established for certain aqueous effluents from petroleum refining; techniques developed for pollution control in these streams may also be applicable to gasification wastewater. Thus, the basic technology for the control of major pollutants in gasifier wastewater streams is already available, although certain modifications may be necessary for the proper application of these technologies. Tables similar to Table 3 are available in the complete report for product gas, tar, and ash streams.

Conclusions

Significant conclusions from this study are:

1. Efficient fixed-bed gasification of caking coals presents problems because of the tendency of these coals to agglomerate in the gasifier, leading to poor gasification rates and heat transfer characteristics.

Table 2. *Comparison of Concentrations [μg/m^3] and Production Factors [μg/g coal]+ of Significant Product Gas Constituents from Different Gasifiers*

	95% Confidence Ranges for This Study	METC (Average for 7 Coals)	Range (3 Coals) Air-Blown Synthane	Wellman-Galusha (Penn.-Anthr. Coal)	Koppers-Totzek (Oxygen-Blown)	Dry Ash Lurgi
Carbon Monoxide	2.0E8-2.8E8	2.6E8	1.3E8-2.0E8	—	7.3E8	—
Benzene	1.6E6-2.6E6 (4.2E3-7.0E3)	—	2.4E6-3.4E6 (4.8E3-7.4E3)	—	—	
Hydrogen Sulfide	3.0E6-6.0E6 (8.4E3-1.8E4)	5.5E6	8.8E5-7.6E6 (2.0E3-1.3E4)	8.9E5 (4.9E3)	4.3E6-2.6E7 (6.6E3-4.6E4)	6.7E6 (9.6E3)
Carbonyl Sulfide	1.4E5-2.8E5 (3.8E2-8.8E2)	—	5.4E4-3.8E5 (1.2E2-7.6E2)	2.2E5 (1.2E3)	5.5E5-1.8E6 (8.0E2-3.2E3)	—
Hydrogen	1.2E7-1.8E7	1.3E7	2.0E7-2.7E7	—	2.9E7	—
Carbon Dioxide	2.2E8-2.8E8	1.9E8	3.7E8-3.8E8	—	2.1E8	—
Thiophene	2.9E4-1.3E5 (7.0E1-4.1E2)		<1.9E4-2.6E5 (<3.8E1-4.4E2)	—	—	—
Methanethiol	1.0E4-1.8E4 (2.6E1-4.4E1)	—	1.7E4-8.6E4 (3.4E1-1.5E2)	— —	— —	— —
Methane	2.1E7-3.1E7	1.8E7	3.1E7-4.0E7	—	0-8.3E5	—
Ammonia	4.9E4-1.3E5 (1.2E2-4.0E2)	—	—	1.2E5 (6.9E2)	—	—
Toluene	3.9E5-2.5E5 (9.4E2-1.7E3)	—	2.9E5-9.0E5 (5.8E2-1.8E3)	—	—	—
Xylenes	1.3E5-6.9E5 (3.4E2-6.2E2)	—	9.5E4-2.8E5 (1.9E2-6.2E2)	—	—	—

continued

Table 2 (continued).

	95% Confidence Range for This Study	Bigas	GFETC (Oxygen Blown)	CO_2 Acceptor	Chapman Wilputte	Woodall Duckham	Winkler	Lurgi (Oxygen Blown-Range for 5 Coals)
Carbon Monoxide	2.0E8-2.8E8	2.8E8	7.5E8	—	2.6E8	3.7E7	2.9E8	2.0E8-2.7E8
Benzene	1.6E6-2.6E6 (4.2E3-7.0E3)	—	—	—	—	—	—	—
Hydrogen Sulfide	3.0E6-6.0E6 (8.4E3-1.8E4)	8.5E6 (3.9E4)	8.7E5 (1.5.E3)	1.6E6 (2.1E3)	4.0E5	—	—	7.0E6-1.6E7
Carbonyl Sulfide	1.4E5-2.8E5 (3.8E2-8.8E2)	3.0E6* (1.3E4)*	7.6E4* (1.3E2)*	7.4E4 (9.7E1)	—	—	—	—
Hydrogen	1.2E7-1.8E7	1.4E7	2.8E7	—	1.4E7	1.6E7	1.3E7	3.7E7-3.9E7
Carbon Dioxide	2.2E8-2.8E8	1.5E8	1.6E8	—	1.8E8	3.7E8	1.5E8	6.0E8-6.5E8
Thiophene	2.9E4-1.3E5 (7.0E1-4.1E2)	—	—	—	—	—	—	—
Methanethiol	1.0E4-1.8E4 (2.6E1-4.4E1)		4.9E4** (8.5E1)**					
Methane	2.1E7-3.1E7	3.2E7	4.0E7***	—	1.1E7	2.0E7	7.5E6	6.7E7-8.4E7
Ammonia	4.9E4-1.3E5 (1.2E2-4.0E2)	3.3E6 (1.6E4)	—	4.2E6 (5.5E3)	—	—	—	—
Toluene	3.9E5-6.9E5 (9.4E2-1.7E3)	—	—	—	—	—	—	—
Xylenes	1.3E5-2.5E5 (3.4E2-6.2E2)	—	—	—	—	—	—	—

+Production factors in parenthesis.
—Blanks in table indicate values not available.
*Includes carbon disulfide.
**Includes other thiols.
***Includes C_2H_4.

2. Glass capillary gas chromatography (GC^2) with specific element detection is a complementary analytical technique to gas chromatography/mass spectrometry (GC/MS) for characterization of complex coal gasifier tar samples. Identification and quantitation of high molecular weight sulfur heterocyclics and primary aromatic amines is difficult because they are present at very low concentrations (~ ppm) in an extremely complex tar matrix.
3. The most significant coal gasification effluent stream from an environmental standpoint is aqueous condensate, followed by tar, product gas, and ash. However, on an equivalent weight basis, the tar stream is more toxic and mutagenic than aqueous condensate based on cytotoxicity and bioassy tests. Polycyclic aromatic hydrocarbons (PAHs) and tar bases are the most mutagenic of the various tar fractions. Coal pyrolysis and gasification at higher temperatures (as in continuous coal feed versus batch feed) leads to reduced tar mutagenicity. The ash stream is very likely nonhazardous under the Resource Conservation and Recovery Act (RCRA) extraction procedure.
4. The most environmentally significant product gas constituents are CO, benzene, H_2S, and carbon sulfide. The product gas represents a "well-known" hazard in the chemical industry, namely that the product (CO) that one is trying to maximize contributes the greatest hazards. The environmentally significant aqueous condensate constituents include phenol/cresols/xylenols (PCX), ammonia, sulfides, thiocyanates, cyanide, arsenic, and chlorides. The environmentally significant tar constituents include various PAHs (especially dibenzo(a,h)-anthracene, benzo(a)pyrene, and benzo(a)anthracene), PCX, and arsenic. The environmentally significant ash elements include arsenic, nickel, beryllium, and selenium.
5. The RTI laboratory gasifier produced pollutant and total stream data which compared very favorably to those reported in the literature for larger-scale gasifiers. Thus, a laboratory gasifier can simulate large-scale gasifiers and serve as a model for studying problems associated with large-scale gasifiers in a cost-effective manner.
6. Coal liquefaction processes produce liquid and solid effluent streams which are qualitatively similar to gasification aqueous condensate, tar, and ash. Combustion of coal produces, in general, far lower quantities of PAHs compared to gasification of coal on a unit coal basis. Coke plant wastewater is qualitatively similar to coal gasification aqueous condensate: pollutant concentrations in the two are within an order of magnitude.

Table 3. *Comparisons of Concentrations[$\mu g/L$]* of Significant Aqueous Condensate Constituents with Concentrations in Effluents from Other Coal Conversion Processes, Ambient Levels, and Regulated Levels

	RTI 95% C.I.	*Coke Plant Waste Ammonia Liquor Range*	*Coal Ash Pond Effluent*	*Coke Plant Liquor*	*SRC-I Wastewater*	*H-Coal Foul Process Water*	*SRC Wastewater*
Cresols	*4.8E5-8.8E5*						*9.4E5*
Xylenols	*1.4E5-3.2E5*						*3.8E5*
Ammonia	*4.8E6-7.8E6*	*1.8E6-4.3E6*	*5.0E1*	*5.0E6*	*5.6E6*		
Sulfides	*0-4.7E5*	*0-5.0E4*		*1.3E6*	*4.0E6*		
Arsenic	*1.6E2-1.2E3*		*2.0E1*				
Thiocyanates	*1.5E5-3.1E5*	*1.0E5-1.5E6*		*1.0E6*			
Phenol	*7.9E5-1.6E6*	*4.1E5-2.4E6*	*<2.5E1*	*1.6E6*	*4.5E6*		*3.9E5*
Cadmium	*0-2.1E2*		*<1.0E2*			*8.0E2*	
Selenium	*2.5E2-1.0E3*						*8.0E2*
Chromium	*3.6E2-3.0E3*					*1.0E2*	*9.0E4*
Cyanide	*1.5E3-7.3E3*	*1.0E4-3.7E4*	*<1.0E1*	*5.0E4*		*1.0E4*	
Iron	*4.7E3-5.1E3*		*5.0E2*			*1.2E3*	*5.6E5*
Chlorides	*9.4E5-2.7E6*			*6.0E6*			
Silver	*0-1.5E2*						
Lead	*1.1E1-2.9E2*		*<5.0E1*			*2.9E3*	

	RTI 95% C.I.	*Wabash River Indiana*	*Fresh Water*	*Rivers & Lakes Mean of 8 Regions*	*Most Stringent Water Quality Standards (U.S.)*	*Most Stringent Effluent Limitations (U.S. & Canada)*	*EPA NIPDWS*^{0}	*Illinois Effluent Standards (all point sources)*
Cresols	*4.8E5-8.8E5*							
Xylenols	*1.4E5-3.2E5*							
Ammonia	*4.8E6-7.8E6*	*8.0E1*			*2.0E1*	*5.0E2*		
Sulfides	*0-4.7E5*					*1.1E1*		
Arsenic	*1.6E2-1.2E3*	*2.0E0*	*4.0E-4*	*8.1E1*	*1.0E1*	*5.0E1*	*5.0E1*	*2.5E2*
Thiocyanates	*1.5E5-3.1E5*							
Phenol	*7.9E5-1.6E6*	*1.0E0+*			*1.0E0+*	*5.0E0+*		*3.0E2*
Cadmium	*0-2.1E2*	*<1.0E0*		*1.2E1*	*2.0E0*	*5.0E0*	*1.0E1*	*1.5E2*
Selenium	*2.5E2-1.0E3*	*1.0E0*	*<2.0E-2*		*5.0E0*	*1.0E1*	*1.0E1*	
Chromium	*3.6E2-3.0E3*	*<1.0E1*	*1.8E-4*		*5.0E1*	*5.0E1**	*5.0E1**	*3.0E2**
Cyanide	*1.5E3-7.3E3*	*0.0E0*			*5.0E0*	*2.0E1*		*2.5E1*
Iron	*4.7E3-5.1E3*	*1.8E3*	*6.7E-1*	*4.3E1*	*3.0E2*	*3.0E2*		*2.0E3*
Chlorides	*9.4E5-2.7E6*	*2.3E4*	*7.8E0*		*1.0E5*	*2.5E5*		
Silver	*0-1.5E2*	*0.0E0*	*1.3E-4*	*2.2E0*	*1.0E-1*	*5.0E1*	*5.0E1*	*1.0E2*
Lead	*1.1E1-2.9E2*	*4.0E1*	*2.2E1*	*2.2E1*	*1.0E2*	*5.0E1*	*5.0E1*	*1.0E2*

**Hexavalent Cr*
+Total Phenols
^{0}*National Interim Primary Drinking Water Standards*

7. The 95 percent confidence intervals for concentration and production of significant pollutants under a variety of gasification conditions (including coal type, temperature, pressure, and coal particle size) are well within an order of magnitude except for sulfides and some trace elements. A major factor probably is the coal type for sulfur compound production since as much as an order of magnitude or greater variation can occur in the amount of sulfur present in the coal itself.

S. K. Gangwal, J. G. Cleland, and R. S. Truesdale are with Research Triangle Institute, P.O. Box 12194, Research Triangle Park, NC 27709.
***N. Dean Smith** is the EPA Project Officer (see below).*

The complete report, entitled "Evaluation of Relative Environmental Hazards from a Coal Gasifier," (Order No. PB 81-217 648; Cost: $11.00, subject to change) will be available only from:

National Technical Information Service
5285 Port Royal Road
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The EPA Project Officer can be contacted at:

Industrial Environmental Research Laboratory
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

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