



Assessment of Secondary Coal Combustion Emissions: The TERESA Study

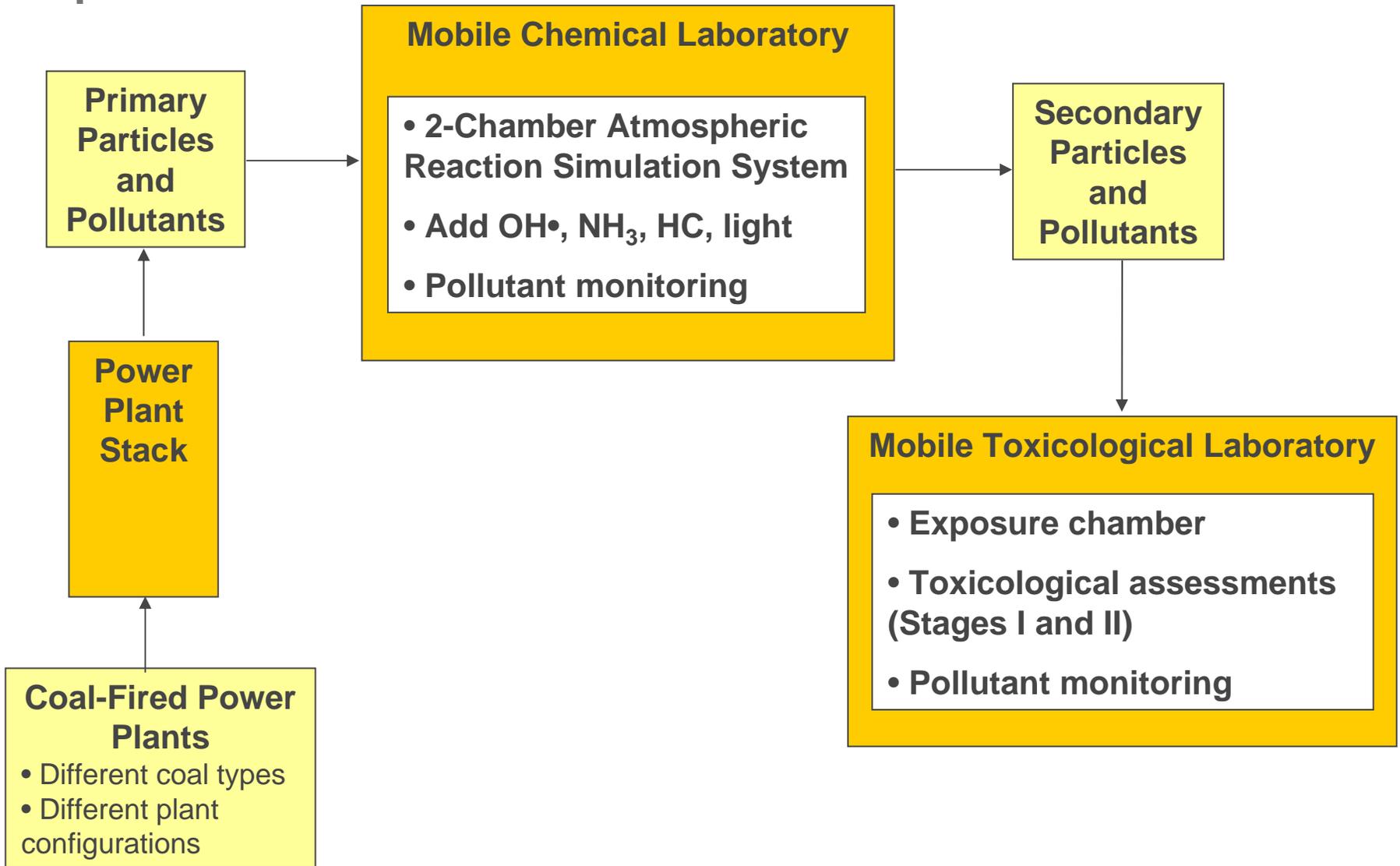
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January 22, 2004

TERESA: Toxicological Evaluation of Realistic Emissions of Source Aerosols

- Primary Objective: Determine the toxicity of *realistic* coal combustion emissions.
- Approach:
 - Evaluate toxicity of *secondary* coal combustion emissions at multiple power plants in the U.S. by exposing laboratory rats to a variety of simulated atmospheric scenarios.
 - Determine relative toxicity of coal combustion and mobile source emissions, as well as ambient PM_{2.5} (concentrated ambient particles; CAPs).

Study Design



Background and Motivation

- **Key issue:** increase our understanding of the *sources* and *components* of air pollution responsible for health effects.
- Two sources of information exist on the health effects of coal-fired power plant PM:
 - Studies examining the health effects of components of coal combustion emissions (e.g., sulfate, sulfuric acid).
 - Studies examining the health effects of coal fly ash.

Coal Fly Ash (CFA)

- Mostly intratracheal instillation studies:
 - Reductions in antibody-forming cells in rats (Dogra et al., 1995), and total/vital capacity in guinea pigs (Chen et al., 1990), changes in lung histopathology in hamsters (Wehrer et al., 1979, 1980).
 - 2, 10, and 50 mg of CFA instilled in rats: minor differences between CFA and TiO₂ (negative control) groups (Arts, 1993).
- *In vitro* studies: acellular OH generation and cytotoxicity in rat epithelial cells (van Maaenen et al., 1999); decreased phagocytic activity in mouse AM (Fisher and Wilson, 1980); little effect on DNA (Prahalad et al., 2000, 2001), effect seemed to be V- and Ni-mediated; some studies do show genotoxic effects.
- Few inhalation studies:
 - MacFarland et al. (1971) and Alarie et al. (1975) in monkeys and rodents: no unique biological effects from CFA exposure.
 - Rats exposed to CFA (0.6 mg/m³, 4.25 mg/m³) 8 h/day for up to 180 days: no effect in the low exposure group and only minor effects related to BAL macrophages in the high exposure group (Raabe et al. 1982).
 - Hamsters exposed to 2 mg/m³ CFA (inhalation exposures) for 180 days: no change in surfactant properties (Nishimura and Negishi, 1995).

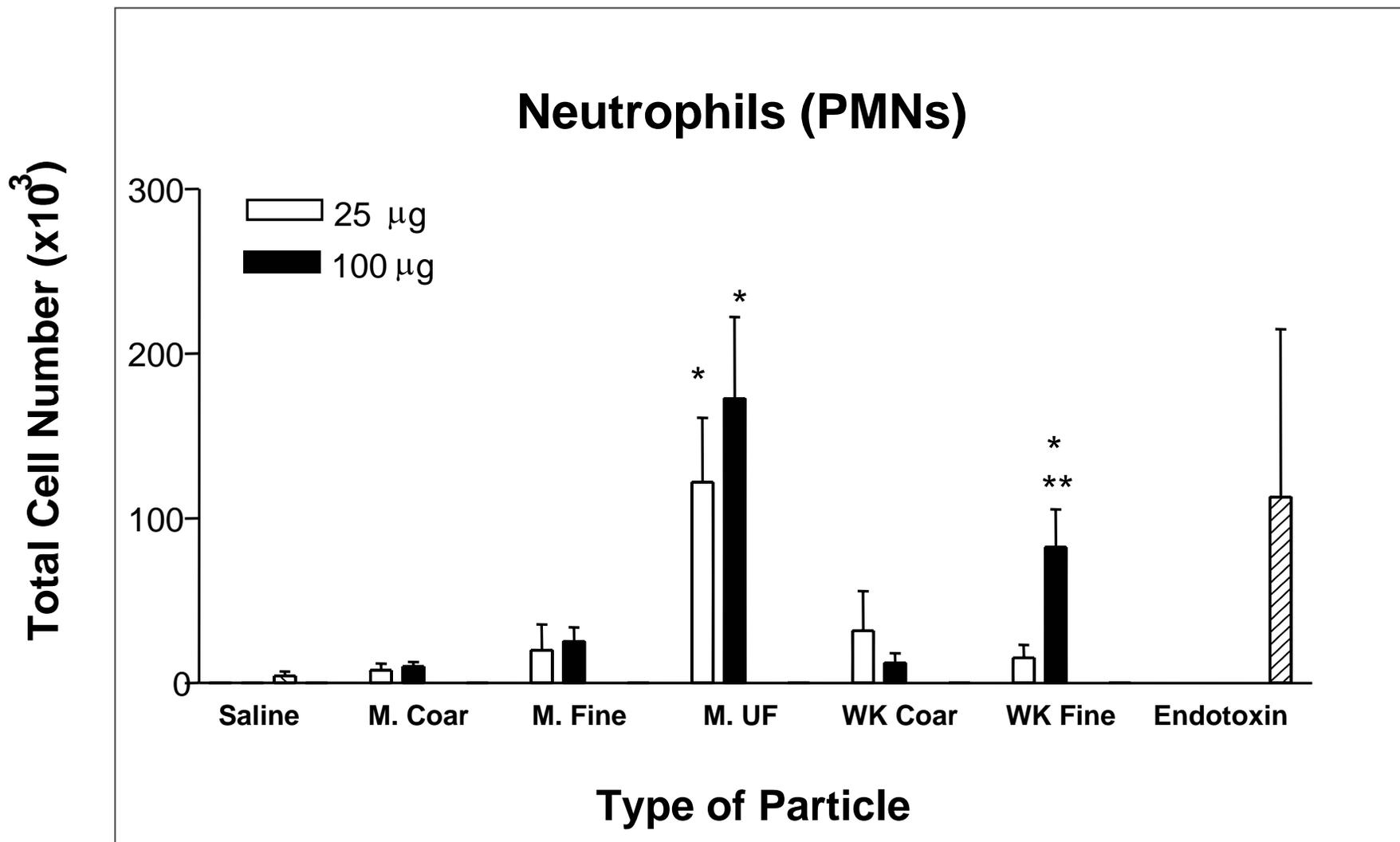
Coal Fly Ash (CFA): Effect of Size and Composition

- Using eastern bituminous coal, PM_{<2.5} was more cytotoxic and mutagenic than larger fractions, and particle size was inversely related to metal content of the ash (Mumford and Lewtas, 1982).
- CFA from bituminous coals appears to be more toxic than lignite coals (Smith et al., 2000).
- Mice exposed to CFA samples with the highest levels of metals showed the greatest effect on enhanced susceptibility to infection (Hatch et al., 1985).
- Higher metal sulfate ultrafine aerosols from a bituminous coal induced greater effects on pulmonary function in guinea pigs than a lower sulfur coal (Chen et al., 1990).
- Importance of ultrafine fraction??

Elemental Analysis of Ultrafine, Fine, and Coarse Coal Fly Ash (Gilmour et al., in press)

Element	MT UF	MT <2.5µm	MT>2.5 µm
µg/g ash			
Si	28,500	156,742	222,875
Al	93,780	103,979	108,800
Ca	82,900	89,858	115,175
Fe	6,920	53,929	30,350
S	39,400	7070	9,130
Mg	14,600	27,721	31,300
Ti	1845	6353	6180
K	1155	9358	5660
Cl	659	1264	1460
Ba	16200	2298	1843
P	10530	1080	979
Sr	7480	3426	3858
V	712	208	108
Ni	330	347	
Nb	910	176	22
Mn	487	1048	907
Cd	1620	463	
Se	565	136	
Ga	460	83	27
Cu	420	320	77
Elements %	22.5	47	54
Oxygen %	16.5	44.5	45
Carbon %	unknown	0.4	0.5

Effect of Coal Fly Ash Instillation on PMN Numbers in Mouse Lungs



Limitations of Coal Fly Ash Studies

- Studies using collected primary CFA (from ESPs or pilot combustors):
 - Low quantities of primary CFA are emitted from U.S. power plants
 - Populations are exposed to *secondary* PM.
 - Possible differences between collected particles and those that penetrate the ESPs into the ambient environment.
 - Instillation and *in vitro* studies tend to involve very high doses.
 - Possible changes in PM characteristics during storage.
- Inhalation exposure studies:
 - All studies have used pilot combustors: emissions from pilot combustors may differ from full-scale plants due to differences in surface area/volume ratios and therefore time-temperature histories.

Knowledge Gaps

- No information on the toxicity of secondary particles formed through SO₂ conversion in the atmosphere.
- No assessment of the toxicity of actual plant emissions.

TERESA Objectives

Primary Goal:

- Investigate and clarify the impact of the sources and components of PM_{2.5} on human health via a set of realistic animal exposure experiments.

Specific Objectives:

- Investigate the relative toxicity of coal combustion emissions and mobile source emissions, their secondary products, and ambient particles.
- Assess the effect of atmospheric conditions on the formation/toxicity of secondary particles from coal combustion and mobile source emissions.
- Evaluate the impact of coal type and pollution control technologies on emissions toxicity.
- Increase understanding of toxicological mechanisms of PM-induced effects.

Plant Selection

Program currently includes 3 coal-fired plants (with additional plants planned):

1. Upper Midwest: PRB coal (low sulfur, low ash).
2. Southeast: low sulfur (<1%) eastern bituminous coal, no scrubber for post-combustion SO₂ removal, with or without selective catalytic reduction (SCR) for NO_x removal.
3. Medium-to-high sulfur (>2-3%) eastern bituminous coal, scrubbed unit, with or without SCR.

Atmospheric Reaction Simulation System

- Critical component of TERESA.

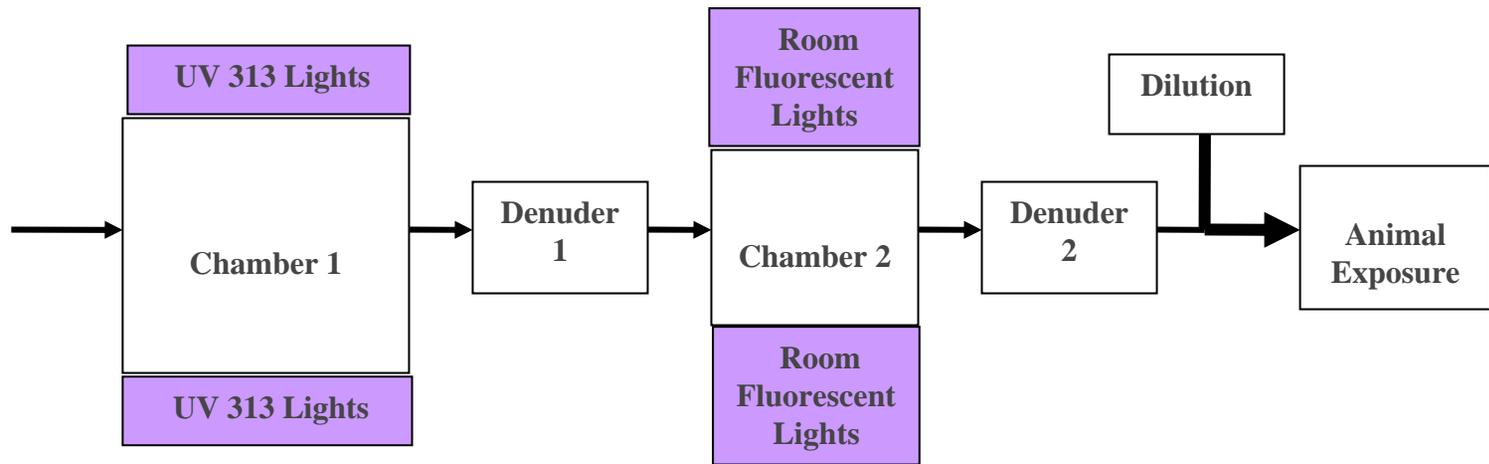
Technical Requirements

- Large, stable, and reproducible aerosol mass concentrations for animal exposures.
- Consistent size distribution across exposures.
- Sufficient flow of aerosol for exposure and characterization.
- Stable output in a short period of time.
- Secondary particles generated using typical atmospheric pathways and conditions (temperature, pressure and RH), without incorporation of compounds not present in the atmosphere.
- Aerosol components (SOA, sulfate, metals) in ratios consistent with typical average values in an aged plume.
- Low concentrations of unreacted gases (SO_2 , NO_x , O_3) during animal exposures.
- Small photochemical chamber for use in mobile laboratory installed in a refurbished bus.
- Minimal particle losses.

Atmospheric Reaction Simulation System

- Two-chamber design.
- Add atmospheric oxidants (OH radicals) to convert SO₂ and NO_x in stack gas to sulfuric acid and nitric acid.
- Chamber 1: Designed to oxidize 20-35% of SO₂ to sulfuric acid with a 60-minute residence time using O₃ photolysis as a source of OH radicals. For some exposure scenarios, NH_{3(gas)} will be added to partially neutralize acidic sulfate particle strong acidity.
- Chamber 2: Designed to coat particles with SOA through addition of VOCs (α -pinene) and ozone. Simulates the formation of SOA from the plume mixing with biogenic emissions.
- Sequential approach simplifies chemistry and avoids complex photochemical oxidation of organics.
- Novel “gas-cleaning system” (nonspecific denuder) uses a gas-permeable membrane to removal excess SO₂, NO_x, ozone, and other pollutant gases while maintaining the secondary particles.

Dual Chamber System



Exposure Scenarios

Scenario	Composition	Simulated Atmospheric Condition
1	Gas- and particle-free air	Sham exposure
2	Primary (un-aged) emissions diluted to the range of 50 µg/m ³ SO ₂ using clean air (same dilution as for 3, 4, and 5 below)	Primary stack emissions
3	Primary emissions + hydroxyl radicals	Aged plume, oxidized stack emissions, sulfate aerosol formation from nucleation
4	Primary emissions + hydroxyl radicals + ammonia	Aged plume, sulfate aerosol partially neutralized by ammonia
5	Primary emissions + hydroxyl radicals + ammonia + VOCs	Aged plume, mixture of neutralized sulfate and secondary organic aerosol derived from biogenic emissions

Plus additional Control Scenario: atmospheric components only, no emissions

Exposure Characterization

- PM mass, number, size distribution (including ultrafines)
- PM components:
 - Sulfate, nitrate
 - EC/OC
 - Ammonium
 - Metals
 - Particle strong acidity
 - Selected organics (eg. PAHs)
- Gaseous pollutants:
 - CO
 - NO₂
 - SO₂
 - Ozone
 - NH₃
 - Formaldehyde

Animal Exposure and Toxicological Assessment

- Conducted in separate mobile toxicological laboratory.
- 4-hour exposures, with 1-hour baseline and recovery periods (room air).

Stage I Assessment (normal rats):

- Pulmonary function/breathing pattern
- *In vivo* oxidative stress via chemiluminescence
- Blood cytology (CBC/differential)
- Bronchoalveolar lavage (LDH, β NAG, total protein)
- Pulmonary histopathology

Stage II Assessment (rat MI model; Wellenius *et al.*, 2002):

- Telemetry: cardiac function (ECG, HR, HRV), BP, temperature
- Blood chemistry (endothelin-1, CRP, IL-1, IL-6, TNF α)
- Pulmonary function/breathing pattern

Mobile Source and CAPs Assessment

- Mobile source assessment:
 - Sample diesel and/or gasoline engines (specific age and type TBD).
 - Methods for atmospheric simulation, animal exposure, and toxicological assessment will be completely analogous to the methods used for coal combustion emissions.
- Concentrated ambient particles (CAPs):
 - Use existing data from the Harvard School of Public Health laboratory.
- Compare toxicities of the three particle sources/types.

Status and Schedule

- Laboratory/methods development work almost complete.
- Outfitting of mobile laboratories almost complete.
- Fieldwork at first plant scheduled for early March.