Phase 1 Report

Project No. DE-FC26-05NT42304

Health Effects of Subchronic Inhalation of Simulated Downwind Coal Combustion Emissions

Phase 1: Development of the Exposure Atmosphere

Joe L. Mauderly, DVM, Project Director
Jacob D. McDonald, PhD, Manager, Exposure and Analytical Chemistry

Lovelace Biomedical and Environmental Research Institute
Albuquerque, NM

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Project Director: ______________________ Date 6/16/06

Joe L. Mauderly, DVM
Director, National Environmental Respiratory Center
President, Lovelace Biomedical and Environmental Research Institute
Lovelace Respiratory Research Institute
2425 Ridgecrest Dr. SE
Albuquerque, NM 87111
505-348-9432
jmauderl@lrri.org

Contents

1 Executive Summary

This Report describes completion of the work under Phase 1 of project DE-FC26-05NT42304 "Health Effects of Subchronic Inhalation of Simulated Downwind Coal Combustion Emissions." This project is being performed as an integral part of a joint government-industry program termed the "National Environmental Respiratory Center" (NERC), which is aimed at disentangling the roles of different physical-chemical air pollutants and their sources in the health effects associated statistically with air pollution. The characterization of the exposure atmosphere and the health assays will be identical to those employed in the NERC protocols used to evaluate other pollution source emissions.

The project has two phases, each encompassing multiple tasks. General guidelines for the generation and composition of the exposure atmosphere were set by consensus from an expert workshop. The capability to generate the exposure atmosphere, and pilot studies of the comparative exposure composition using two coal types, was accomplished in Phase 1. In Phase 2, the toxicological study will be conducted using one of the coal types tested in Phase 1. This project provided 50% support for the work in Phase 1. This report summarizes the approach and results of Phase 1 and provides a recommendation for the atmosphere to be used for the animal exposures.

A system was constructed for generating a complex mixture simulating the key components of "downwind" population exposures to emissions from coal-fired power plants. Duplicate combustion systems were developed to provide redundancy for the eight months of daily exposures, and systems were added to modify the emissions and provide additional components to meet the target composition. The composite system was assembled, modified, and tested to achieve the required operating capability. Combustion trials were conducted using two coal types, Powder River Basin low-sulfur sub-bituminous coal (PRB) and Central Appalachian low-sulfur bituminous coal (CALS). Representative coals were obtained and processed. The final design involves feeding pulverized coal to electric "drop tube" furnaces using a screw feeder. A system for adding sulfate involves aerosolizing sulfuric acid, followed by heat vaporization and re-condensation. Gases are metered into the mixture from compressed gas tanks. The components are mixed and reacted in a mixing chamber, from which the atmosphere is routed to a dilution-distribution-exposure system. The latter was constructed and analyses of the pilot exposure atmospheres were conducted in the actual exposure system, although construction of the system was actually scheduled for Phase 2.

In its final configuration, the generation-exposure system has proven robust and suitable for daily operation over prolonged periods. Generation trials demonstrated that the target exposure composition could be achieved using either coal type. However (as observed by others), the PRB coal proved less difficult to generate consistently, largely because it is less "sticky" and thus easier to feed at a constant rate. The only difference between the final exposure atmospheres using the two coals would be in the composition of the ash component. Because coal ash will constitute only approximately 1% of the total particulate matter in the exposure, it is judged unlikely that small differences in ash composition would influence biological responses. Because PRB coal provides operating advantages and the difference between the ashes is not likely to influence biological outcomes, we recommend proceeding with Phase 2 using PRB coal. This approach was also recommended by NERC advisors.

2 Strategy

Information on past laboratory studies of the health impacts of coal emissions, and the need for a new study, were reviewed in detail in the April 30, 2004, competitive application, "Health Effects of Subchronic Inhalation of Simulated Downwind Coal Combustion Emissions." That application proposed the conduct of a toxicological study of the key components of coal-fired power plant emissions to which populations are exposed at some distance downwind. Further, it proposed that the study would be conducted as an integral part of the National Environmental Respiratory Center (NERC) program, which is aimed broadly at disentangling the components of complex air pollution mixtures that cause the respiratory and cardiovascular health effects associated statistically with ambient air pollution. The animal study protocol was to be consistent with protocols used for other NERC studies of source emissions. This strategy would: (1) provide a comprehensive, contemporary assessment of the health hazards associated with coal emissions; (2) prove a direct comparison with hazards of other source emissions; and (3) together with similarly-developed data from other source emissions, provide a database that could be analyzed with multivariate statistical strategies to determine which physical-chemical components of air pollution mixtures co-vary most closely with the different health effects.

Coal emissions were recommended by the NERC External Scientific Advisory Committee (ESAC) as one of the key source emissions for study by the program. Accordingly, we began gathering information on approaches to conducting such a study. The opportunity to respond to the Department of Energy/National Energy Transfer Laboratory (DOE/NETL) competitive solicitation for coal emission health research came when the NERC pre-planning process had advanced to the point that a solid strategy could be proposed. The work described herein was accomplished according with that strategy, with approval from both the NERC ESAC and DOE/NETL. The DOE/NETL grant involves two phases. Phase 1, the completion of which is reported here, encompassed the development, testing, and refinement of the system for generating the animal exposure atmosphere, and determination of operating characteristics using two coal types. Phase 2, to be initiated upon approval by DOE/NETL (and already approved by the NERC ESAC), encompasses the animal study and analysis and publication of all results.

Incremental progress under Phase 1 has been described in quarterly progress reports. Some, but not all, of the contents of those reports are reproduced in this report. The purpose of this report is to summarize our experience during Phase 1, present results of our preliminary generation trials, and propose a strategy for progressing on to Phase 2.

a. Recommendation from Expert Workshop

The general approach taken in this project followed the recommendations resulting from a workshop convened on February 27–28, 2003, to elicit current views of academic, federal, industry, and other technical experts concerning the most appropriate exposure atmosphere to simulate for toxicological studies and the most appropriate methods for generating the atmosphere. The workshop was attended by the individuals listed in Table 1. The participants represented a range of technical expertise and experience encompassing the nature of human exposures to coal emissions and their atmospheric transformation products, and the operation and comparative emissions characteristics of full-scale and laboratory-scale coal combustors.

Table 1. Workshop Participants

Bill Aljoe	DOE/National Energy Technology Laboratory
Ed Barr	LRRI, NERC Exposure Manager
Steve Benson	University of North Dakota
Lung-Chi Chen	New York University
Paul Chu	Electric Power Research Institute
Tom Grahame	DOE/Fossil Energy
John Jansen	Southern Company
JoAnn Lighty	University of Utah
Bill Linak	EPA/ORD (Manager of planned EPA coal exposures)
Joe Mauderly	LRRI, NERC Director (workshop moderator)
Jake McDonald	LRRI, NERC Atmospheric Chemist
Jim Meagher	NOAA
Bruce Miller	Pennsylvania State University
Larry Monroe	Southern Company
Niki Nicholas	Tennessee Valley Authority
Matt Reed	LRRI, NERC Study Director
Annette Rohr	Electric Power Research Institute
Roger Tanner	Tennessee Valley Authority
John Watson	Desert Research Institute
Richard White	LRRI, NERC Exposure Supervisor

Several experimental goals provided a framework for discussion, including:

- Simulate the most realistic human exposure scenario, realizing that a single atmosphere cannot possibly encompass the full range of emission mixtures and reaction products;
- Simulate contemporary exposures, but not from the most advanced technology;
- Exposures must cause measurable effects at the highest concentration, and;
- Exposures must fit within the requirements of animal management, and the experimental protocol.

A set of consensus recommendations resulted from the workshop. In addition to the general target compositional parameters listed in Table 2, it was recommended that:

- The most practical approach to laboratory-scale combustion is the “drop tube” electric furnace.
- Although ash will be a very minor component of the total exposure, different coal types will yield different ash composition. The two most representative coal types for the current steam market are Powder River Basin low-sulfur sub-bituminous (PBR) and Central Appalachian low-sulfur bituminous (CALS).

- Experience at the University of North Dakota Energy and Environment Research Center (UND/EERC) has shown that PRB coal will be the easier to aerosolize and feed to a drop-tube furnace.

Table 2. Consensus Recommendations for Atmosphere Composition

Sulfate particulate matter (PM) to ash PM ratio ≈ 100:1;

Carbon content of ash ≈ 5–10% (10% maximum);

SO_2:SO_4 molar ratio ≈ 1:1;

Total sulfur:NO_y molar ratio ≈ 4:1 in emissions, we want ≈ 2:1 in exposure;

Among nitrogen species ("NO_y"), we want ≈ 20% nitric oxide (NO), 55% nitrogen dioxide (NO_2), 15% nitric acid (HNO_3), and 10% peroxyacetyl nitrate (PAN) (later recommended against PAN);

Don't bother with O_3 or other secondary reaction products in this study (including PAN);

PM size cut of 2.5–3.0 μm is OK, at least cut to a size respirable by rodents; and

Measure, but don't attempt to manipulate, Hg (participants acknowledged that Hg is a key current issue, but recommended against using it as a target variable for a respiratory/cardiovascular study)

b. Work Plan and Schedule

According to the plan proposed in the application and approved by the NERC ESAC, the exposure development Phase included: (1) procurement of system components, including two drop-tube furnaces for redundant operation; (2) procurement of processed PRB and CALS coals in amounts permitting both pilot studies and conduct of the subchronic animal exposure; (3) iterative assembly, testing, and refinement of the generation system; and (4) development of sufficient operating and compositional information to support key "go-no go" decisions. The latter decisions included: (1) practicality of conducting daily exposures over a period of several months; and (2) which coal type to use for the exposure.

The schedule followed for this work was that proposed in the application. The system assembly and refinement and coal procurement efforts began in the winter of 2004, and was to be completed by the end of August 2005. Combustion trials with PRB coal were to begin at that time and be completed by the end of December 2005. Combustion trials with CALS coal were to begin at that time and be completed by the end of April 2006. This report was scheduled to be submitted by the end of May 2006.

Our goal for completing the developmental Phase of the project (Phase 1) was to conduct sufficient generation trials to determine whether or not either coal could be used, without expending the additional effort to make the final fine-tuning adjustments to meet the exposure targets precisely with both coals. It did not make sense to expend that final effort until the coal type was selected. We have accomplished that goal.

Overall, the work has progressed on nearly the original schedule. Certain aspects of the system required more time than anticipated to develop, and a major problem with the robustness of the coal feed system required a re-design. Regardless, the submission of this report is less than a month later than the original target date, which is not bad for an approximately 18-month effort to develop an entirely novel system. Moreover, we took advantage of a personnel-related opportunity to construct the full animal exposure system, which had originally been scheduled for Phase 2. Not only was that work completed early, but it also allowed us to make measurements of the exposure atmospheres in the actual system to be used for exposures, rather than in the upstream mixing chamber.

3 Work Accomplished and Results

a. Procurement, Processing, and Storage of Coal

The CALS coal was obtained from the Jones Fork blending/processing plant in Knott County, Kentucky. This plant acquires coal from multiple underground mines in the Hazard District of Eastern Kentucky, mining primarily the Hazard 4 and Elkhorn 3 seams. The coal beds and seams are geologically and stratigraphically consistent with low-sulfur bituminous coals from the Kentucky-West Virgina area. The selection of this source was reviewed and ratified by Bill Aljoe of DOE/NETL and Larry Monroe of Southern Company. The identification and procurement of this coal was facilitated by Steve Winter of CONSOL Energy, to whom we are indebted.

The coals were shipped to UND/EERC and processed under the supervision of Jason Laumb using methods they had previously used for coals for their own research and the coal used at EPA/RTP. At UND/EERC, the coals were pulverized, mixed, sampled for analysis, packaged under nitrogen, and shipped to Lovelace Respiratory Research Institute (LRRI).

For long-term storage at LRRI, the coals are contained in closed containers under nitrogen to purge oxygen that might oxidize the coal. We developed an enclosure that provides for storage of the coals in an inert atmosphere. The coal containers have a constant stream of high purity nitrogen flowing (at 1 liter per minute) through the headspace at all times. The nitrogen is generated from a PEAK nitrogen generator that produces 99% pure nitrogen.

b. Characterization of Coal

The PRB and CALS coals were characterized by standard proximate and ultimate analyses at Geochemical Testing Inc. based in Somerset, PA. This analytical strategy and source were recommended by UND/EERC, and provide a standard ASTM method-based characterization comparable to that used throughout the industry.

The "proximate" analysis gives moisture content, volatile content (when heated to 950^{o}C), the free carbon remaining at that point, the ash (mineral) in the sample, and the high heating value based on the complete combustion of the sample to carbon dioxide and liquid water. The "ultimate" analysis gives the weight percentage of hydrogen, carbon, nitrogen, sulfur and oxygen, and ash.

The results of proximate analysis for PRB and CALS coal are listed in Tables 3 and 4, respectively. The ASTM method numbers are indicated following the descriptors. The composition of both coal samples fell well within the range of values typical of the types as provided to the steam market. The sulfur content of the "as received" PRB and CALS coals were 0.49% and 1.39%, respectively, consistent with the anticipated values, and reflecting the relative desirability of PRB coal to meet sulfur emission standards. The ash content was slightly higher for the CALS coal than for PRB, as was the volatile matter. The heat content was higher for the CALS coal as well, reflecting its relative desirability for steam generation.

Table 3. Proximate-Ultimate Analysis of PRB Coal

	Weight% As Received	Dry	Dry Ash-Free
Proximate Analysis D3172			
Moisture	16.91	–	–
Ash	6.56	7.9	–
Volatile Matter	36.69	44.16	47.94
Fixed Carbon	39.84	47.94	52.06
Total	*100*	*100*	*100*
Ultimate Analysis			
Hydrogen D5373	6.32	5.33	5.79
Carbon D5373	57.51	69.21	75.15
Nitrogen D5373	0.71	0.85	0.93
Sulfur D4239-02	0.42	0.51	0.55
Oxygen D3176	28.48	16.2	17.58
Ash D3174-02	6.56	7.9	–
Total	*100*	*100*	*100*
Heating Value (Btu/lb; D5865)	10000	12035	13067
Free Swelling Index D720-91	0.0		

Table 4. Proximate-Ultimate Analysis of CALS Coal

	Weight% As Received	Weight% Dry	Weight% Dry Ash-Free
<u>*Proximate Analysis D3172*</u>			
Moisture	1.53	–	–
Ash	7.71	7.83	–
Volatile Matter	38.29	38.88	42.19
Fixed Carbon	52.47	53.29	57.81
Total	*100*	*100*	*100*
<u>*Ultimate Analysis*</u>			
Hydrogen D5373	5.06	4.96	5.39
Carbon D5373	76.08	77.26	83.83
Nitrogen D5373	1.51	1.53	1.66
Sulfur D4239-02	1.39	1.41	1.53
Oxygen D3176	8.25	7.01	7.59
Ash D3174-02	7.71	7.83	–
Total	*100*	*100*	*100*
Heating Value (Btu/lb; D5865)	13550	13761	14930

c. Assembly and Final Configuration of the Generation System

To set the specifications for the electric furnaces, discussions were first held with Dr. L.C. Chen and other investigators at New York University, who reported the only experience with exposing animals using a drop-tube furnace. Drs. Mauderly and McDonald also visited UND/EERC and EPA/RTP to gain a first-hand view of their electric furnaces and to discuss operating experience. Advice from UND/EERC was especially helpful in setting specifications for LRRI's unique application, identifying potential vendors, and eliciting a operating advice and precautions.

After learning the best features of existing drop-tube furnaces and coal feeding devices, and defining key operating issues and problems to avoid, we developed specifications, contacted suppliers, and determined the best vendor on the basis of technical competence to provide a custom-designed unit, cost, and delivery schedule. We purchased two identical split-tube furnaces and associated control systems from Thermcraft, Inc. (Winston-Salem, NC).

The furnaces have 4-inch internal diameters to accommodate 3-inch ceramic tubes in which the coal will be combusted. The 12780-watt furnaces have silicon carbide heating elements and an operating temperature range of 500 – 1600^{o}C. The furnaces are insulated with light-weight ceramic fiber and have steel outer shells that are arranged in two halves that can be opened to service the heating elements and combustion tube. The heating elements are controlled by Yokogawa UP350 digital controllers which sense temperature with ceramic-sheathed thermocouples and provide current to the furnaces from a 240-volt, 3-phase, 60 Hz power supply. The design of the furnaces is illustrated in Figure 1.

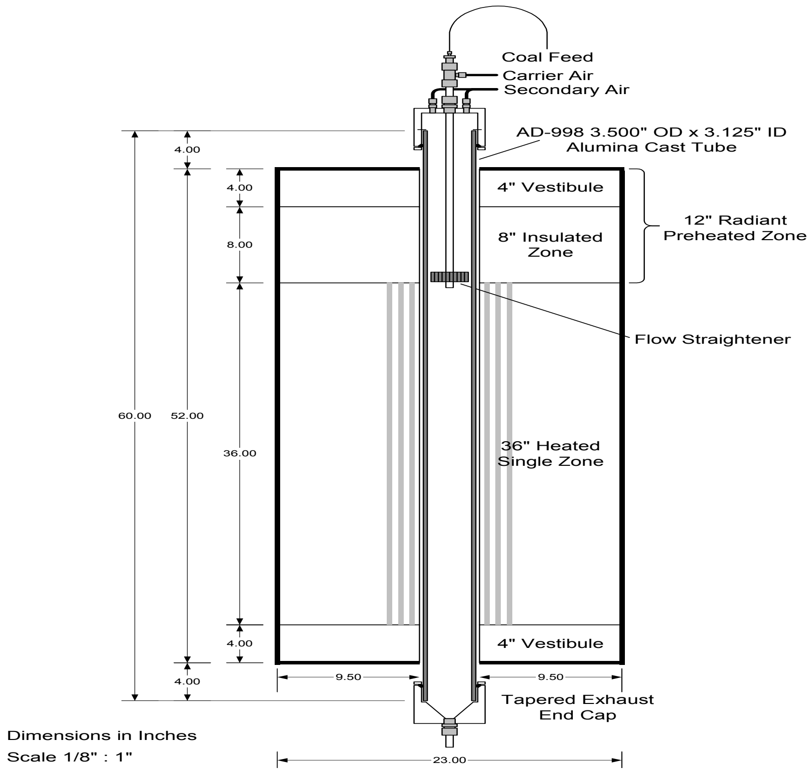

Figure 1. Cross-Section of LRRI Drop-Tube Electric Furnace

The furnaces were mounted on stands providing space above for coal and air-feed devices and below for emissions collection and conditioning (Figure 2).

Figure 2. Furnace being Mounted on Stand (See 6-ft. ladder for scale.)

The rest of the system was constructed after the furnaces were in place. The components were developed, tested, and refined iteratively, with operating parameters evaluated at each step. The general design of the final system is shown in Figure 3.

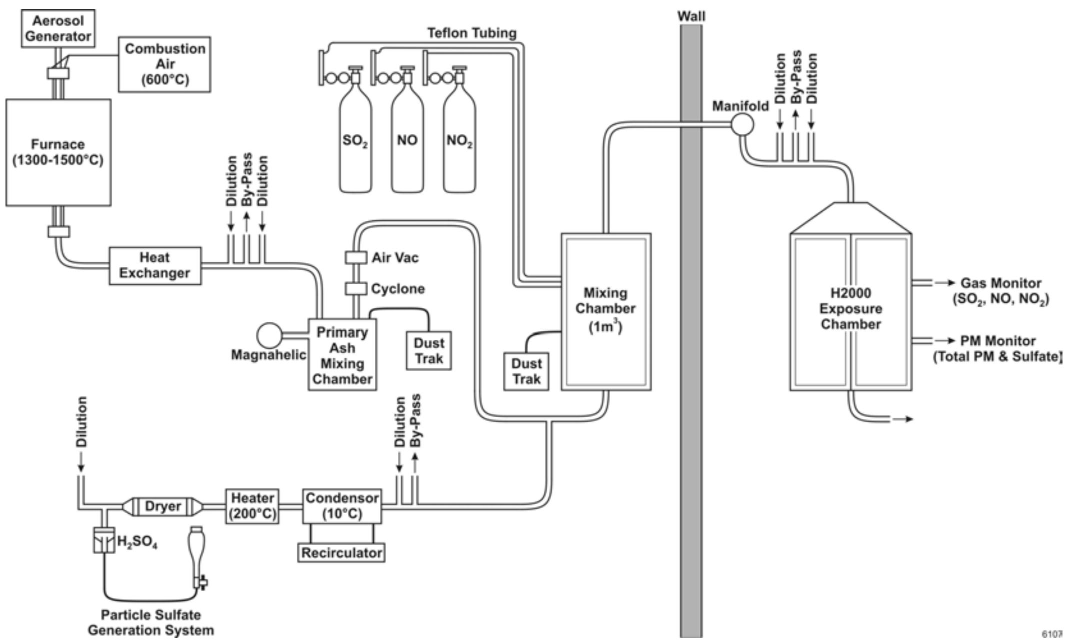

Figure 3. Schematic Diagram of Components of the Generation-Exposure System

Aerosolized coal and heated combustion air are fed into the top of the furnace. The rate of production of emissions and the burn-out of carbon are controlled by the coal feed rate, furnace temperature, and air flow rate (residence time). The combustion air is heated to 600°C by a small electric furnace (Figure 4).

Figure 4. Electric Furnace for Pre-Heating Combustion Air

The aerosol and combustion air flow rates were approximately 1 and 15 liters per minute. At these flows, the residence time in the furnace was approximately 2 – 3 seconds. The furnace combustion zone was heated to 1400°C for both the PRB and CALS trials. Furnace emissions are cooled using a heat exchanger, and the cooled emissions are diluted and routed to a primary ash mixing (or buffer) chamber. Material is drawn from the 30-liter stainless steel buffer chamber and through a cyclone to ensure that only fine, respirable particles are included in the exposure. An air pressure-driven "Air Vac" extractor is used to move the aerosol through the cyclone and into the larger mixing chamber. The modified ash aerosol stream then joins a sulfate aerosol stream before entering the larger (1.0 m^3) mixing chamber.

Adding sulfate is necessary to achieve the target sulfate content of the exposure atmosphere. Sulfate is added by merging a sulfate aerosol with the ash aerosol stream, as diagrammed above. An aerosol of sulfuric acid is generated from solution using a Hospitak™ nebulizer, which produces a relatively large (1 – 2 micron) particle size. The aerosol is dried through a diffusion dryer and then heated at ~200°C to vaporize the sulfuric acid. Vapors are then condensed through a heat exchanger (10°C) to create an aerosol with a targeted bimodal particle size at ~ 100 and 1000 nm. This particle size matches the ambient air bimodal size distribution of sulfate observed in $PM_{2.5}$. Figure 5 shows the sulfate generation system. In this photo, the furnace is at the upper right, and the emissions cooling system is shown under the furnace. The control panel at the left monitors pressures and flows throughout the system (including ash delivery). The large stainless steel mixing chamber (actually an animal exposure chamber) is in the center background.

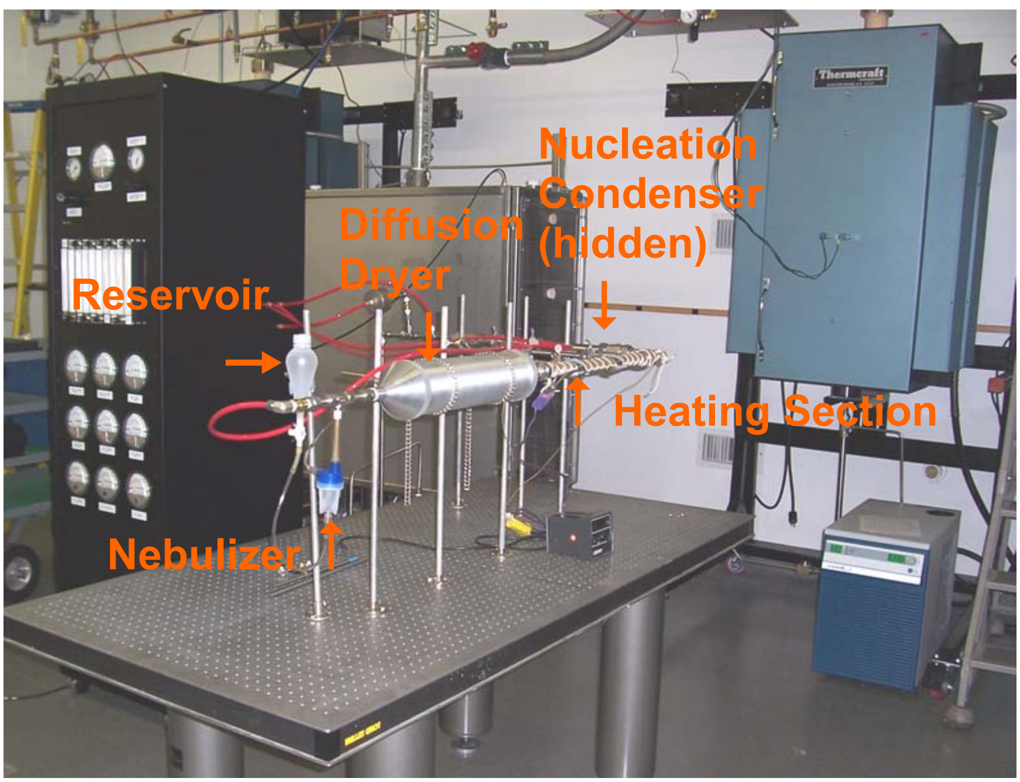

Figure 5. Sulfate Generation System

It is also necessary to add SO_2, NO_2, and NO to the mixture to achieve the desired composition. These gases are purchased in compressed gas cylinders, and metered into the mixing chamber.

The first device used for aerosolizing pulverized coal and feeding the aerosol into the top of the furnace was based on the design used successfully by EPA/RTP for short-term animal exposures (Figure 6). Coal dust from the surface of bulk material placed in a small reservoir tube was entrained as an aerosol into air flowing into the reservoir tube and out through a small-diameter extraction tube placed very close above the surface of the dust. For operation, the reservoir tube was loaded, the extraction tube lowered to an appropriate height above the coal surface, airflow was initiated at an appropriate rate, and the extraction tube was maintained at a constant height above the coal dust by attachment to a syringe pump (worm gear device). Operating variables determining the feed rate are the airflow rate and the speed at which the syringe pump advances the extraction tube. This system was assembled, tested, and optimized, and used for the first generation trials.

Figure 6. Initial Coal Aerosolizing Device, Based on EPA Design

It was discovered that, although the coal aerosolization device worked, it required very close observation and frequent adjustment to obtain a constant feed rate. Such a personnel-intensive and sensitive system was not considered sufficiently robust for daily use over a period of many months, so alternative designs were considered. After alternatives were evaluated, a screw-feeder device was selected, tried, and proven effective. This device (Figure 7) uses a motor-driven auger to feed dust from a hopper into the air stream immediately above the furnace. It was found that PRB coal fed very well using the screw feeder, and that the feed rated was stable over prolonged periods with little operator interaction other than loading the hopper (Figure 8). The CALS coal can also be fed successfully using the screw feeder; however because of its “stickier” nature, it tends to feed in larger clumps of particles than PRB coal.

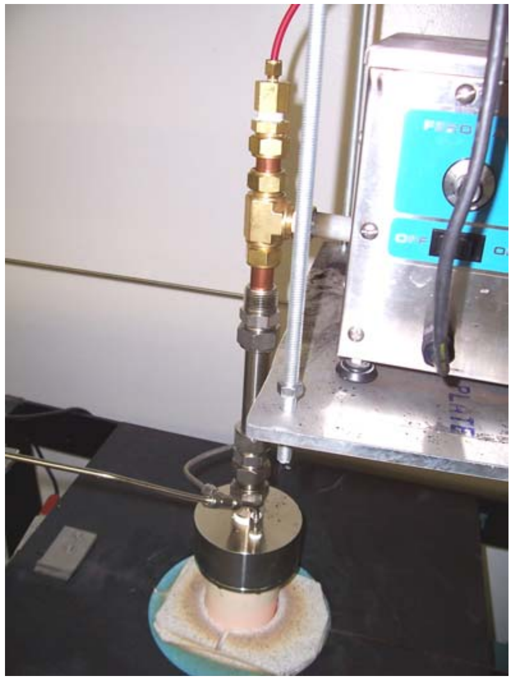

Figure 7. Junction between the Outlet of the Screw Feeder and the Air Stream Entering the Top of the Furnace. The line carrying the pre-heated combustion air enters from the left.

Figure 8. Technician Loading the Hopper of the Screw Feeder. The combustion air pre-heater is to the left of the top of the furnace.

The exposure atmosphere is routed from the mixing chamber into the next room housing the dilution-distribution-exposure system. The atmosphere is distributed (Figure 9) throughout a horizontal plenum (A), from which portions are extracted and diluted to the final concentration for each chamber (C). One chamber houses animals exposed to each of four dilutions. Providing material to the distribution plenum at a concentration exceeding that of the highest exposure level allows for close adjustment and control of each exposure concentration. A separate extraction point (B) serves each chamber, and the material is diluted to the appropriate concentration. The exposure chambers are shown in Figure 10.

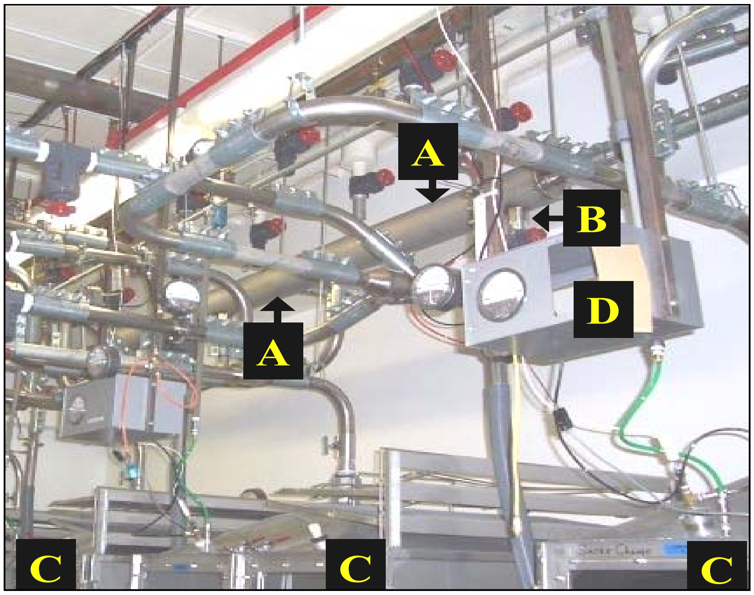

Figure 9. Distribution System in the Exposure Room

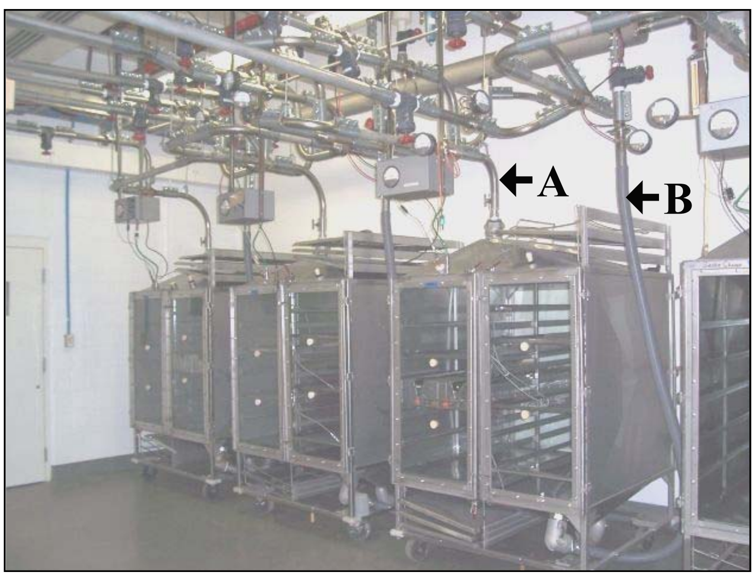

Figure 10. Animal Exposure Chambers, Showing Inlet (A) and Exhaust (B)

d. Iterative Generation Trials

Generation trials were initiated using PRB coal, because a small amount of processed PRB coal was provided by EPA/RTP so work could begin before we received the study batch. The results shown below, however, were obtained using the study coals. Coal emissions, sulfate, and gases were added as described above. The workshop recommended including some neutralized sulfate, but our experience indicates that production of ammonia by the animals (despite twice daily chamber cleaning) will provide for sufficient neutralization (~50% neutralization based on ammonia concentrations measured with similar animal numbers in previous studies) without adding ammonia. After approximating the exposure composition targets using PRB coal, we switched to CALS coal.

e. Composition of Atmospheres using PRB and CALS Coals

The data from each iterative generation trial are not included in this report. The key issue is our final ability to achieve the target exposure mixture, and the answer is represented by our most recent summary data with each coal. The data that follow are those that were presented at the May 2006 NERC annual meeting, and which served as the basis for the NERC ESAC recommendation to proceed.

(1) Particle Size

The particle size exiting the furnace was bi-modal, as expected. This was the reason that the cyclone was included — to remove the larger particles. The particle size distribution in the ash buffer chamber while burning PRB coal is shown in Figure 11. During this trial, the two size populations in the bi-modal distribution centered on 0.17 and 3.7 μm.

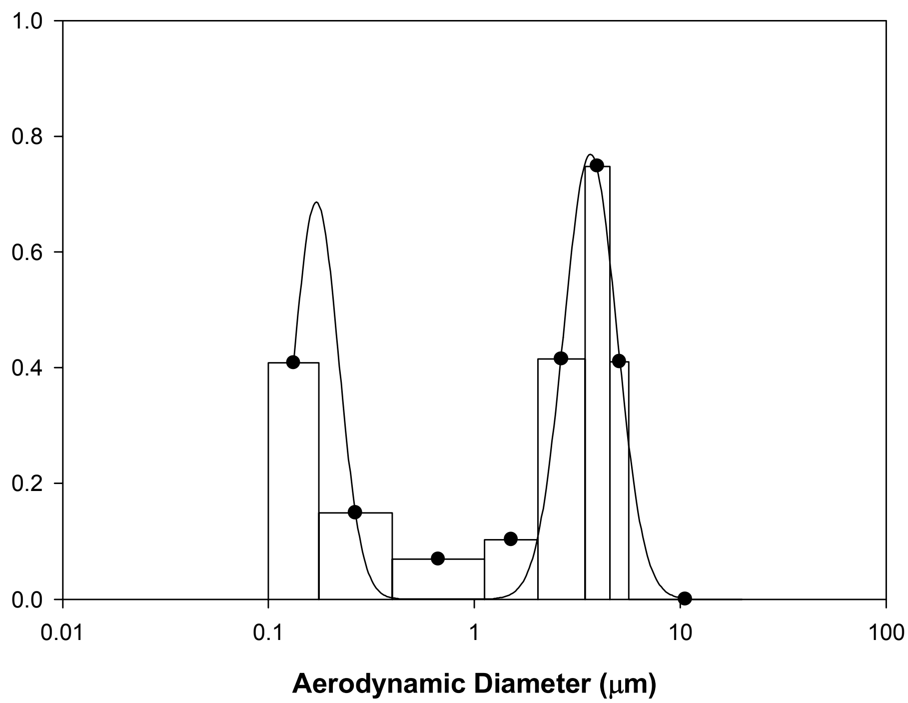

Figure 11. Particle Size Distribution in Ash Buffer Chamber during PRB Coal Combustion Trial

The cyclone removes the largest particles, yielding a size distribution within the range that can be deposited in lungs of rodents (upper bound of approximately 3 µm) and that better represents ash that may penetrate emissions control devices (e.g., electrostatic precipitators) in the environment. The adjusted size distribution is illustrated in Figures 12 and 13, which show results from the animal exposure chamber. Two different techniques that measure two separate size ranges were used to analyze the particle size distribution in the exposure chambers with the target atmosphere. An aerodynamic particle sizer (APS, 3321, TSI, St. Paul, MN) measures aerodynamic size from 0.5 – 20 microns by time of flight and an optical detector. Data taken using the APS sampler (Figure 12) shows the larger portion of the aerosol that centers on approximately 0.7 – 1.0 µm, with an upper bound of approximately 2.5 µm. The smaller size fraction was analyzed by differential mobility/condensation nuclei counter (SMPS, Model 3080) that measures particle size from ~0.012 – 0.5 microns. Data taken using the SMPS sampler (Figure 13) shows that the small fraction of mass in the lower size mode centers on approximately 70 nm, or 0.07 µm. These results demonstrate that the system has the targeted bimodal size distribution that mimics the size of sulfate (the primary particle component in the atmosphere) in the environment.

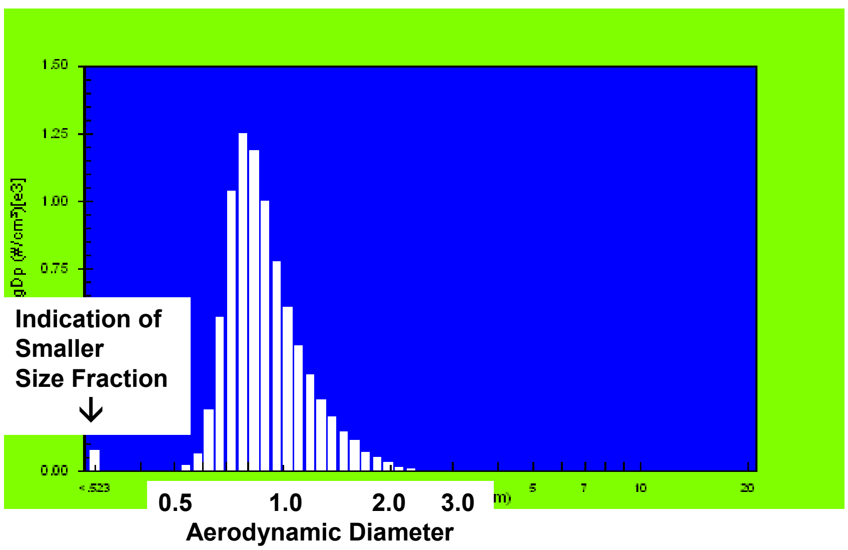

Figure 12. Particle Size Distribution in Exposure Chamber during PRB Combustion (size in µm)

(2) Particle Composition

The degree to which carbon is burned out of the ash particles is one parameter that reflects the adequacy of the generation system to mimic full-scale combustion. Power plants strive to burn out as much carbon as possible to maximize use of the potential heat value of the coal. Burnout, or combustion efficiency, is reflected by low carbon monoxide, a change in color of the particulate, and carbon content (determined by thermogravimetric analysis or thermal/optical reflectance). For color change of the particulate, the blackness of ash parallels residual carbon. We found it easier to achieve burn-out with PRB than with CALS coal, as predicted by UND/EERC and other technical experts. PRB coal ash is red-orange after satisfactory carbon burn-out, and Figure 14 illustrates that we achieved that color.

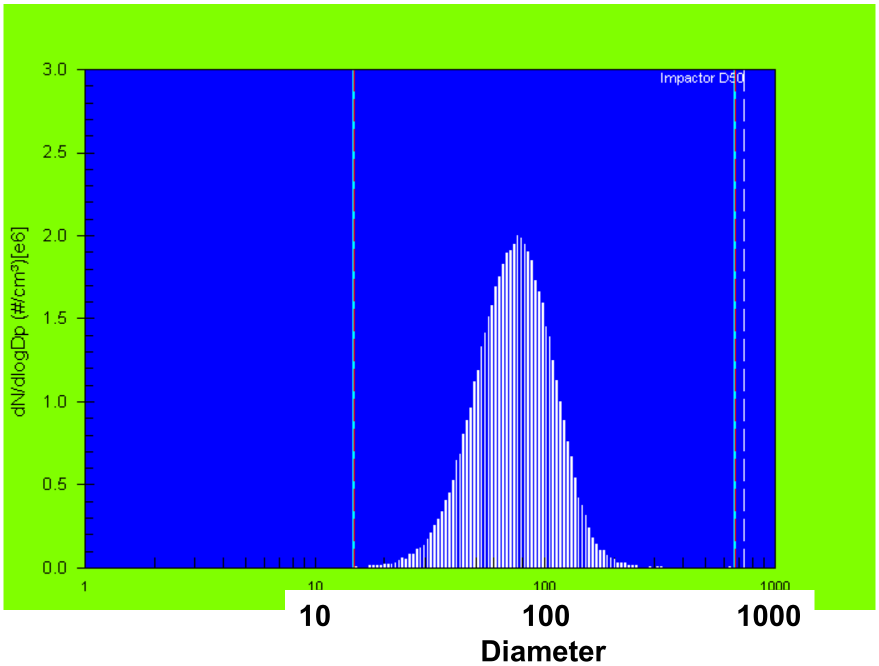

Figure 13. Size Distribution of Smaller Particle Population during PRB Combustion (size in nm)

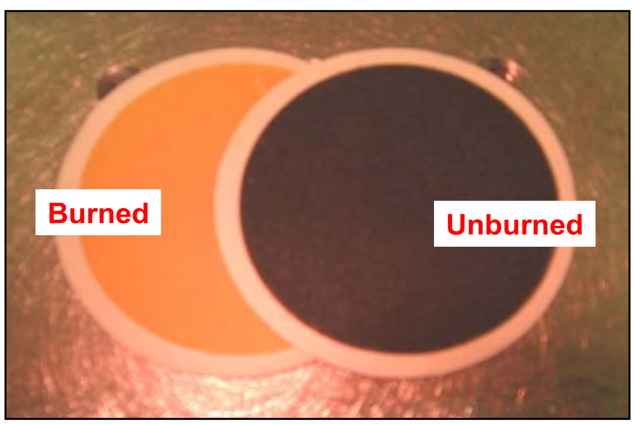

Figure 14. Color Change Reflecting Appropriate Carbon Burn-Out of PRB Coal. The filter on the left contains PRB ash, and the filter on the right contains unburned PRB coal.

The relative carbon burn-out of PRB and CALS coals is illustrated in Figure 15. The carbon and ash are a sum of measured elements (determined by x-ray fluorescence) and carbon (determined by thermal/optical reflectance) obtained from the primary mixing chamber. Without final fine-tuning of dilution-time-temperature profiles, we achieved approximately 80 – 85% burnout with PRB coal, but only approximately 75 – 80% burnout with CALS coal. Further tuning will increase burn-out, but we are not confident that we can achieve a burn-out with CALS coal equal to that of CALS coal.

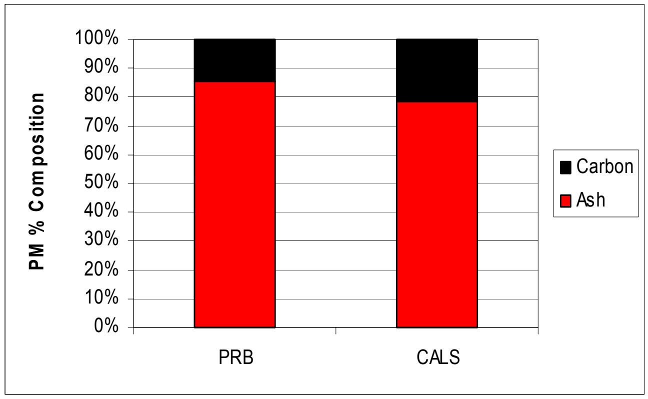

Figure 15. Relative Carbon Burn-Out with PRB and CALS Coals

The relative elemental compositions of the ashes from the two coals are illustrated in Figure 16. The most striking difference is the much greater iron content of the CALS ash, as expected.

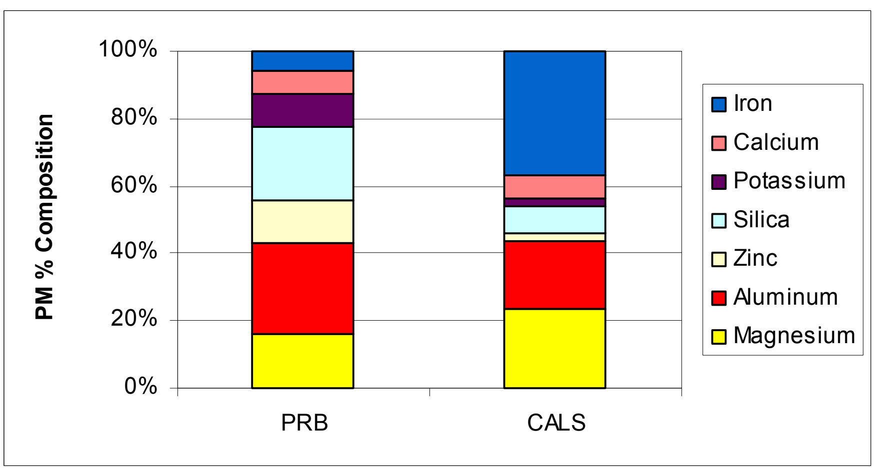

Figure 16. Elemental Content of PRB and CALS Coal Ashes

PM in the animal exposure atmosphere, however, will not be just ash. Indeed, ash will constitute a very small portion (approximately 1%). For example, in the high-level exposure chamber the ash content will be approximately 10 μg/m^3, and the total particulate mass (remainder is sulfate) will be 1000 μg/m^3. Thus differences in ash concentration result in only minor differences in the composition of the atmosphere (e.g., 4 x increase in iron would change concentration from 1 – 4 µg/m3. This is illustrated in Figure 17, which shows the composition of the total particulate mass in atmospheres generated from the two coals. From this perspective, it is clear that the PM will be primarily sulfate, and that differences attributable to the two coals would be almost negligible. This does not guarantee that the ash composition would have no impact on health outcomes, but it suggests that may very well not influence the outcomes.

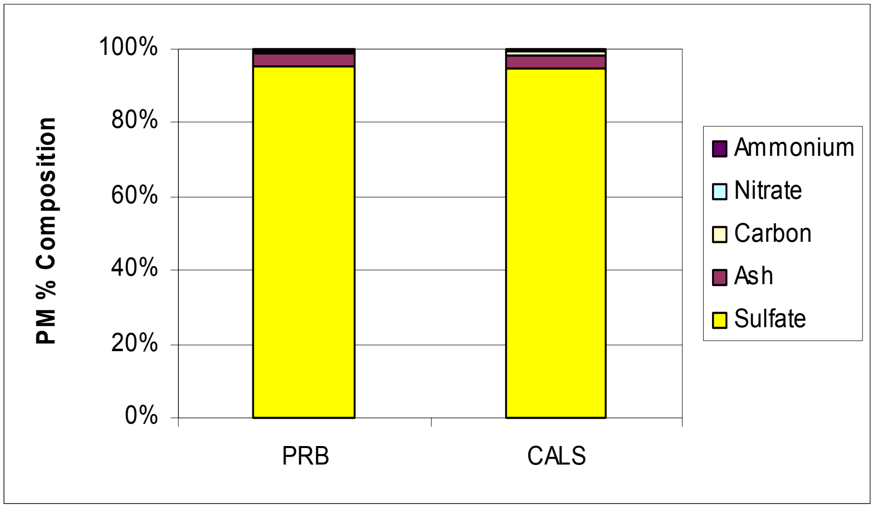

Figure 17. Relative Composition of Total PM in Exposure Atmospheres Generated Using PRB or CALS Coal

(3) Overall Approximation of Target Atmosphere

The extent to which the overall target exposure atmosphere composition was approximated in the generation trials to date is illustrated for PRB and CALS coals in Figures 18 and 19, respectively. The targets were met with very good accuracy, considering the demands of the complex natures of the target atmosphere and the generation system. These results demonstrate that the exposures could be conducted with either coal. We expect that final fine-tuning after selection of the study coal will bring the atmosphere even closer to the target values.

Figure 20 presents results for additional non-particle components that were not included as specific targets. Carbon monoxide was higher for CALS than for PRB coal at the same particle concentration, probably reflecting the lesser carbon burn-out. Total vapor-Phase hydrocarbon (THC) was essentially equal for the two coals, and in both cases the vapor hydrocarbon was near background.

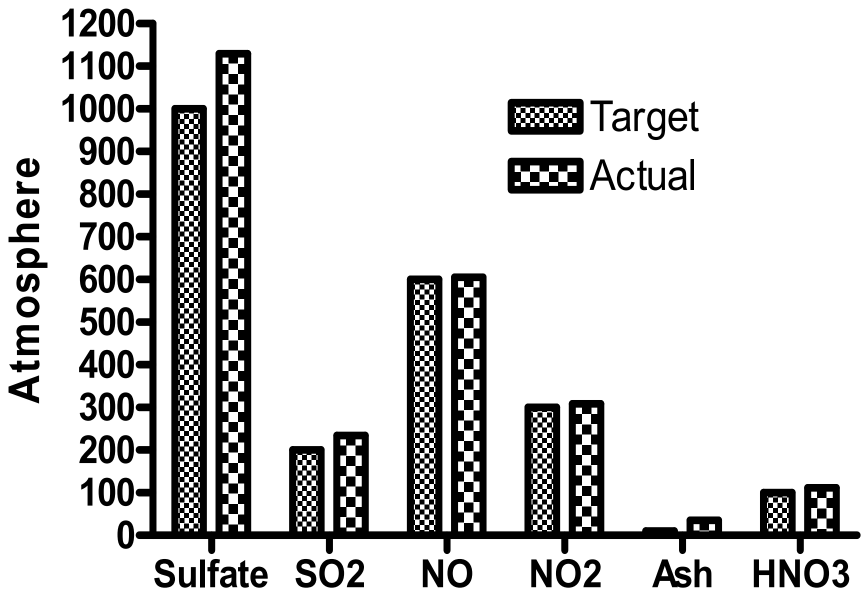

Figure 18. Approximation of Target Exposure Atmosphere using PRB Coal

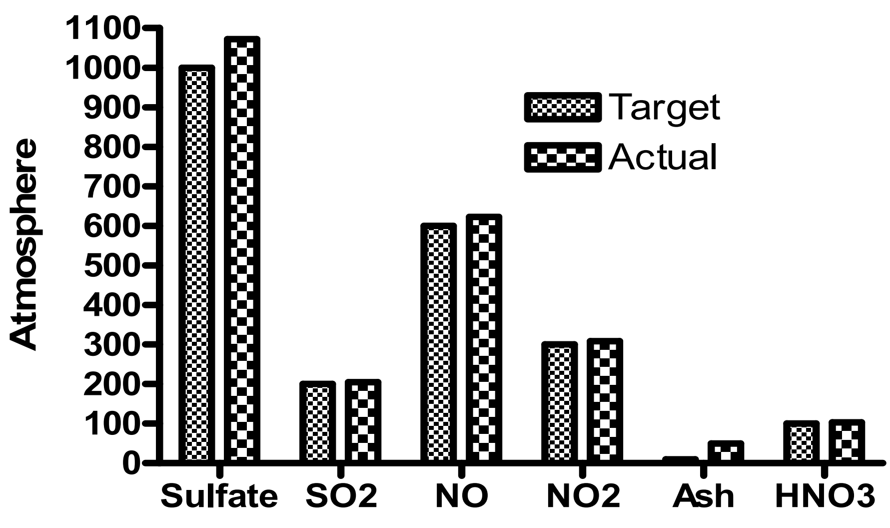

Figure 19. Approximation of Target Exposure Atmosphere using CALS Coal

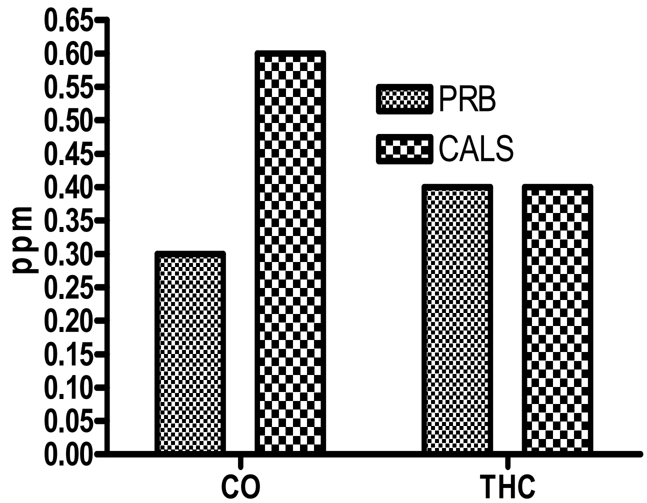

Figure 20. Comparative Carbon Monoxide and Total Hydrocarbon at the Same Particle Concentration for PRB and CALS Coals

The above results demonstrate that the feasibility of achieving an exposure atmosphere meeting the target composition. Moreover, we have developed sufficient operating experience that we are confident that a subchronic exposure can be carried out successfully. Pending a decision to move forward, we will conduct additional fine-tuning of the atmosphere using the selected coal. That can readily be accomplished while the animal protocols are being developed and other logistics of initiating the animal study are being managed.

4 Recommendations for Phase 2

Based on the above experience and results, and consistent with the recommendations of the NERC ESAC, we recommend progressing to Phase 2 at this time, using PRB coal for the study.

The ESAC's guidance from the May 2006 meeting included a recommendation to proceed with planning and initiation of the animal study using PRB coal. Of course, that recommendation does not preclude the need for DOE/NETL to make its own "go-no go" decision. We stand ready to participate in any discussions that are necessary to facilitate a decision.

The systems for generating the exposure atmospheres and conducting the exposures have been successfully developed and tested. We found that, although either coal could be used for the exposure, PRB coal provides a greater level of assurance that the coal feed rate will be consistent, provides a more complete burn-out of carbon, and requires less maintenance of the feed lines. The heavy dependence of Phase 2 of this project on the 80% co-funding from other sponsors under the auspices of the NERC program would necessitate additional interaction with the rest of the NERC constituency if DOE/NETL opted to proceed to Phase 2 using CALS coal.

There are no important technical obstacles at this time to proceeding with Phase 2. Pending DOE/NETL approval to proceed, we will focus on using the selected coal to make the final adjustments necessary to fine-tune the system to meet exposure composition targets as precisely as possible. To begin the animal study, protocols have to be developed and approved and the initiation of exposures must be coordinated with other animal procurement and management activities. If a “go” decision is made, the sooner it is made the sooner we can schedule and begin those activities.

5 Publications, Presentations, and Technology Transfer

The work described in this report has not been reported in public venues, either by publication in journals or presentation at scientific meetings. The strategy, progress, and results were described at the May 2005 and May 2006 annual meetings of the NERC External Scientific Advisory Committee, sponsors, and investigators. The proceedings of those meetings were not made available to the public. The DOE/NETL Project Officer attended both meetings, received all presentation materials, and participated in the discussions. Progress was also summarized briefly in the November 2005 NERC Annual Report, which was provided to the Project Officer. Although the Annual Report is not Distributed to the public, it is available on the NERC web site (www.nercenter.org).

No technology transfer activities have resulted from this project to date.