NO_x, Fine Particle and Toxic Metal Emissions from the Combustion of Sewage Sludge/Coal Mixtures: A Systematic Assessment.

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By

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DISCLAIMER

ABSTRACT

This research project focuses on pollutants from the combustion of mixtures of dried municipal sewage sludge (MSS) and coal. The objective is to determine the relationship between 1) fraction sludge in the sludge/coal mixture, and 2) combustion conditions on a) NOx concentrations in the exhaust, b) the size segregated fine and ultra-fine particle composition in the exhaust, and c) the partitioning of toxic metals between vapor and condenses phases, within the process. To this end we shall use an existing 17kW downflow laboratory combustor, available with coal and sludge feed capabilities. The proposed study will be conducted in concert with an existing ongoing research on toxic metal partitioning mechanisms for very well characterized pulverized coals alone. Both high NOx and low NOx combustion conditions will be investigated (unstaged and staged combustion). The proposed work uses existing analytical and experimental facilities and draws on 20 years of research on NO_x and fine particles that has been funded by DOE in this laboratory. Four barrels of dried sewage sludge are currently in the laboratory. Insofar as possible pertinent mechanisms will be elucidated. Tradeoffs between CO_2 control, NO_x control, and inorganic fine particle and toxic metal emissions will be determined.

For the Second Quarter of this project we present our data on particle size distributions obtained from combustion of MSS and Gas, MSS and Coal and Coal and Gas alone.

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INTRODUCTION

Municipal sewage sludge (MSS) is formed during wastewater treatment. Sludge is the largest by-product of wastewater treating plants and it's processing and disposal represent one of the most environmentally challenging aspects of the wastewater treating process (Werther and Ogada, 1999). As discharged, the sludge is contained in a dilute (0.25 to 12 wt%) aqueous suspension and contains hazardous compounds. A description of how sludge is generated can be found in Werther, and Ogada (1999).

One of the options currently being considered to improve the treatment of this sludge is a process involving heat treatment (to render the sludge biologically inactive) followed by dewatering, drying, pulverizing, and combustion (Hall, 1992). The advantages of this combustion process are: a large reduction in disposal volume, thermal destruction of toxic organics, and recovery of the energy value of the carbon sources in the sludge (Werther and Ogada 1999). However, there are a number of potential disadvantages including NOx formation (due to the high nitrogen content of most MSS), handling of the raw material (due to the toxicity of metals and dioxins in the feedstock), possible boiler slagging and fouling problems (due to the high inorganic content of the feedstock), and the release of toxic trace elements into the environment (Cenni, et. al., 1998).

A study was conducted to investigate the partitioning characteristics of selected trace elements during the combustion of a dried, treated (Swiss Combi Process), and pulverized MSS at commercially relevant temperatures (i.e. combustion zone and post-combustion zone temperatures similar to those expected in commercial combustors. The objectives of this study were:

- to quantify the partitioning of selected major inorganic (silicon, iron, calcium, and aluminum) and trace elements (zinc, arsenic, selenium, and antimony) during combustion of MSS in a natural gas flame
- to quantify the partitioning of selected major inorganic (silicon, iron, calcium, and aluminum) and trace elements (zinc, arsenic, selenium, and antimony) during co-combustion of MSS with a well-characterized bituminous coal (Ohio 5/6/7 blend)
- to identify mechanisms where possible for trace element vapor-to-solid phase partitioning
- to explore synergistic effects for co-combustion of MSS with coal

EXPERIMENTAL FACILITIES

Experimental work was conducted in a 6m tall, 0.15m ID vertical downflow aerosol furnace (Figure 1) operated at a feed rate of 12.5kJ/s (42,800 BTU/hr) for all feed sources. The furnace is designed to simulate the time/temperature histories and complex particle interactions of commercial-scale combustors while still providing a flow stream that is sufficiently well characterized to allow extraction of rates and mechanisms. This furnace is described in detail elsewhere (Seames, 2000). Natural gas was premixed with air in the body of a laminar flame burner prior to discharge and ignition. MSS was fed using a volumetric screwfeeder. The MSS was blown off the screw tip and into the furnace using a metered stream of 5 psig transport air and then premixed first with the combustion air and then with the natural gas prior to ignition in the furnace. For coal combustion, coal was also fed using a volumetric screwfeeder. Coals was blown off the screw tip and into the furnace using a metered stream of 5 psig, transport air, then mixed with the primary air prior to discharge into the furnace. When both coal and MSS were burned, the primary air was first mixed with the coal, then with the MSS in the burner prior to discharge and ignition in the furnace. Coal and MSS were blended as a 50-50 mixture based on heat load (i.e. ~ 6.3 kJ/s for each fuel feedstock).

All of the experiments were run at a naturally evolving temperature profile and 20% excess oxygen in the combustion air. Fly ash particles were withdrawn from a sampling port (Port 14) located 4.3 meters below the burner. Samples collected at Port 14 represent typical conditions late in the post-combustion zone.

Particle sampling was performed using a portable water-cooler aspirated sampling probe. Dilution nitrogen is fed to the sample tip and a vacuum pump is used to draw the nitrogen plus the furnace sample from the centerline of the furnace. The dilution nitrogen plus the cooling jacket on the probe serves to freeze subsequent reaction in the probe. It also condenses any species that will condense above atmospheric conditions (Davis, et. al., 1998). The sample is routed to a Berner-type low-pressure impactor (BLPI; Hillamo and Kauppinen, 1991). The bottom stage of the BLPI acts as a sonic orifice and controls the total flow rate through the impactor. By measuring this flow rate with a wet test meter prior to sampling, an isokinetic sampling rate can be calculated and then maintained by adjusting the amount of dilution nitrogen routed to the probe. This sampling system, shown in Figure 2, and its utilization with the University of Arizona combustor is described in detail elsewhere (Seames, 2000).

Sample sets that most closely duplicated the typical ash mass particle size distribution were selected for elemental analysis. Arsenic, selenium, zinc, and antimony analyses were performed by graphite furnace atomic absorption spectroscopy (GFAA) at Arizona. Major species (i.e.. silicon, calcium, iron, aluminum, and magnesium) were analyzed by flame atomic absorption spectroscopy (AAS) at Arizona while sodium was analyzed by flame atomic emission spectroscopy (AES) at Arizona.

EXPERIMENTS PERFORMED

MSS was prepared and provided by the Institute for Process Engineering and Power Plant Technology at the University of Suttgart (K. R. G. Hein, Director) by W. Scheurer and is representative of a typical German urban MSS (Scheurer, 1999).

A series of experiments were performed over a three-day period to meet the experimental objectives of this test program. A summary of the experiments performed is given in Table 1. Experimental conditions for the particle sample sets collected are summarized in Table 2. For the natural gas baseline experiment, one impactor set was collected at Port 14 (see Fig. 1) without using the inlet cyclone on the BLPI. For the other three test conditions (gas + MSS, coal only, coal + MSS), two matched impactor sets were collected at each test condition. A matched sample set consists of five membranes from a 1st collection test for particulate on atmospheric stages -- 6-10[1] -- of the BLPI without the preseparator cyclone attached to the BLPI inlet and six membranes from a 2nd collection test performed immediately before or after the 1st test collecting particulate on the subatmospheric stages -- 1-6 -- of the BLPI with a cyclone.

The following sample sets were selected for inorganic analysis:

Natural gas only:	FB-1
Gas + MSS:	MSS-2
Coal Only:	MSS-6
Coal + MSS:	MSS-4

EXPERIMENTAL RESULTS

Proximate/ultimate/heat of combustion analysis and detailed elemental characterization of the Ohio 5/6/7 blend coal used in this study were performed by a consortium of researchers under DOE program DE-AC22-95PC95101, “Toxic Substances from Coal Combustion – A Comprehensive Assessment”, (Senior, 1998). These data are shown in Table 3 (extracted from Seames, 2000).

Proximate/ultimate/heat of combustion analysis of the MSS was performed at Galbraith laboratories (Knoxville, TN). Inorganic elemental analysis was performed at Arizona. These data are shown in Table 4.

Temperature Profiles

Furnace temperature profiles were obtained for each test condition (excluding the gas only test condition[2]). These are shown in Figure 3. The similarity of these curves is not surprising and

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[1] A small number of particles were collected on the inlet membrane, stage 11. These were discarded and are not included in the results of this study.

[2] A typical gas-only temperature profile obtained at a similar heat load for the same combustion furnace can be found in Fig. 4.2 of Davis (1999).

verifies that all experiments were performed at nearly identical heat loads. Most importantly, the maximum combustion temperature for each test is similar (within 60K). As the flue gas travels away from the burner, the rate of the temperature decrease measured is qualitatively consistent with the differences in the heat capacity of the three flue gas streams[3].

Particle Size Distributions

The differential total ash particle distributions (PSDs) for all four experimental conditions are shown in Figure 4. These distributions were developed following a method originally developed by Markowski and Ensor (1977) for reporting impactor results. The mass collected on an impactor stage is divided by the volume of gas passing through the impactor during the sampling period. Using this concentration value for each impactor stage, the differential concentration is divided by the differential log10 of the particle size. This value is plotted versus the average particle size[4] associated with the differential particle size used. In Figure 4, four different size scales are shown from the same data in order to evaluate different attributes of the same data.

Four distinct particle regions are evident for the “Coal only” and “Coal + MSS” results: a vapor-phase region, a submicron fume region, a micron-sized “fragmentation” region, and a bulk (> 3mm diameter) fly ash region. By contrast, the “Gas only” distribution shows only two distinct regions – a vapor-phase region and a particle region while the “Gas + MSS” distribution shows three regions – a vapor-phase region, a submicron region, and a supermicron region.

For the sampling system employed in this work, Davis, et. al. (1998) demonstrated that (for flue gases generated by the combustion of gas flames doped with 100 ppmv semi-volatile metals) the smallest particle sizes (below about 0.1 microns) in the profile primarily represent vapor-phase material that nucleates in the sample probe. The vapor-phase material represents a greater fraction of the total material for the “Gas only” sample compared to the other distributions. More noteworthy, the co-firing of MSS with coal increases the vapor-phase material compared to coal alone or MSS fired with gas assist.

The presence of a submicron fume region from pulverized coal combustion has been explored in detail by previous researchers. The major refractory elements (Fe, Si, Al, and Ca) are believed to vaporize as suboxides formed by reduction with CO in the immediate vicinity of the burning char particle (Linak and Peterson, 1984; Senior and Flagan, 1982, McNallan, et. al., 1981). As the molecules diffuse out of the reducing atmosphere of the char, they are oxidized. These oxides are not very volatile and will nucleate while still in the flame zone to form a submicron fume (Mulholland and Sarofim, 1990; Sarofim, et.al., 1977; Natusch, et.al., 1974; Taylor and Flagan, 1982; Linak and Peterson, 1984; Helble and Sarofim, 1989, McNallan, et. al., 1981).

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[3] Flue gas with higher particle loading with have higher heat capacity; for these tests MSS+Gas < Coal Only < Coal + MSS particle loading.

[4] All of the impactor particle size data reported here is reported as the 50% cutoff aerodynamic particle diameter. Collection efficiency curves of the BLPI are reported elsewhere (Hillamo and Kauppinen, 1991). The relationship between the real diameter of a fly ash particle and the aerodynamic diameter is described elsewhere (Raask, 1981).

The fractionation of larger particles to submicron-sized particles also contributes to the submicron fume (Smith, 1980, McNallan, et. al., 1981, Helble and Sarofim, 1989). The contribution from fractionation is highly fuel dependent. The submicron fume is typically only 1-7% of the total fly ash mass but has a very high particle number density (McNallan, et. al., 1981, Wall, et. al., 1979).

Profiles in the submicron region are similar for the “Gas + MSS”, “Coal only”, and “Coal + MSS” test conditions. Previous work has shown that during combustion of the Ohio 5/6/7 blend coal, the mechanisms responsible for submicron particle generation are consistent with the submicron fume theory described above (Seames, 2000). Thus the similarity in profiles between these three test conditions suggests that submicron fume theory is applicable during the combustion of MSS co-fired with gas or coal.

The near micron-sized “fragmentation” region has only been recently observed (Linak, et. al., 2000; Seames, 2000) during pulverized coal combustion. This regime is believed to form by the swelling and fragmentation of larger particles (e.g. hollow cenospheres) during char burnout. Seames (2000) demonstrated that the partitioning mechanisms for volatilized trace elements (i.e. arsenic and selenium) could be different in the fragmentation region compared to the bulk fly ash region for some coal/combustion condition combinations. Co-combustion of MSS with coal results in a smaller fragmentation region compared to coal alone. This suggests that only the coal char particles are participating in the fragmentation mechanism. The absence of a fragmentation mode in the “Gas + MSS” PSD supports the conclusion that MSS ash particles are not subject to significant fragmentation leading to the generation of a micron-sized particle region.

The bulk fly ash profiles for the “Gas + MSS”, “Coal only”, and “Coal + MSS” experiments are similar.

Detailed information related to the particle size distributions from the sample sets collected during this test program is included in the Appendix.

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TABLES 1 through 4 on Pages 13 through 19

TABLE 1
A SUMMARY OF THE MSS EXPERIMENTS

Test Run #	Gas Feed Rate (Nm^3/hr)	Coal Feed Rate (kg/hr)	MSS Feed Rate (kg/hr)	Sample Set #	Sampling Times
MSS-1	1.12[5]	--	--	FB-1	60 mins
	1.12	--	1.36	MSS-1c	10 mins
				MSS-1	30 secs
				MSS-2c	10 mins
				MSS-2	30 secs
MSS-2	--	1.26	1.36	**MSS-3c**	10 mins
				MSS-3	25 secs
				MSS-4c	10 mins
				MSS-4	1 min 15 secs
MSS-3	--	1.58	--	MSS-5c	15 mins
				MSS-5	6 mins
				MSS-6c	15 mins
				MSS-6	6 mins

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[5] Assumes heating value of 1080 Btu/scf

TABLE 2
SUMMARY OF EXPERIMENTAL SAMPLING CONDITIONS

SAMPLE SET #	TOTAL COMBUSTION GAS RATE (slpm)	**SAMPLING RATE (slpm)**	**SAMPLING TEMP.** (^{o}K at Port where Sample was taken)	PORT 4 O_2 CONC (%)	PORT 4 CO_2 CONC (%)	PORT 14 O_2 CONC (%)	PORT 14 CO_2 CONC (%)	PORT 14 NOx CONC (ppm)
FB-1	261	2.48	891	2.9	10.2	5.1	9.1	127
MSS-1	265	2.58	905	3.1	12.3	4.7	11.3	812
MSS-2	265	2.58	905	3.1	12.3	5.6	10.4	780
MSS-3	262	2.17	1014	3.2	15.6	3.4	15.9	505
MSS-4	262	2.17	1014	2.9	15.8	3.4	15.9	505
MSS-5	250	2.03	916	3.4	15.8	3.5	15.9	640
MSS-6	250	2.03	916	3.4	15.8	3.5	15.9	640

TABLE 3a
PROXIMATE/ULTIMATE ANALYSIS OF THE OHIO COAL[6]

Proximate	wt%
Fixed Carbon	48.78
Volatile Matter	39.19
Moisture	2.33
Ash	9.70
Heating Value kJ/kg	29,900
Ultimate	
Carbon	71.07
Hydrogen	5.07
Nitrogen	1.37
Sulfur	2.62
Oxygen	10.17
Chlorine	0.0974
Moisture	2.33
Ash	9.70

TABLE 3b
OHIO COAL
ASH CHARACTERIZATION[1]

Component	wt%[7]
SiO_2	38.09
Al_2O_3	39.41
Fe_2O_3	13.03
TiO_2	2.04
CaO	2.24
MgO	1.02
Na_2O	0.92
K_2O	2.55
BaO	0.20
P_2O_5	0.51

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[6] From Senior, et.al. (1998)

[7] Sulfur-free basis

TABLE 3c

Mineralogical Characterization[8]

XRD[9] DETERMINATIONS	wt %
Quartz	19
Kaolinite	43
Illite	21
Pyrite	12
Siderite	0.5
Feldspar	tr[10]
Bassanite	tr
Sphalerite	tr
Marcasite	tr

Table 3d

Trace Element Forms of Occurrence[11]

Element	Form	Wt%
Arsenic	Organic	0
	Mono-sulfides	30
	Silicates	5
	Pyrite/sulfides	35
Selenium	Organic	40
	Mono-sulfides	5
	Silicates	0
	Pyrite/sulfides	55
Antimony	Organic	
	Mono-sulfides	
	Silicates	
	Pyrite/sulfides	

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[8] From Senior, et. al., (2000)

[9] XRD = X-ray Diffraction

[10] tr = trace

[11] Determine by selective leaching, from Senior, et.al., (1998)

ELEMENTAL CONCENTRARTIONS

Chemical Composition[12]	MIT/NAA (ppmw)	USGS[13] (ppmw)
Si	nd[14]	14096
Al	11100	6751
Ti	670	430
Fe	15100	10906
Ca	1400	816
Mg	760	252
Na	350	208
K	2500	1187
P	nd	200
Cl	880	nd
Li	nd	8
Be	nd	2.6
B	nd	64
Sc	3.90	2.8
V	23.0	19
Cr	19.0	11.4
Mn	19.0	15
Co	5.8	3.64
Ni	nd	13
Cu	nd	7.1
Zn	39	16
Ga	11.0	4
Ge	nd	7.3
As	19.0	13.2
Se	1.40	1.32
Br	23.0	13.9
Rb	21.0	8.4
Sr	100	74
Y	nd	7.2

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[12] From Senior, et. al. (1999)

[13] Optimal results from ICP-MS, ICP-AES, NAA, Hydride (Se), and Cold Vaporization (Hg)

[14] nd = not determined by this method

Chemical Composition	**MIT/NAA (ppmw)**	**USGS (ppmw)**
Zr	nd	10
Nb	nd	2.2
Mo	7.6	3.6
Ag	nd	<0.2
Cd	0.40	0.1
In	0.06	nd
Sn	nd	1
Sb	2.30	1.44
Te	nd	<0.2
Cs	0.63	0.56
Ba	102	49
La	9.40	7.29
Ce	14.0	13.7
Nd	11.0	nd
Sm	2.00	1.61
Eu	0.38	0.317
Tb	nd	0.237
Yb	1.10	0.73
Lu	0.22	0.102
Hf	nd	0.45
Ta	nd	0.1
W	nd	0.8
Au	3.e-3	<1
Hg	0.15	0.13
Tl	nd	1.0
Pb	nd	6.8
Bi	nd	<0.2
Th	2.70	1.59
U	**--**	0.93

TABLE 4
CHARACTERIZATION OF MUNICIPAL SEWAGE SLUDGE

ANALYSIS	VALUE	UNITS
Heat of Combustion	2858	**Btu/lb**
Proximate		
Moisture	5.12	wt%
Volatile Matter	39.20	wt%
Fixed Carbon	1.89	wt%
Ultimate		
Carbon	19.70	wt%
Hydrogen	3.08	wt%
Nitrogen	2.22	wt%
Chlorine	0.041	wt%
Sulfur	0.59	wt%
Ash	53.79	wt%
Ash Elemental Analysis		
SiO_2[15]	36	wt%
CaO	8.8	wt%
Na_2O	1.8	wt%
MgO	4.1	wt%
Fe_2O_3	15	wt%
Al_2O_3	19	wt%
Arsenic	6.7	ppmw
Selenium	3.3	ppmw
Zinc	664	ppmw
Antimony	7.2	ppmw

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[15] Major elements converted to oxides on a sulfur-free basis; data reported is not normalized

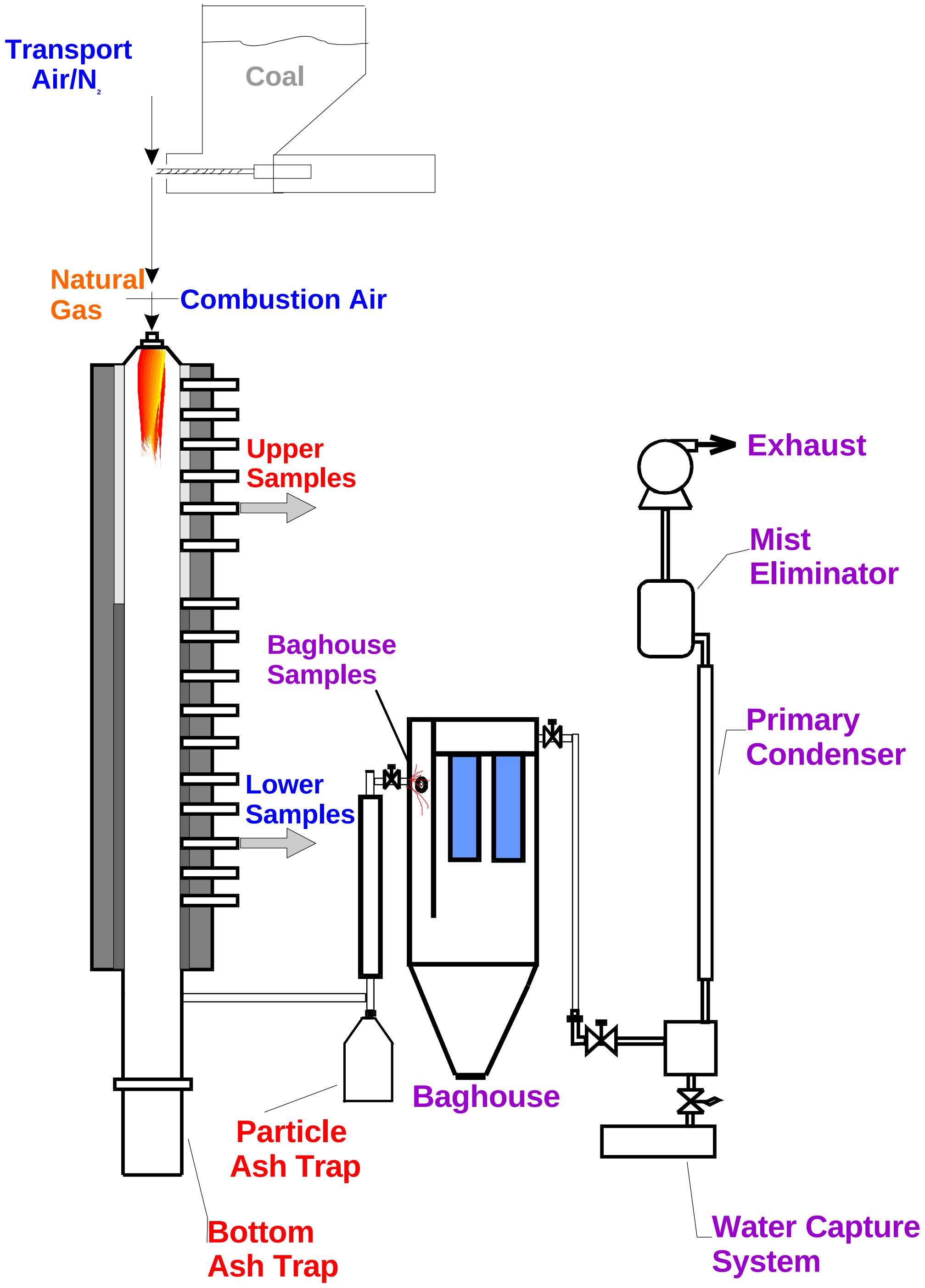

Figure 1. Overall schematic of University of Arizona Combustion System

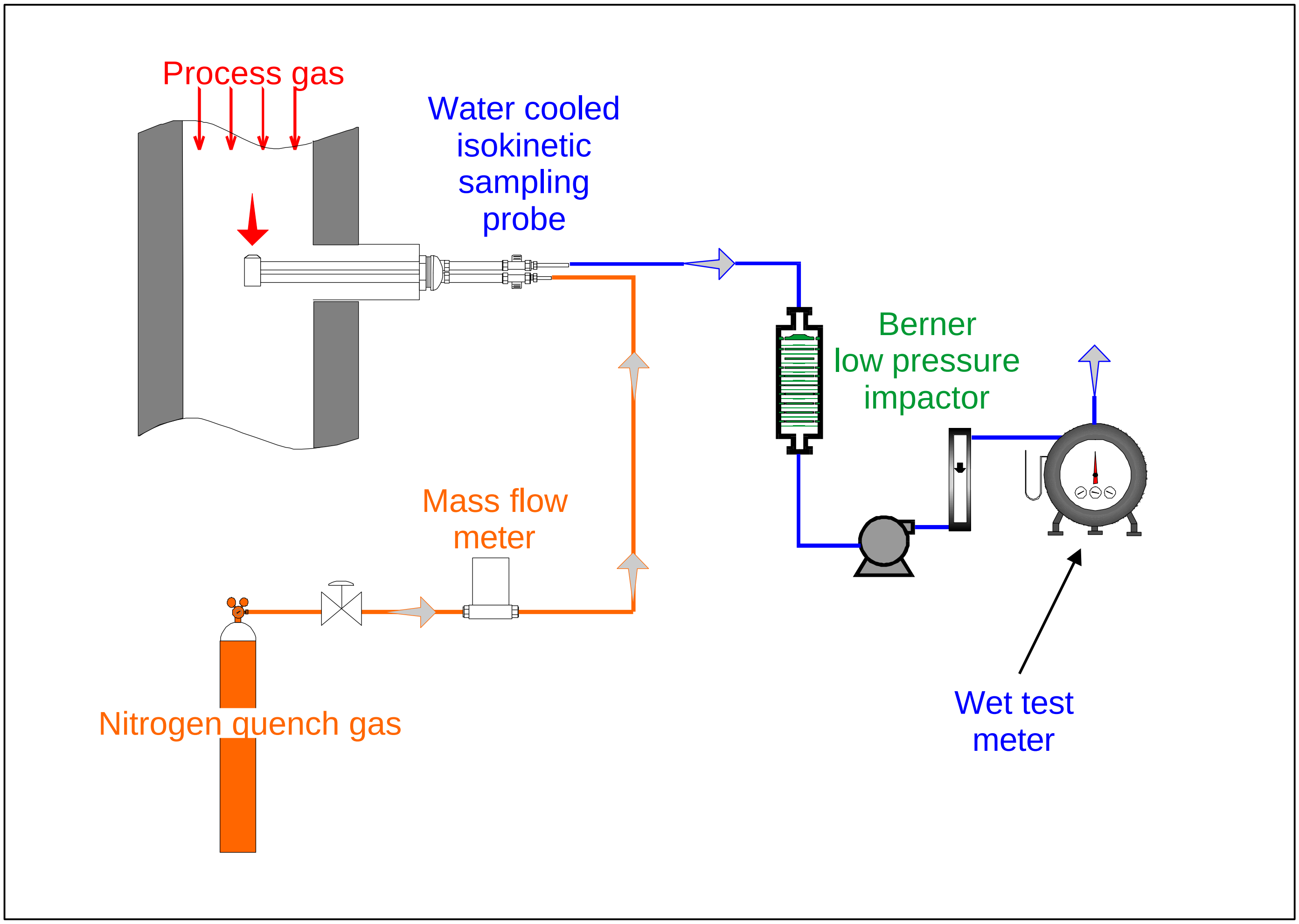

Figure 2. Simplified schematic of particle sampling system

Figure 3
Typical Naturally Evolving Temperature Profile
At Each Experimental Combustion Condition

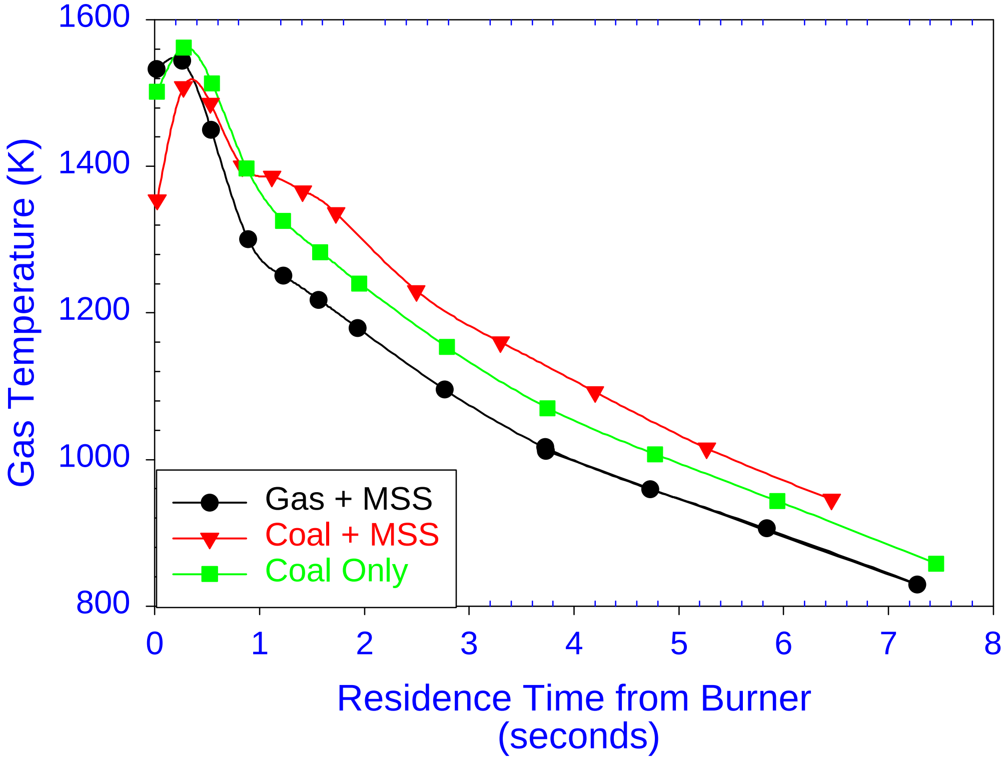

FIGURE 4
PARTICLE SIZE DISTRIBUTIONS FOR MSS EXPERIMENTS