FEASIBILITY OF APPROACHES FOR GENERATING INHALATION EXPOSURES TO COAL COMBUSTION EMISSIONS FOR TOXICOLOGICAL STUDIES TO SIMULATE EXPOSURES OF POPULATIONS

A Report Developed by the

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for the

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I. Executive Summary

The feasibility of generating exposure atmospheres simulating key components of human exposures to emissions from coal-fired power plants was evaluated using a combination of literature review, discussions with organizations having relevant experience, a workshop of technical experts, and discussion with the external advisory committee of a major air pollution health research program. Although emissions from coal-fired power plants and their atmospheric reaction products contribute to environmental air pollution and are often cited as among the important sources of pollution-related health risks, there has been surprisingly little toxicological evaluation of the health hazards of breathing coal emissions or of the influences of coal type, operating variables, emission reduction strategies, and atmospheric reactions. Virtually no toxicological research has been done to place "downwind" (i.e., rather than "top of stack") emissions into context regarding population health risks.

The present review addressed the feasibility of generating meaningful, reproducible, repeated inhalation exposures of sufficient numbers of animals for a sufficient time to characterize health hazards, and the exposure variables modifying those hazards. Although no program involving inhalation exposures to fresh or reacted coal combustion emissions was underway at the time of this report, two programs anticipated such exposures. The U.S. Environmental Protection Agency (EPA) was dosing animals with particulate material collected from a laboratory-scale combustor, and anticipated conducting inhalation exposures in the future. The Electric Power Research Institute (EPRI) was preparing to conduct brief inhalation exposures of animals to fresh and reacted emissions taken directly from the stacks of operating power plants. The most recent study was done by New York University over 10 years ago using single inhalation exposures of animals to fresh emissions from a laboratory-scale "drop-tube" furnace.

It was concluded that useful exposures could be generated within the bounds of plausible technology and funding. The workshop produced a consensus of general specifications for the key components of a "downwind" exposure model, including:

- Sulfate particulate matter (PM) to ash PM ratio is $\approx$ 100:1.
- Carbon content of ash is $\approx$ 5–10%.
- Sulfur dioxide (SO_2):sulfate (SO_4) molar ratio is $\approx$ 1:1.
- Total sulfur to nitrogen species molar ratio is $\approx$ 2:1.
- Among nitrogen species, we want $\approx$ 20% nitric oxide (NO), 55% nitrogen dioxide (NO_2), 10% peroxyacetyl nitrate (PAN), and 15% nitric acid (HNO_3).
- It is not worth attempting to fully model ozone or other secondary reaction products resulting in part from coal emissions.
- The upper bound of particle size should be limited to the respirable range for the species to be exposed.

It was determined that a suitable exposure atmosphere might be developed beginning either by combusting coal or by resuspending ash collected from a power plant, although neither approach would provide the desired atmosphere without modification by adding components and/or aging and reacting the emissions. Although opinion is divided regarding the preferred approach, the author's preference and that of the External Scientific Advisory Committee of the

National Environmental Center was to begin by combusting coal. It was determined that the most feasible laboratory-scale combustion apparatus for this purpose was the “drop-tube furnace.

II. Background

A. Report Task

The National Energy Technology Laboratory contracted with the Lovelace Respiratory Research Institute (LRRI) to conduct a review and develop a report on the feasibility of, and the most appropriate methods for, conducting inhalation exposures of animals to coal combustion emissions for the purpose of toxicological studies of the potential health effects of human exposures to emissions from coal-fired power plants. This request was predicated on the continued uncertainty about the contribution of power plant emissions to the health effects associated with population exposures to air pollution and the paucity of toxicological data for effects of realistic exposure scenarios. The purpose of this review was to evaluate the feasibility of generating exposure atmospheres for toxicological studies that might improve the understanding of health hazards of coal emissions and their key atmospheric transformation products. The task was described as follows:

> *“—provide a written evaluation of approaches for the laboratory-scale generation of coal combustion emission atmospheres that optimize the simulation of human exposures to emissions and their key atmospheric transformation products. The report should be an inclusive, but concise, summary of the methods used to gather information (including a listing of the technical experts consulted), the plausible options that were identified, the recommended “best” option, the rationale for the recommended option, and (if possible) the extent of consensus among the experts consulted regarding the recommended option.”*

B. Approach

The first step involved conducting a review of the published literature on studies of the health effects of exposure to coal combustion emissions—studies of the effects of the three major components of human exposures to emissions: 1) sulfur dioxide and sulfur-containing environmental aerosols; 2) nitrogen oxides, nitric acid, and peroxyacetyl nitrate; and 3) coal fly ash and reports of the composition of coal emissions. Searches for publications during the last 30 years were performed using standard databases. Although it was not ascertained that the literature search encompassed all published technical reports as well as journal articles, the search was considered sufficient to encompass the range of methods used to generate exposure atmospheres for experimental studies involving either humans or animals. Although many of the citations reported the effects of exposure, this review did not address the heath effects *per se* (not included in the Statement of Work). The bibliographies produced by the review are presented in Appendices A, B, C, and F, and the scanty literature on exposures to “whole” coal combustion emissions is also included in the body of the report.

The second step involved convening a peer workshop of individuals having expertise in areas relevant to the evaluation, including the composition of actual population exposures, atmospheric chemistry of coal emissions and their reaction products, full-scale power plant combustion technology and emissions, laboratory-scale simulation of power plant emissions, and animal exposures to coal combustion emissions. The goal of the workshop was to elicit information useful for specifying the composition of an appropriate exposure atmosphere and to elicit recommendations and consensus, if possible, on the best method for generating laboratory exposures.

The third step involved obtaining specific information on the methods proposed by two organizations planning to conduct inhalation exposures of animals to coal emissions within current programs: the U.S. Environmental Protection Agency (EPA) and the Electric Power Research Institute (EPRI). The goal of this effort was to determine the proposed methods and their rationale.

The fourth step involved presenting the results of the review and workshop to the External Scientific Advisory Committee and sponsors of the National Environmental Respiratory Center (NERC) at its June 5–6, 2003, annual meeting. The NERC program is undertaking animal exposures to a range of common source emissions and intends to conduct exposures to coal emissions in the future. This forum was considered a useful "third-party" assessment because the advisory committee consists of a cross-section of highly qualified health and atmospheric experts, and the Center's sponsors include regulating and regulated entities (including the EPA and the electric power industry).

III. Literature Review of Methods for Generating Exposure Atmospheres

In addition to reviewing methods used for exposures to whole fresh or reacted emissions from coal combustion, it is of interest to review methods used in previous experimental exposures to the three classes of materials that are likely to be incorporated as major components of the desired exposure atmosphere: 1) fly ash; 2) sulfur dioxide and sulfur-containing particles; and 3) nitrogen oxides and nitric acid. Although a thorough literature search was conducted using available databases, it was not assured that all technical reports or more obscure publications were identified. Regardless, the substantial number of citations obtained was considered to adequately encompass the recent (30-year) historical range of study types and generation methods.

A. Experimental Exposures to Coal Combustion Emissions or Coal Fly Ash

1. *Exposures to Coal Combustion Emissions*

There have been astonishingly few reported studies of experimental exposures of cells or animals directly to whole, diluted, fresh or reacted emissions from the combustion of coal and no reports of experimental exposures of humans. There is a sizable literature on experimental exposures of cells and animals to fly ash from conventional pulverized coal or fluidized bed coal combustors, but no report of such exposures of humans. There are a few clinical reports of respiratory disorders resulting from extreme occupational exposures to fly ash, but those are not considered relevant to methods for experimental exposures.

The bibliography in Appendix A includes six citations related to exposures of animals to coal combustion emissions; however, most are not relevant to the subject at hand. Three citations (Rittinghausen et al., 1997; Heinrich et al., 1986; Mohr et al., 1986) resulted from a series of studies in which animals were exposed to flue gas from a device alternately described as a "coal oven" or "domestic coal furnace," which was used for its production of a high content of polycyclic aromatic hydrocarbons (PAHs). The concentrations of PAHs were further increased by the addition of effluent from pyrolysis of coal tar pitch and/or vapors of benzo(a)pyrene. Because the resulting exposures might more closely represent coke oven emissions than coal-fired power plant emissions, this series of studies is not considered relevant to the subject of this report.

The Liang et al. (1988) publication reported a study in which mice and rats were exposed in a manner that attempted to model open burning of coal in dwellings in rural southwestern China. Bituminous coal ("Gongchong coal ore") was burned in a shallow floor pit in the center of a room open via doors and windows to the ambient outdoor air, and the animals were placed in cages around the interior walls of the room. Exposures were 12 hours/day for 15 months for mice and 19 months for rats. Exposure concentrations were only reported for Benzo(a)pyrene (506 $\mu g/m^3$) and SO_2 (180 $\mu g/m^3$). Lung tumors, the primary effect of interest, were markedly increased in both mice and rats. These methods were not considered relevant to the goals of this report.

Two studies were considered relevant to the goals of this report because they involved exposures of animals to inhaled emissions from laboratory-scale coal combustion in a manner that could conceivably be employed to generate exposures relevant to full-scale processes. The first (Kirchner et al., 1980) addressed emissions from fluidized bed combustion, which has not gained significant commercial use. The second (Chen et al., 1990) employed a "drop-tube" furnace, which has some potential for use in modeling emissions from boilers currently in wide use.

a. Kirchner et al. (1980) Exposure of Mice to Emissions from Laboratory-Scale Fluidized Bed Combustion of Coal

Kirchner et al. (1980) exposed mice continuously for 500 to 1000 hours to diluted effluent from an "experimental process development-scale" fluidized bed combustor burning high-sulfur bituminous coal (5.5% sulfur "Sewickly" coal) and using calcitic limestone as the SO_2 sorbent. The 6-inch diameter vertical combustion chamber was operated at 2 kg coal/hour at 3% excess oxygen. Effluent was passed through cyclones to remove "50% of particles over 10 µm in diameter," diluted with clean air, passed through an "atmospheric effects simulator," and then introduced into a whole-body animal inhalation exposure chamber. The atmospheric effects (reaction) chamber irradiated the effluent with ultraviolet lights during a residence time of 15 minutes, but no additional reactants were added. The fly ash reaching the exposure chamber had a mass concentration varying between 15 and 25 mg/m^3, and a mass median aerodynamic diameter of 1 µm. Concentrations of measured gases included: SO_2 = 33 ppm, NO = 21 ppm, CO = 18 ppm, and THC = 20 ppm.

No peer-reviewed journal publication containing final results from the study was identified; the only citation located was a DOE technical report containing the proceedings of the DOE-sponsored October 1979 Hanford Life Sciences Symposium. That report described exposure-related reductions in food intake and body weight gain, a reduction of the *in vitro* uptake of *Staphylococcus aureus* by pulmonary macrophages removed from the lungs, and reduced hematopoietic (blood cell-forming) cell colonies after intravenous injection with bone marrow cells. The lungs had histological changes typically associated with heavy exposures to particles, including increased macrophages and epithelial cell hyperplasia in bronchioles and alveoli and perivascular cuffing with lymphoid aggregates. Exposure also altered the metabolism of organic compounds by the liver, with some metabolites increased and some decreased.

Overall, the high exposure levels, limited documentation of the composition of the exposure atmosphere, limited health response endpoints, and scanty reporting of results severely limit the report's utility for framing potential health hazards from exposure to emissions from fluidized bed coal combustion. Indeed, the "effective" fly-ash exposure concentration cannot be determined with confidence, due to the large upper bound of particle size. Because 5 µm is the approximate upper bound of particle size that can be inhaled by rodents (Miller, 2000), much of the particle mass in the exposure atmosphere would have been "wasted" in regard to inhalation and potential deposition in the lung, and thus the "effective" exposure concentration could have been quite different than the total particle mass concentration. The study did, however, demonstrate that a laboratory-scale combustor can be successfully operated daily for up to 40+ days.

b. Chen et al. (1990) Exposure of Guinea Pigs to Emissions from a Drop-Tube Furnace Burning Bituminous Coal or Lignite

Chen et al. (1990) Exposed guinea pigs (nose-only) for either one or two hours to effluent from a laboratory-scale "drop-tube" furnace burning either Illinois No. 6 bituminous coal or Montana lignite. Both coals were pulverized and dry sieved to achieve a particle diameter between 53 and 63 µm, and fed in a stream of nitrogen into a downdraft laminar flow furnace having a combustion zone temperature of 1250°K. The furnace is diagrammed in Figure 1 in Section V.D.1 below. The combustion effluent was quenched in argon in a cooling section, passed through a three-stage virtual impactor to limit upper bound particle size to 1 µm, and then passed into the exposure chamber. The average exposure mass concentration was 5.8 mg/m^3 and the average particle size was 0.21 µm. The concentration of SO_2 in the exposure chamber varied between 4 and 13 ppm, with an average of 7.6 ppm. Oxygen was added to the exposure chamber to maintain the concentration between 18% and 20%.

The health endpoints consisted of respiratory function, which was measured immediately after exposure and at times ranging up to 96 hours after exposure. For testing, the animals were anesthetized, and measurements consisted of lung volumes and CO diffusing capacity (alveolar-capillary gas exchange efficiency). One-hour exposures to emissions from the bituminous coal produced no changes in respiratory function, but 2-hour exposures caused reduced lung volumes and diffusing capacity. Lung volumes had returned to normal by 48 hours after exposure, but diffusing capacity still remained reduced at 96 hours. Two-hour exposures to emissions from Montana lignite also temporarily reduced lung volumes but did not reduce diffusing capacity.

This study demonstrated that 2-hour exposures to high concentrations of fresh, unreacted emissions from combustion of bituminous coal or lignite caused measurable changes in the lung function of animals, which is not surprising. Similar exposures caused lesser changes from low-sulfur lignite than from high-sulfur bituminous emissions, which was presumed to result from differences in sulfate levels. Although only single, acute exposures were performed, and the health response endpoints were limited, the study demonstrated that: 1) different coal types may present different types or magnitudes of health hazards, and 2) the "drop-tube" furnace can be used to generate emissions from combustion of pulverized coal in a controlled manner on a bench-top scale.

Although the exposures performed by Chen et al. (1990) did not fully meet the criteria described in following sections, the report serves as a "proof-of-concept" demonstration of the potential for using the "drop-tube" furnace as a starting point for laboratory-scale exposures.

2. *Exposures to Coal Fly Ash*

The bibliography in Appendix A contains 73 reports of exposures of cells or animals to coal combustion fly ash collected from combustion sources and used later for toxicological studies and none reporting experimental exposure of humans. The exposure method for many of these studies involved simply mixing the ash in cell culture medium or instilling it as an aqueous suspension into the tracheas of animals. The studies involving exposures to aerosolized ash employed a range of aerosol generation techniques, which were often poorly described. For example, the most recent citation involving aerosolized fly ash is that of Dormans et al. (1999), who describe the generation device only as an "atomizer."

There are currently several alternate approaches to generating aerosols from dry particles such as fly ash, including some using high-energy air jets or ultrasonic vibrations to disagglomerate particles and produce an aerosol. Others, as exemplified by the Wright dust feeder, use various mechanical techniques to introduce small amounts of material from a reservoir into an air stream. Contemporary dust generation methods were reviewed by Moss and Cheng (1995). If coal fly ash was to be aerosolized today, it is more likely that an air jet mill technique, rather than any of the methods used in earlier studies, would be employed; thus, the historical methods are not summarized here.

B. Experimental Exposures to Sulfur Dioxide and Sulfur-Containing Environmental Aerosols

A considerable body of literature exists on experimental exposures to sulfur compounds. The bibliography in Appendix B includes 67 citations describing experimental exposures of cells or animals to sulfur dioxide (SO_2), and 51 describing experimental exposures of humans. It also contains 64 citations describing exposures of cells or animals to sulfur-containing aerosols that might be found in the environment (albeit not all likely to be related to coal emissions) and 12 describing experimental human exposures to sulfur-containing aerosols.

All of the experimental exposures to SO_2 were generated by dilution of concentrated SO_2 with clean air. Tanks of compressed SO_2 are readily available from commercial sources, and the concentrated gas can be metered accurately to yield the desired concentrations. Methods for generating gaseous exposure atmospheres from compressed gas sources are straightforward, and were reviewed by Wong (1995).

The sulfur-containing aerosols were nearly all generated by nebulizers, which generate droplet aerosols from aqueous solutions of the desired compound. The exposures were to either diluted wet aerosols or to solid aerosol particles produced by drying or heat-treating wet aerosols. One citation (Anderson et al., 1992) described human exposures to carbon particles coated with sulfuric acid and sulfate using vapors from fuming sulfuric acid. Methods for generating aerosols from solutions were reviewed by Moss and Cheng (1995).

C. Experimental Exposures to Nitrogen Oxides, Nitric Acid, and Peroxyacetyl Nitrate

A tremendous amount of research has been conducted on the effects of nitrogen oxides. The bibliography in Appendix C includes 172 citations describing experimental exposures of cells or animals to nitrogen dioxide (NO_2) or nitric oxide (NO) and 41 citations describing exposures of humans. Much less work has addressed other nitrogen compounds that result from coal combustion. The bibliography contains 14 citations describing exposures of cells or animals to nitric acid (HNO_3) or peroxyacetyl nitrate (PAN) and 6 citations describing human exposures to those compounds.

Both NO and NO_2 can be purchased as compressed gases at a range of concentrations and diluted with air to the desired exposure concentrations. Vapors of HNO_3 can be generated by heating or nebulizing aqueous solutions. Nebulization is the preferred approach for generating aerosols. Desired exposure concentrations are achieved by adjusting the concentration of the original solution, the flow rate through the vaporizer or nebulizer, temperature, and dilution.

PAN is much more difficult to work with than the other compounds. It is unstable at molar concentrations and is not available commercially. It must be synthesized in the laboratory, and recent studies have used the synthesis procedure described by Kleindienst et al. (1990). PAN is generated by a step-wise liquid phase process using a non-volatile solvent such as tridecane as a medium, and by adding reactants sequentially to produce PAN. Bubbling air through the PAN/tridecane solution generates high concentrations of gaseous PAN, which can be diluted with air to the desired final concentration.

IV. Review of Current Programs Involving Animal Exposures

At the time of this report, there were two ongoing programs that included plans for toxicological studies of inhaled coal combustion emissions. Although neither of these programs planned to use approaches that were considered to fully meet the need of the research envisioned by this report, they are described below as background.

A. U.S. Environmental Protection Agency

The EPA Office of Research and Development's intramural research program was conducting toxicological research using materials collected from in-house laboratory-scale oil and coal combustors. At the time of this report, no real-time inhalation exposures to coal emissions had been conducted; rather, particulate matter collected from the combustor effluent was being instilled into the lungs of rodents or used for dosing cultured cells. The current effort was in part an extension of previous work with residual oil fly ash, which demonstrated that much of the toxicity was associated with soluble transition metals. Accordingly, at the time of this report, most of the effort on coal emissions had been directed toward collecting and testing the ultrafine fraction of particulate because it was enriched in metals compared to larger particle fractions. Extension of the effort to include inhalation studies of whole diluted unreacted and reacted emissions was envisioned. Coal emissions had been generated to-date using a pilot-scale down-fired tunnel furnace, and EPA's current plans were to use the same combustor for inhalation studies.

A summary of EPA's in-house coal emission toxicological research was presented by Dr. Andy Miller at the June 5–6 annual meeting at LRRI of the NERC advisory committee and sponsors. That presentation is reproduced in full in Appendix D.

B. Electric Power Research Institute

At the time of this report, the EPRI had launched a toxicology research program termed "Toxicological Evaluation of Realistic Emissions of Source Aerosols" (TERESA). The first source emission to be addressed by the program was coal combustion emissions, with the studies to be conducted by Harvard University under the direction of Dr. Petros Koutrakis. No studies or pilot exposures had yet been conducted at the time of this report. The planned approach involved conducting inhalation exposures at power plant sites using emissions taken from the stack or ductwork leading to the bottom of the stack. It was planned to pass the emissions though a reaction chamber to model the aging and atmospheric reactions of stack emissions, using high concentrations of reactants and ultraviolet light in a relatively small reaction chamber to achieve reactions that would take longer in the environment.

At the time of this report, exposures at three power plant sites had been envisioned, and one had been confirmed for study in the fall of 2003. The confirmed site was an upper Midwest plant burning Powder River Basin (PRB) coal. The plant had two operating units: one with SCR treatment and one without. Plans included a Midwest plant burning medium to high-sulfur eastern bituminous coal and a Southeastern plant burning high-sulfur bituminous coal.

To accommodate conducting the exposures on-site, only very short (hours) single exposures of small numbers of animals and evaluation of rapidly occurring effects were planned. The planned exposure atmospheres included fresh emissions, "aged" emissions, "aged" emissions reacted with ammonia, "aged" emissions reacted with volatile organic compounds, reactants alone, and clean air (controls). The planned toxicological protocol included measurements of breathing pattern, *in vivo* oxidative stress by chemiluminesence, bronchoalveolar lavage, hematology, electrocardiogram, and histopathology.

A summary of EPRI's TERESA program was presented by Dr. Ron Wyzga at the June 5–6 annual meeting at LRRI of the NERC advisory committee and sponsors. That presentation is reproduced in full in Appendix E.

V. Workshop on Strategies for Generating Exposures

A. Background and Preparations for the Workshop

Judging the feasibility of conducting meaningful research on the toxicology of coal combustion-origin emissions to which substantial populations are actually exposed (i.e., in contrast to "top-of-stack" emissions, to which few if any people are exposed) involves consideration of a range of issues. Accordingly, eliciting expert opinion spanning the range of key issues was viewed as a seminal step toward identifying and weighing options. These issues encompass population exposures, atmospheric chemistry of stack emissions, emissions reduction technologies, combustion technologies, coal type, and laboratory-scale generation of the exposure atmosphere. It is not apparent that an effort to elicit this range of opinion in an organized, interactive manner had heretofore been undertaken.

An expert peer workshop was conducted on February 27–28, 2003, to elicit current views of, and create discussion among, 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 held at the LRRI facilities in Albuquerque, NM, and consisted primarily of moderated, topic-oriented discussions. Other than the workshop introduction and presentation of a "straw-man" strategy, there were no formal presentations; some participants shared specific data as they pertained to issues during the discussion.

Several steps were taken in preparation for the workshop. A literature review on the composition of coal emissions was conducted to survey the scope of literature and areas of research focus and to determine whether "specifications" for the composition of an appropriate exposure atmosphere could be derived from the literature with confidence. The resulting bibliography is contained in Appendix F. Although there is a substantial literature on coal emissions, it was determined that a "straw-man" exposure composition could not be synthesized from the literature with confidence.

Discussions were held with individuals having experience in operating laboratory-scale coal combustors and resuspending coal fly ash, including Dr. Lung-Chi Chen of New York University, Bill Linak of the EPA, Jost Wendt, of the University of Arizona, and Dr. Adel Sarofim of the University of Utah. In January 2003, Dr. Mauderly was involved in detailed discussions of related EPRI research as a member of the Technical Advisory Committee of the EPRI TERESA program. In addition to the above contacts, Dr. Mauderly talked with Dr. Thomas Grahame of the Office of Fossil Energy at DOE Headquarters and Dr. William Aljoe and colleagues at the DOE National Energy Technology Laboratory in Pittsburgh, PA, to better frame the range of issues bearing on exposure decisions and to identify potential workshop participants. Dr. Mauderly then recruited workshop attendees from an extensive list of potential candidates. At the same time, Dr. Jake McDonald of LRRI prepared a "straw-man" proposal for an approach to generating an exposure atmosphere as a starting point and catalyst for the workshop discussions.

B. Workshop Participants and Agenda

The workshop was attended by the 20 individuals, 15 from outside the Institute and 5 from LRRI, as listed in Table 1. Contact information for the participants and the workshop agenda are presented in Appendix G. The participants were selected to represent 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

External	
Bill Aljoe	DOE/National Energy Technology Laboratory
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
Jim Meagher	NOAA
Bruce Miller	Pennsylvania State University
Larry Monroe	Southern Company
Niki Nicholas	Tennessee Valley Authority
Annette Rohr	Electric Power Research Institute
Roger Tanner	Tennessee Valley Authority
John Watson	Desert Research Institute
LRRI	
Joe Mauderly	Project Manager
Ed Barr	Exposure Manager
Jake McDonald	Analytical Chemistry Manager
Matt Reed	Toxicology Study Director
Richard White	Lab Supervisor, Exposure Operations

The workshop began Thursday morning, February 27, and ended at noon on Friday, February 28, 2003. The meeting on the first day was held at the LRRI inhalation facility on Kirtland AFB. After introductions and a presentation by Dr. Mauderly of background information on the project and constraints posed by the requirements of animal exposures, the group toured the LRRI wood smoke and engine emissions exposure laboratories to gain a sense of the nature of animal inhalation exposure systems and the logistics of animal exposures and health measurements. Dr. McDonald then presented a “straw-man” strategy for generating the coal emissions exposure atmosphere to summarize the information gathered by the Institute to-date and provide a starting point for discussion. The remainder of the workshop consisted of roundtable discussions moderated by Dr. Mauderly encompassing (in order): 1) the composition of real-world human exposures to coal emissions and reaction products, 2) general specifications for an appropriate exposure atmosphere, and 3) approaches to generating the desired atmosphere. The latter discussion examined in sequence two general starting approaches: 1) resuspending fly ash and 2) burning coal. It was acknowledged that either approach would require modification of the initial atmosphere to model the composition of predominant coal-origin air contaminants to which large numbers of people were exposed. After methods, advantages, and disadvantages of each approach were reviewed and general agreement reached, consensus was tested regarding the preference of the attendees for one of the two general approaches.

C. Key Underlying Considerations and Constraints

The conduct of toxicological studies of human exposures to key components of coal combustion emissions and their major transformation products necessarily involves several considerations and constraints. These constraints frame the plausibility of different approaches that might be taken.

1. *Desire for Exposures to Represent a Realistic Human Exposure Scenario*

Simulating population exposures to "coal emissions" presents more difficulty than does simulating exposures to many source emissions such as engine emissions or wood smoke. All people are exposed to fresh, diluted emissions from engines and wood stoves, and many spend time in locations that are continuously infused with fresh emissions; thus, diluted "end-of-tailpipe" or "top-of-chimney" emissions are plausible, albeit certainly not the only, exposure scenarios for those sources. In contrast, few, if any, people are exposed to "top-of-the-stack" coal emissions, and not many are sufficiently near to stacks to receive exposures to coal combustion-source pollutants that are not predominated by reaction products (e.g., much of the SO_2 is converted to sulfate before large numbers of people are exposed).

These considerations point toward the need to create an exposure atmosphere that is not simply a dilution of fresh coal combustion emissions, but rather represents the major pollutants of coal combustion origin as they are encountered by substantial numbers of people at some point downwind from the source. Of course, this necessarily incurs several compromises. Nobody is exposed downwind to coal emission materials and their immediate transformation products alone. Similarly, the chain of atmospheric reactions between coal emissions, sunlight, heat, and air contaminants from other sources is a continuum along which it is hard to draw firm lines - yet, lines of practicality must be drawn to select a single exposure composition. For example, one would clearly choose that the exposure atmosphere contain a substantial sulfate component but may not choose to include ozone, to which coal emissions contribute a lesser portion of reactants. Other decisions lie intermediately between those two more obvious choices.

The two coal-related human exposure scenarios currently raising the most questions from a public health viewpoint are : 1) eating game fish in which mercury is concentrated through the food chain and 2) repeated exposures to inhaled air contaminants over extended times (i.e., not a single exposure of only a few hours). Because this report considers only the latter scenario, it is considered that a thorough toxicological evaluation would include exposing animals repeatedly over periods ranging from several days to several months.

2. *Desire to Simulate Contemporary Exposures but Not the Cleanest or Most Advanced Technology*

The most appropriate study for evaluating current health hazards would be exposures that simulate downwind population exposures to emissions from combustion systems that represent coal types, combustion technologies, and emissions reduction technologies that predominate coal combustion demographics at present and in the recent past. Coal-fired power plant emissions are evolving in composition and concentration, as are other source emissions, but simulating only the cleanest contemporary (or future) emissions would not provide the information most useful in today's debates about causality of the effects of air pollution. Of course, it also would be very useful to document reductions in health hazards associated with reductions of emissions, but doing so would require establishing a "baseline" case for comparison. It is envisioned that the most appropriate initial study would therefore be aimed at establishing a reasonable and current baseline case.

3. *Need for Exposures to Produce Biological Effects*

Determining the exposure-response relationships for biological effects is at least as important to a solid research strategy as simply producing significant effects. It has been demonstrated over and over that significant toxicological responses can be elicited by exposure to essentially any material at some dose (exposure concentration x time factor), but simply demonstrating an effect is seldom an appropriate goal in itself. On the other hand, without knowledge of exposure-response relationships for adverse effects (i.e., the slope of the dose-response curve), one cannot place the magnitude of the hazard into context or estimate the gains that might be made by reducing exposure. Two conditions are necessary in order to evaluate exposure-response relationships: 1) effects must be tested at multiple exposure levels and 2) at least the highest exposure level must produce measurable (statistically significant) effects. It is not necessary that all exposure levels cause detectable biological effects; indeed, it is desirable that the lowest exposure level produce no effects, so that the level of realistic hazard and the threshold for effect can be bracketed.

4. *Requirements Imposed by Animal Exposure Protocols and Animal Management Criteria*

The total length of animal exposures has not been decided, but a good toxicological research strategy would certainly involve times ranging from several days to several months to determine the effects of repeated exposures. Such an exposure regimen necessitates the ability to expose daily without down time, which means that the exposure generation system must be reliable and that the resulting exposure atmosphere must be reproducible from day to day. There is no report of an experimental exposure of animals to a complex coal emissions atmosphere over periods of weeks, and no laboratory-scale combustors have been run continuously for that length of time. Either a back-up generation system must be available, or sufficient parts and an ability to maintain and repair the generator within non-exposure hours must be in place.

The requirement for prolonged, repeated exposures limits the range of suitable approaches. Along with the need to expose substantial numbers of animals (adequate group sizes and multiple exposure levels) housed under well-controlled conditions, this requirement essentially eliminates consideration of conducting the exposures at the site of a power plant. Repeated exposures at a power plant could be performed, but are not very feasible considering the animal exposure and maintenance facilities that would have to be constructed, and the fact that coal feedstocks and operating conditions may vary over time.

The animals must be maintained in a controlled environment with ready access to adjunct facilities. The exposure room must be large enough to accommodate multiple inhalation exposure chambers (at least one per exposure level); the facility must be controlled for adequate protection against spurious infections and stress; and there must be facilities within the controlled-access building for washing exposure chambers and storing food and other supplies. The exposure room must be near facilities for receiving, quarantining, and conditioning animals, performing necropsies and collecting samples, conducting some assays of collected samples immediately, and measuring some effects in real time (e.g., cardiac monitoring by telemetry). The animal housing and exposure rooms must be kept isolated from external contamination, and within specified light, noise, temperature, and humidity limits. In addition to the scientific aspects of conducting well-controlled studies, several of the above requirements are established by federal animal research regulations.

5. *Funding Constraints*

No specific level of funding was set as a limitation for the approach; however, funding is a ubiquitous limitation, and unlimited funding was not considered an appropriate assumption for this effort. For example, it was not considered practical to envision either setting up a full-scale coal-fired power plant at a toxicology research laboratory or constructing the necessary toxicological research facilities at a power plant. The latter, while incurring the lesser cost, would still incur costs on the order of $ millions even before the actual research began. With this in mind, a pre-determined constraint for the workshop discussion was the need to conduct studies in an existing toxicology laboratory with modest costs for renovation and capital equipment.

D. LRRI "Straw-Man" Approach

1. *Combustor Type*

It was assumed that the exposure atmosphere would be generated by burning coal in a combustor that struck a reasonable compromise between commercial-scale temperature/time profiles and clean-up strategies, and simplistic bench-scale burning of coal. Among the several sub-commercial-scale coal combustion systems published or currently in use, the "drop-tube" furnace was identified as the only system likely to be practical. "Pilot-scale" simulations of power plant combustion systems are larger and more costly than the program would likely accommodate, and the extent to which they accurately mimic commercial combustors would still be debated. Other "bench-scale" approaches are few and cruder, and were considered to depart too far from realism.

The combustion system that has come to be known generally as the "drop-tube" furnace was developed at MIT in the early 1980s by Dr. Adel Sarofim and colleagues

(Quann et al., 1982). Fundamentally, it consists of a vertical tube (i.e., "drop tube") for injecting pulverized coal (it has been used also with other materials) in a stream of gas downward through an electrically heated combustion section followed by apparatus to quench combustion, cool and dilute the effluent, and adjust the particle size distribution (Figure 1).

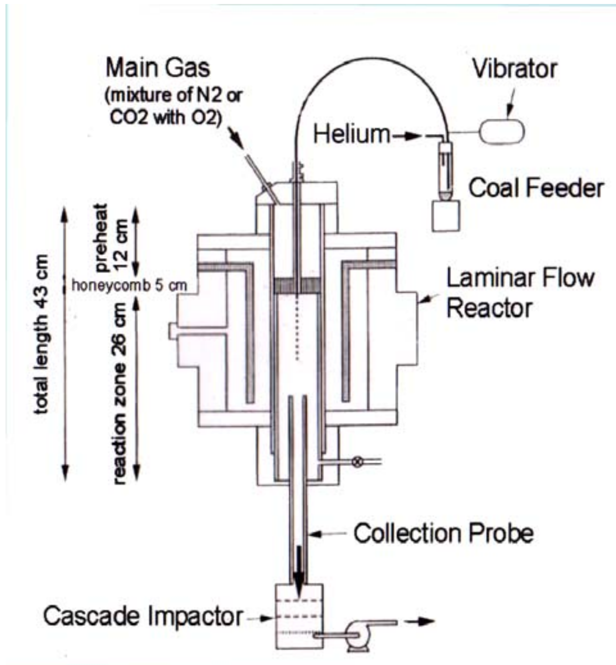

Figure 1: Drop-Tube Furnace

The drop-tube furnace was used initially to examine the composition of coal combustion products from different coal types. The first accompanying toxicological studies did not use the direct effluent, but rather used SO_2 and zinc oxide to model key components of interest (Amdur et al., 1986). In a subsequent study (Chen et al., 1990), the effluent from the drop-tube furnace was used directly for exposures of animals to combustion products of Illinois No. 6 and Montana lignite coals. The animals were exposed by nose only and only for up to two hours.

As typically configured, the drop-tube furnace is roughly nine feet tall when fully assembled, so it could fit into a typical high-bay exposure room. The feed rate of approximately 0.05 grams/minute is likely to produce sufficient effluent to supply multiple exposure levels. The operating temperature can range broadly which, along with varying dilution, quenching, and cooling, would allow operating parameters to be optimized. As a default, the same operating conditions used by Chen et al. (1990) might be used as a starting point. Although this device has been used by different investigators and is a "mature" technology, it has never been operated daily for prolonged periods; thus, operating trials, duplication of furnace units, and/or maintaining an inventory of key spare parts would be important considerations.

2. *Coal Type*

Powder River Basin (PRB) sub-bituminous coal was proposed. It has a large market share because it allows generating plants to meet sulfur emissions targets with less or no sulfur removal after-treatment. It has a lower sulfur content, a relatively higher content of earth metals (e.g., sodium, potassium, calcium), and a lower content of transition metals than most eastern bituminous coals. Its composition was viewed as an advantage in obtaining the final exposure composition that was likely to be desired. For example, sulfur removal may not be necessary if PRB coal is used, but might be required for higher-sulfur coal. In addition, PRB coal has a high reactivity and will combust easily in a drop-tube furnace, and will not have as many problems with carbon conversion as eastern bituminous.

3. *Modification of Emissions*

It was proposed that a cyclone be used to produce an upper-bound particle size cut of approximately 3.0 μm. Because health responses will be measured in rats and mice, particles larger than those respirable by rodents would not contribute to the "exposure" of interest. Failing to limit particle size to the rodents' respirable range could result in incorrect conclusions regarding the relationship between particle mass (and composition) and health effects. Anatomic differences and obligatory nasal breathing in rodents create differences in particle inhalabilty between humans and rodents. For example, only approximately 85%, 77%, 71%, and 65% of 2, 3, 4, and 5 μm particles are inhaled by rodents (Ménache et al., 1995).

The straw-man proposal did not include further treatment or modification of the emissions stream, other than the cyclone and dilution with clean air to the appropriate concentrations.

4. *Dilution Indicator and Exposure Concentrations*

Different exposure concentrations would be produced by parallel dilution with clean air of the effluent from the cyclone, resulting in identical ratios of the air contaminants at different concentrations. Setting and controlling exposure concentrations requires: 1) selecting a primary dilution indicator for specifying exposure levels, 2) selecting the concentrations of the primary indicator, and 3) selecting a controlling dilution indicator (if necessary) for managing the exposures. Any component of the exposure atmosphere could potentially be selected as the basis for setting exposure concentrations.

Because particles would be a substantial component of the coal emissions exposure atmosphere, and because particulate matter is an air pollutant receiving considerable current regulatory attention, it was proposed that respirable particle mass concentration would be the primary indicator for setting exposure concentrations. The particle mass could be measured as changes in filter weight measured under standardized conditions daily and averaged over the total length of exposure. It was proposed that four log dilutions be used to facilitate characterization of exposure-response relationships and the significance of exposure-related trends in health effects. To facilitate comparisons with results from the most extensive current program to compare effects of other source emissions (NERC), it was further proposed that the particle concentrations be set at 1000, 300, 100, and 30 $\mu g/m^3$. Any component of the exposure atmosphere could be selected as the basis for making daily adjustments to the dilution system, and it may prove more efficient to use real-time monitoring of a gas (e.g., SO_2 or CO) rather than particles for minute-by-minute monitoring.

E. Summary of Workshop Discussions and Recommendations

1. *Desired Composition of the Exposure Atmosphere*

The discussion of the desired exposure atmosphere was begun by asking several individual participants for their initial thoughts regarding key points to consider in bounding the composition of the exposure. The resulting discussion involved the entire group, served to elicit specific data and examples from participants to identify and discuss alternate views, and to move the group toward consensus regarding key parameters and target values. No attempt was made to record the entire discussion in detail (i.e., no transcript was made); rather, the key points that resulted were considered the "product" of the workshop.

All participants readily agreed at the outset that "top-of-the-stack" emissions were not the desired exposure. The following succinctly summarizes the key points offered by respondents to the first round of questions (in order of questioning).

- *Tom Grahame:* The particulate matter (PM) should be comprised of a sulfate/ash ratio of ≈ 100:1, and the sulfate should be ≈ 50% ammonium sulfate.

- *Jim Meagher:* Very little ash is detectable 20 km downwind of a power plant. On the other hand, exposures should not model emissions immediately downwind from a plant. Perhaps simulating 50 km downwind would be reasonable.

- *John Jansen:* You should have approximately 0.3–0.8 μg sulfate/ppb SO_2. The sulfur is not all neutralized at the simulated exposure location; use an ammonium-to-sulfate ratio of ≈ 1.6–1.8. The PM should contain little carbon; you certainly don't want to add carbon to the atmosphere.

- *Roger Tanner:* The desired PM would be relatively acidic. Although metallic components are minor constituents, they may still be important.

- *John Watson:* The "straw-man" proposal would result in largely a sulfate study. One might consider using a coal having a more toxic composition than PRB. (It was noted in discussion however, that PRB coals are usually used in unscrubbed systems. The more toxic coals [higher sulfur as well as trace elements] are typically fired in plants that have wet scrubbers that remove much, if not most, of the sulfur and trace species.)

It became evident from the discussion generated by the above individual comments that considerable agreement was evolving regarding the desired composition of the exposure atmosphere. As a next step, the group as a whole was asked to create a list of specific parameters and target values. A number of specific parameters were proposed, discussed, and agreed upon. The resulting list can be considered consensus guidelines—that is, points that evolved from the discussion and were listed, with which general agreement and no strong disagreement was voiced:

- Sulfate 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), 10% peroxyacetyl nitrate (PAN), and 15% nitric acid (HNO_3)
- Don't bother with O_3 or other secondary reaction products
- PM size cut of 2.5–3.0 µm is OK, at least cut to a size respirable by rodents
- Measure, but don't attempt to manipulate, Hg (participants were aware that Hg is a highly visible issue, and although it was not considered a target variable in the NERC study, they wanted it noted that it was discussed; i.e., that it was not overlooked)

2. *Approach to Generating the Exposure Atmosphere*

It became clear as the discussion progressed that there was a dichotomy of thought among the group regarding the best approach to use as a starting point for generating the desired exposure atmosphere. Given expression of valid rationales for both: 1) beginning by resuspending fly ash and 2) beginning by burning coal, these alternatives were discussed individually with no priority given to their order of discussion. The aim of the discussion was to identify the optimum approach for each alternative, making the prior assumption that the alternative under discussion would be selected. Although the participants remained divided to the end regarding the favored approach, both approaches were discussed thoroughly and constructively, and there was general agreement on the following summary points.

a. Begin Developing the Exposure Atmosphere by Resuspending Collected Fly Ash

1) Collection Site and Comparison to Stack Ash

Collect ash from the final hopper of the electrostatic precipitator (ESP) of an operating power plant boiler. ESPs typically contain a series of charged plates with collection hoppers located sequentially downstream. The final hopper would contain the ash having the smallest size distribution and a composition most similar to that of ash emitted from the stack. However, ESP ash could still be quite different chemically from stack ash (e.g., it may be less alkaline). Substantial quantities are collected, allowing the conduct of a study using ash from a single collection and generated under uniform plant operating conditions. Obtaining the ash would be technically simple.

Do not use collected ash from a bag filter after the ESP. Although many plants are configured this way and the particle size might be smaller, baghouse ash would have much more exposure to flue gas (e.g., SO_x, NO_x), and thus would likely differ chemically from emitted stack ash.

Perform a comparison of resuspended ESP ash to stack ash emitted from the same plant and at the same time as the ESP ash was collected. This will be necessary both to determine the resuspension process optimizing the simulation of stack ash and to determine the extent to which stack ash is actually simulated. The ESP ash should be mixed thoroughly (there is an ASTM method for mixing powders), aerosolized in the laboratory, size-selected (before and/or after aerosolization), and sampled as an aerosol for physical-chemical characterization.

Stack ash should be collected using a high-volume sampler followed by a mini-ESP or high-efficiency cyclone. While the easiest way to capture a large sample of post-ESP ash might be to use a "mini-baghouse," this would incur the same problem of prolonged exposure of the ash to flue gases as collecting from the baghouse in the first place.

The similarity achievable between resuspended ash and stack ash would determine whether this approach should be taken, and if so, would also help place the results of the study into context in comparison to "real" emissions.

Indeed, it is possible that collection from the stack by high-volume sampling might provide enough ash for the inhalation study (10–20 kg), although the collection time would be several days and perhaps a few weeks. If this proves possible, it would provide a sample preferable to ESP ash.

2) Source

Although ash from a pilot-scale combustor could be used, most participants viewed ash from a full-scale operating power plant as being preferable. Paul Chu offered assistance in identifying an appropriate plant and assisting with obtaining necessary permissions. Obtaining permission might be fairly straightforward if the utility and power plant could remain anonymous, and it should be possible to publish results without naming the plant if the coal and combustion system could be adequately described. It was suggested that it may be possible to obtain ash from a plant involved in the TERESA study, which would provide the potential advantage of linking, to at least some extent, results from the two programs.

3) Coal Type

The relative merits of ash from bituminous versus PRB sub-bituminous coal were debated. Overall, a majority preference was expressed for "low-sulfur Southern Appalachian" bituminous (LSSA) coal ash. LSSA coal ash would yield an exposure containing a more acidic particulate having higher sulfate and metal contents. However, it would be more "sticky" and thus harder to aerosolize than the less-sticky PRB coal ash. PRB coal ash would be more alkaline, have lower metal content, and would probably have a smaller size.

4) Adjustment of Sulfate

The sulfate content of the exposure atmosphere will probably have to be manipulated to achieve ratios of species typical of downwind exposures. The particulate sulfate should have an aerodynamic diameter centered on approximately 0.3 μm (i.e., accumulation mode), and should consist of approximately 55% ammonium sulfate, 40% ammonium bisulfate, and 5% sulfuric acid.

5) Adjustment of Nitrogen Species

The nitrogen species content of the exposure atmosphere will probably also have to be manipulated. Ideally, the atmosphere would contain the four principal components of "NO_y": NO, NO_2, PAN, and HNO_3. A mixture of NO and NO_2 can be obtained as bottled gas, and should be relatively easy to introduce. HNO_3 can be generated as a gas. PAN is problematic because it needs to be generated starting with a hydrocarbon mixture. It was generally agreed that adding PAN would probably not be practical, and could be justifiably disregarded as a secondary reaction product somewhat like ozone, which was not recommended for this exposure.

b. Begin Developing the Exposure Atmosphere by Burning Coal

1) Coal Type

Both PRB sub-bituminous and LSSA bituminous coals have advantages and disadvantages for this approach. Although a preference was expressed for LSSA if all factors were equal (for the compositional reasons stated above), it was recognized that PRB may be the better choice if coal is to be burned in the laboratory. The coal should be stored under inert gas at a cool temperature (ultra-low temperature freezing would not be necessary).

PRB coal has a higher moisture content than LSSA. It burns quickly; indeed, its flammability is a consideration for safe handling and storage. Its lower sulfur content may make hitting the desired sulfur targets easier. The high sulfur content of LSSA may yield too much primary sulfate, and it should be easier to hit target values by adding than by removing sulfate. Moreover, the higher carbon content of LSSA may be problematic if primary emissions are used as the starting point, whereas it may not be a problem when starting with fly ash.

2) Combustor Type and Operating Conditions

It was generally conceded that the drop-tube furnace was probably the only practical choice for combusting coal in the laboratory for toxicology studies. Larger-scale combustors of designs more similar to full-scale combustors may be preferable for their closer simulation of real-world emissions, but a combustor sufficiently large to achieve reasonable simulation would probably be beyond the physical and financial limitations of the program. However, it was considered very unlikely that a drop-tube furnace could be used without modification of the effluent to achieve the desired ratios of components in the exposure atmosphere.

Modification of the portion of the drop-tube system downstream from the heating section to change the time-temperature profile should be considered. If a drop-tube furnace is used, it will be important to consider the adequacy of its simulation of the time-temperature profile of a coal-fired power plant. As developed originally at MIT, the furnace simulated only the initial combustion of fuels. However, some drop-tube furnaces constructed more recently (e.g., at Pennsylvania State University and the University of North Dakota EERC) have residence times and gas cooling rates that provide more realistic temperature profiles.

Care should be taken to characterize the effluent (both particles and "flue gas") from the drop-tube furnace and compare it to stack effluent from a full-scale power plant. It is relatively easier to control the consistency of feed rate in a full-scale plant than in a laboratory combustor, and variations in feed rate could lead to less incomplete combustion in the laboratory. If this occurs, the flue gas will differ from the full-scale situation (e.g., carbon monoxide and polycyclic aromatic hydrocarbons may be emitted in higher concentrations). Variations in the oxygen content in the feed gas could lead to differences in combustion temperature that, in turn, could lead to different amounts of vaporization and later re-condensation of trace metals. Variations in oxygen content could also influence the amount of sulfuric acid on primary particles emitted from the furnace. This would be less of a concern with PRB coal than the higher-sulfur bituminous coal, because particle-borne sulfur would be largely neutralized when using PRB coal.

The effluent from the drop-tube furnace will need to be modified to achieve the desired exposure. The particle size distribution will probably need to be restricted on both the high and low ends, rather than just on the high end as in the straw-man proposal. Data were shown demonstrating that power plant ESPs are more efficient at removing particles below 0.2 μm and above 0.8 μm than they are intermediate-sized particles. Cutting only the upper size at 3.0 μm would likely result in too much ash mass in relation to the other exposure components and also too great a portion of mass below 0.2 μm. There was discussion regarding the relative health importance and abundance of ultrafine ash compared to larger ash, but the take-home message was that, as described above for other parameters, achieving a realistic ash size distribution is also an issue for comparison to an operating power plant. It is also possible that even if the drop-tube furnace is used with PRB coal (as recommended), SO_2 and NO_x may have to be reduced to hit the desired exposure targets.

Several operating issues were discussed. The furnace will take several hours to come to the operating temperature necessary for exposures. For daily use, the furnace should be left on 24 hours/day, although the temperature can be ramped down outside of exposure hours. Feeding the PRB coal to the drop-tube may prove to be challenging for multiple-hour exposures because of its moisture content, and potential feeding strategies were discussed. The necessity of duplicate furnaces was discussed. If an assortment of key back-up parts is maintained, most problems could probably be fixed between exposures. Because of time required for cooling for maintenance and re-heating, repairs will probably require 10–12 hours of down-time. Overall, it appeared plausible to conduct the exposure using a single drop-tube system. The system is expected to cost between \$30 and \$50 thousand, and is not available "off the shelf" (i.e., it must be constructed).

c. Preferences for Resuspension of Ash versus Burning Coal

At this point in the discussion, the 15 non-LRRI participants were polled for their preferences for beginning the development of the exposure atmosphere by either resuspending collected fly ash or burning coal. Everyone was asked to express a preference, regardless of how weakly or strongly the preference was held. Only a total count was made; no attempt was made to record the votes of individuals. Many, but not all, agreed that a useful study could be done either way, given sufficient attention to ensuring that the final composition of the atmosphere met the desired criteria. However, the participants were approximately equally divided regarding the approach most likely to result in the best exposure (seven for resuspending collected ash versus eight for burning coal). Importantly, the attendees were in unanimous agreement that regardless of approach, such a study would provide useful information despite its inevitable limitations.

3. *Suggestions for Moving Forward*

A path forward was suggested and encouraged by several participants. Unless the method of generating the exposure is determined on some other basis, it was suggested that a comparison of resuspended ESP ash with stack ash be undertaken as a first step toward making the decision. The comparison would include size-specific chemical speciation. The suggestion was predicated on the views that: 1) this comparison was likely to resolve uncertainties, one way or the other, about the extent to which stack particulate emissions could be simulated and 2) this exploratory effort could be undertaken at modest cost without committing financially to either strategy. In contrast, it was acknowledged that if a drop-tube furnace were set up as an exploratory activity, the expenditure of time and money would probably preclude exploring the resuspension approach, and thus pre-determine the outcome.

4. *Name of the Exposure Atmosphere*

It was generally agreed that terming this exposure atmosphere "coal combustion emissions" was problematic. It was the consensus of the participants that this label would imply exposure to direct, un-reacted emissions (e.g., as in the Chen et al. 1990 study), rather than an exposure aimed at simulating emissions that had traveled downwind and were partially reacted. A few suggestions were made, but no strong consensus developed. It was the general view that no name stood out as clearly the best, and that any name implying something other than direct emissions would suffice. Suggestions included the following, with slight predominance of preference for the second:

"Aged Coal Atmosphere"
"Simulated Aged Coal Plume"

VI. Opinion of the NERC Advisory Committee

The National Environmental Respiratory Center (NERC) is a government-industry program to improve our understanding of the contributions of individual air contaminants and their combinations to the health effects associated statistically with air pollution. Its strategy involves conducting identically designed inhalation toxicology exposure-response studies of selected source emissions, using very detailed protocols for both characterizing the composition of exposure and for characterizing a broad range of health effects. The combined database of composition-response results from the individual studies will allow statistical analyses disentangling the roles of pollutant species and their combinations, in lieu of the ability to develop a database having the same scope and detail by field sampling and epidemiology.

Because the NERC program is contemplating the inclusion of coal combustion emissions among its source-specific exposures, the evaluation described above was discussed with the Center's External Scientific Advisory Committee and sponsors at the June 5–6, 2003 annual Center meeting. The composition of the Committee, which is heavily weighted toward health experts familiar with air quality issues, but also includes a renown atmospheric chemist (Chow) is listed in Table 2.

Table 2: NERC External Scientific Advisory Committee

Morton Lippmann, PhD, Chair New York University	John Vandenberg, PhD U.S. Environmental Protection Agency
Michael Bird, MSc, PhD, DABT, C.Chem, FRSC International Agency for Research on Cancer	Sverre Vedal, MD, MSc National Jewish Medical and Research Center
Bill Bunn, MD, JD, MPH International Truck & Engine Co.	Ron White, MST Johns Hopkins University
Judith Chow, ScD Desert Research Institute	Ron Wyzga, MS, ScD Electric Power Research Institute
Gerald van Belle, PhD University of Washington	

The NERC committee enthusiastically agreed that a meaningful study of "downwind" coal emissions could be conducted using approaches framed by the workshop and recommended that such a study be undertaken by NERC using an experimental design identical to that of the other NERC studies completed or underway (diesel emissions, gasoline emissions, hardwood smoke). Upon reviewing options for generating the coal emissions exposure atmosphere, the Committee expressed its strong view that the exposure should be developed beginning by combusting coal, and agreed that the drop-tube furnace was the most appropriate combustion device for such a study. It was the Committee's view that, although interesting research could be conducted beginning by resuspending collected ash, research results sufficiently convincing and broadly accepted to have an impact on regulatory decision-making would most likely need to be obtained by exposing animals to atmospheres generated by combusting coal. In that context, the Committee recommended against expending resources to compare ESP ash to stack ash.

VII. Conclusions

It is concluded that it is feasible to generate inhalation exposure atmospheres suitable for conducting meaningful inhalation toxicology studies of the health hazards of population exposures to coal combustion emissions. However, as might be expected, the most appropriate approach depends on the specific human exposures to be modeled and the specific study design. The approaches taken by EPA and EPRI may be well-suited to their specific research interests, but there is a need for research providing information that will not be obtained by those programs. Specifically, there is a need for subacute (or subchronic, depending on preferred terminology), repeated exposures of sufficient numbers of animals to allow study of a range of health effects. Such exposures would be conducted five days a week at a minimum, and preferably daily, for at least a few weeks, if not months. The health assays would incorporate evaluations of respiratory and cardiac effects using methods that produce information relevant to the effects purported on the basis of epidemiology to be attributable to air pollution.

It is concluded that research having the greatest impact, and perhaps also the greatest scientific validity, would be conducted using an exposure atmosphere generated by burning coal, followed by modification of the effluent to achieve the desired target composition. Further, it is concluded that the drop-tube furnace is currently the most appropriate device for laboratory-scale generation of coal combustion emissions despite its obvious differences from full-scale combustors. Finally, it is concluded that the target composition parameters outlined by consensus of the workshop participants frames an appropriate final composition for the initial inhalation toxicology studies. The exposure would simulate exposures to key "downwind" products of coal-fired power plant emissions, and might most appropriately be termed "downwind coal combustion emissions."

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