**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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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) NO_x 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 work is progress using 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 NO_x and low NO_x 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 CO₂ control, NO_x control, and inorganic fine particle and toxic metal emissions will be determined.

Progress in the Sixth Quarter (January 1, 2002 through March 31, 2002) was slow because of slagging problems in the combustor. These required the combustor to be rebuilt, a job that is not yet complete. A paper describing our results heretofore has been accepted by the Journal Environmental Science and Technology.

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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 its processing and disposal represent one of the most environmentally challenging aspects of the wastewater treating process [1]. 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 [1].

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 [2]. 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 [1]. However, there are a number of potential disadvantages including NO_x 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 [3].

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

These have been described in the Second Quarterly Report 40838R02.

PROGRESS TO DATE

Experimental progress to date has been described in Quarterly Reports 40938R02 through 40938R05. These results have allowed the fine particle emissions from combustion of MSS alone and MSS combined with pulverized coal to be determined. Mechanisms describing trace metal partitioning have been described. Results will also be published in a paper entitled: “*A Study of Fine Particulate Emissions from Combustion of Treated Pulverized Municipal Sewage Sludge*” by Wayne S. Seames, Art Fernandez, and Jost O.L. Wendt, which has been accepted for publication by *Environmental Science and Technology*, the premier journal in the environmental field.

This progress has now been halted, due to a breakdown in the experimental combustor. It was found in early January that there were significant deposits attached to the walls just downstream of the refractory portion of the combustor. Attempts to dislodge these deposits resulted in destruction of portions of the wall. This caused large leaks of air into the combustor, and made staging studies impossible. We attempted to rebuild just a portion of the furnace, with existing Zircar cylinders, but this attempt was ultimately unsuccessful, because it still resulted in leaks. We then decided to rebuild the entire furnace. This will delay the project and require a no-cost extension. The paperwork for this is being submitted.

A second low pressure impactor has been built. This will allow us to gather more particulate samples more rapidly.

FUTURE WORK

The new furnace will be rebuilt during the coming quarter. Then experimentation on the effects of staged combustion on fine particle and NO_x emission from co-combustion of coal and MSS will commence.

An additional task will involve calibration of the new low pressure impactor against our existing one.

A Study of Fine Particulate Emissions from Combustion of Treated Pulverized Municipal Sewage Sludge

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Municipal sewage sludge (MSS) is formed during wastewater treatment, and its processing and disposal represent one of the most environmentally challenging aspects of the wastewater treating process. One disposal option currently being considered is a process involving heat treatment (to render the sludge biologically inactive) followed by dewatering, drying, pulverizing, and combustion. This research focuses on fine particle emissions from the combustion of dried, treated, MSS, cofired with either natural gas or pulverized Ohio bituminous coal as a supplemental fuel. These fuels were burned at 13 kW in a downflow laboratory combustor designed to replicate time/temperature histories and particle concentrations typical of practical combustion units yet also sufficiently well defined aerodynamically to allow elucidation of mechanisms. Size-segregated particle size distributions were obtained by isokinetic sampling followed by dilution/quenching and passage into a Berner Low-Pressure Impactor. Major and trace elements were analyzed by flame and graphite furnace atomic absorption spectroscopy. Four particle size regions were identified: furnace vapor-phase material that formed ultrafine particles either in or just before the sampling probe, submicron-sized particles formed during the combustion process, micron-sized fine particles, and larger supermicron sized bulk fly ash particles. The fuel mix appears to influence trace metal partitioning routes and the composition of fine particulate matter in the exhaust. Cofiring of MSS with coal increases the ultrafine/submicron particle emission compared to firing coal alone. This increase in ultrafine/submicron particles is most likely due to an interaction between species derived from MSS (possibly alkali metals) and those from coal (possibly sulfur and/or chlorine). Vapor-to-solid phase partitioning of arsenic and selenium is controlled by surface reaction with active surface sites during MSS combustion with either gas or coal. Co-combustion of MSS with the Ohio bituminous coal allows the arsenic and selenium to be reactively scavenged by calcium, thus changing the speciation of the trace metal emitted. Ohio bituminous coal alone contained insufficient calcium to accomplish this same scavenging effect.

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Introduction

Municipal sewage sludge (MSS) also known, and commonly referred to, as municipal biosolids waste (or simply as biosolids) is formed during wastewater treatment. Sludge is the largest byproduct of wastewater treating plants and its processing and disposal represent one of the most environmentally challenging aspects of the wastewater treating process (1). 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 (2). The advantages of this combustion process are as follows: a large reduction in disposal volume, thermal destruction of toxic organics, and recovery of the energy value of the carbon sources in the sludge (1). However, there are a number of potential disadvantages including NO_x formation (due to the high nitrogen content of most MSS, although the usual high volatile nitrogen content is easy to convert to N₂ and may be even be exploited for reburning purposes), possible boiler slagging and fouling problems (due to the high inorganic content of the feedstock), and the release of fine toxic fly ash particles into the environment (3).

A study was conducted to investigate the factors associated with fine fly ash particle emission resulting from the combustion of a dried, treated (Swiss Combi Process), and pulverized MSS at commercially relevant temperatures (i.e. combustion zone and postcombustion zone temperatures similar to those expected in commercial combustors). In addition to determining the effect of MSS addition to fuel on the overall fly ash aerosol size distributions, this study also focused on possible partitioning mechanisms for three semivolatile regulated trace elements: arsenic, selenium, and antimony. Attempts were also made to investigate zinc partitioning since recent work (4) indicates that the presence of zinc in submicron particles has a negative impact of lung cell tissue integrity. Unfortunately, background interferences rendered the zinc data suspect and so it is not presented here.

Experimental Facilities

Experimental work was conducted in a 6 m tall, 0.15 m ID vertical downflow aerosol furnace (see Figure 1). This furnace, described in detail elsewhere (5, 6), 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. The upper section of the furnace consists of a slag resistant refractory (Thermal Ceramics HPM-95) coupled to a lower section made of fibrous alumina (Zircar) cylinders. Combustion gases and solid particles travel down the furnace following laminar streamlines. Natural gas was used to heat the furnace above the autoignition temperature of coal (typically as close to the steady-state combustion conditions as possible).

Coal and/or MSS, blown off the screw tip of a volumetric screwfeeder, was premixed with the combustion air and fed to a laminar-flame burner located at the top of the furnace. The furnace, operating at a feed rate of 12.5 kW (42 800 BTU/h) and a stoichiometric ratio, SR = 1.2 (i.e. at 20% excess air) for all feed sources, was allowed to reach its self-sustaining steady-state naturally evolving temperature profile prior to sampling. When both coal and MSS were burned, the primary air was first mixed with the coal and 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 kW for each fuel feedstock).

FIGURE 1. University of Arizona downflow laboratory combustor.

Fly ash particles were withdrawn isokinetically (as determined using NO_x measurements) using a portable water-cooled, aspirated sampling probe. Dilution nitrogen was fed to the sample tip, and a vacuum pump was 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 (7). The sample was routed through a short length of 3/8 in. polyethylene tubing to a Berner-type low-pressure impactor (BLPI) (8). No attempt was made to quantify particle losses in the polyethylene tubing due to electrostatic charging. A high performance vacuum was used to reduce the pressure at the BLPI outlet to below 55.2 kPa. The BLPI utilizes low pressure and high jet velocities to allow size segregation of particles in the submicron range. The BLPI consists of 11 collection plates (stages) designed to collect particles of decreasing size as follows: 15.7, 7.33, 3.77, 1.98, 0.973, 0.535, 0.337, 0.168, 0.0926, 0.0636, and 0.0324 (50% aerodynamic cutoff diameter – in μm). The bottom stage of the BLPI acts as a sonic orifice to control the total flow rate from the sampling probe. This particulate sampling and size segregation system is described in detail elsewhere (5, 6).

Samples were withdrawn from the same physical sample port because this provided the most constant furnace residence time within the physical limitations of the experimental furnace. This sample port, located near the end of the combustor but sufficiently far upstream from the uninsulated ash trap to avoid temperature profile impacts, is representative of conditions in the late postcombustion zone of a commercial combustor. Residence times (seconds) and gas temperatures (K) at the sampling location were as follows: gas + MSS, 5.8 and 905; coal only, 5.9 and 943; and coal + MSS, 5.3 and 1015.

Proximate/ultimate/heat of combustion analysis and major inorganic elemental characterization of the Ohio bituminous coal (blend of Ohio coal seams 5, 6, and 7) used in this study were performed by Senior et al. (6). MSS was prepared and provided by the Institute for Process Engineering and Power Plant Technology at the Universitat

TABLE 1. Characterization of Coal and MSS Feedstocks

analysis	pulverized Ohio 5/6/7 blend coal (5)	pulverized and treated swiss municipal sewage sludge
heat of combustion (kJ/kg)	29900	6640
Proximate (wt %)		
moisture	2.33	5.12
volatile matter	39.19	39.20
fixed carbon	48.78	1.89
ash	9.70	53.79
Ultimate (dry wt %)		
carbon	72.77	20.76
hydrogen	5.19	3.25
nitrogen	1.40	2.34
sulfur	2.68	0.62
chlorine	0.0997	0.043
oxygen	10.41	16.29
ash	9.93	56.69
Ash Elemental Analysis (dry wt %)		
SiO_2	38	36
Al_2O_3	39	19
Fe_2O_3	13	15
CaO	2.2	8.8
MgO	1.0	4.1
Na_2O	0.92	1.8
As (ppmw)	19.0	6.7
Se (ppmw)	1.4	3.3
Sb (ppmw)	2.3	7.2

Stuttgart (Prof. Dr-Ing. K. R. G. Hein, Director) and is representative of a typical Swiss urban MSS (9) (a polluted sewage sludge, within regulated limits, from a location with medium to high fraction derived from industrial sources).

Proximate/ultimate/heat of combustion analysis of the MSS was performed at Galbraith laboratories (Knoxville, TN). The MSS ash elemental analysis was performed in-house.

Data for both fuels are shown in Table 1. Compared to the coal, the MSS contains a higher fraction of ash and alkali metals (Na, Mg, and Ca). By contrast, the coal has a higher sulfur content than does the MSS.

Arsenic, selenium, and antimony analyses of the size-segregated fly ash samples collected during the experiments were performed by graphite furnace atomic absorption spectroscopy (GFAA). Major species were analyzed by flame atomic absorption spectroscopy (AAS) or atomic emission spectroscopy (AES).

Results and Discussion

Replicate experiments were performed with (1) MSS fired with natural gas assistance ("gas + MSS"), (2) the Ohio bituminous coal without MSS ("coal only"), and (3) MSS cofired with the Ohio bituminous coal ("coal + MSS"). Sample sets that most closely duplicated the typical ash mass particle size distribution were selected for elemental analysis.

Furnace temperature profiles are shown in Figure 2. The similarity of these curves verifies that all experiments were performed at nearly identical heat loads. Most importantly, the maximum (uncorrected) measured gas temperature for each test is similar (within 60 K). 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 (flue gas with higher particle loading have higher heat capacity; for these tests $\text{MSS} + \text{gas} < \text{coal only} < \text{coal} + \text{MSS}$ particle loading).

The differential total ash particle distributions (PSDs) for all four experimental conditions are shown in Figure 3. These distributions were developed following a method originally developed by Markowski and Ensor (10) for reporting

FIGURE 2. Typical naturally evolving temperature profile at each experimental combustion condition.

FIGURE 3. Particle size distributions for MSS experiments.

impactor results. The sampling and collection system employed in this work generated a profile with four distinct regions. As shown by Davis et al. (7), the smallest diameter impactor stages collect an ultrafine vaporization mode centered at $0.1\ \mu\text{m}$ which is a young, barely coagulated aerosol generated by fresh particles nucleating either in the probe or shortly before sampling.

Slightly larger impactor stages collect submicron particles generated during the combustion process. The presence of this submicron particle size region from pulverized coal combustion has been explored in detail by previous researchers. Major refractory elements (Fe, Si, and Ca) can vaporize as suboxides in the immediate, oxygen-deficient, vicinity of the burning char particle (13–15). Upon subsequent oxidation these metals nucleate to form a submicron fume (12–13, 15–19). The fractionation of larger particles to submicron-sized particles may also contribute particles to the submicron size region (15, 19–20). The contribution from fractionation is highly fuel dependent. For coal combustion, the submicron size region is typically only 1–7% of the total fly ash mass but has a very high particle number density (13, 20–21).

A fine fragmentation mode is collected at $1\text{--}2\ \mu\text{m}$. Particles in this size range appear to exist for most coals and is the subject of another communication (11) and has recently been observed (5, 22–23) in other work. This regime is believed

to form by fragmentation of larger particles (e.g. hollow Cenospheres) during char burnout either by impaction or swelling. Micron particles are commonly observed to have submicron particles fused to the surface of larger, irregularly shaped particles (11, 24).

The ultrafine, submicron, and fine fragmentation regions are classified as fine particles less than $2.5\ \mu\text{m}$ in diameter. These particles are difficult to collect in air pollution control equipment, and so an increased fraction of fine particulates has air quality repercussions. Also, particles have been linked to respiratory distress and lung damage (12).

The coal only and the coal + MSS PSDs clearly reflect the four particle regions just described, while the gas + MSS PSD has only a weak fine fragmentation shoulder mode. The most important result from Figure 3 is that the combination of coal plus MSS produces a much greater amount of ultrafine/submicron particles than either coal or MSS (+gas assist) alone. This increase is most likely due to an interaction between species derived from MSS (possibly alkali metals) and those from coal (possibly sulfur and/or chlorine). The shape of the “coal only” and “coal + MSS” curves in the submicron region are similar. However, co-combustion of MSS with coal results in a smaller mass in the fine fragmentation region compared to coal alone. This suggests that only the coal char particles significantly participate in the fragmentation mechanism. The smaller fragmentation “shoulder” mode in the “gas + MSS” PSD supports the conclusion that MSS ash particles are not as subject to significant secondary fragmentation as has been observed for coals.

The bulk fly ash profiles for the “gas + MSS”, “coal only”, and “coal + MSS” experiments are similar. These results suggest that the pyrolysis and char burnout processes affecting mineral exclusions and large inclusions for the MSS and coal are similar.

Partitioning of Major Elements (Si, Al, Fe, and Ca).

Differential elemental distributions for four key major inorganic elements (Si, Al, Fe, and Ca) plus three important semivolatile trace elements (As, Se, Sb) are shown in Figure 4. The low concentrations of calcium and aluminum in the Ohio bituminous coal shown in Figure 4 (note: some of the concentrations in the submicron region are below the detection limit of the analytical method employed) are consistent with previous results for this coal (6). The addition of MSS to the coal greatly increases the calcium content of the submicron ash. The relatively high concentrations of Si in the submicron fume resulting from the combustion of MSS are significant. These concentrations imply that the Si reduction/vaporization mechanism is more important for MSS than for coal, possibly because of the increased presence of silicates rather than aluminosilicates, in the MSS (as inferred from Table 1).

Trace Element Partitioning Mechanisms. As shown in Figure 4, all three trace element (As, Se, and Sb) profiles have a similar shape for the “gas + MSS” results indicating similar partitioning mechanisms. The selenium profile for the “coal + MSS” experiment is different from the arsenic and antimony profiles (which are similar). In the “coal only” experiment the As, Se, and Sb profiles are all similar. These results suggest that there may be differences in the partitioning mechanisms of some of these trace elements when MSS is cofired with coal compared to coal-only combustion.

Surface Reaction Mechanisms. A substantial portion of these semivolatile trace elements is volatilized in the vicinity of burning carbon compounds and then partition back to the solid phase through surface reaction in the combustor (5, 12). For the reaction of volatilized trace elements with sites on supermicron particles (larger than the mean free path), it can be shown (11–18) that, if the process is gas film transport controlled, there will be a $1/dp^2$ dependence of the

FIGURE 4. Elemental differential distributions.

FIGURE 5. Supramicron region particle size dependence for trace elements in MSS + coal fly ash.

volatile metal concentration in the particle, while if the process is external surface reaction, or pore diffusion controlled, there will be a $1/d_p$ dependence (11). Figure 5 shows results testing these dependencies for As, Se, and Sb in the coal + MSS data. Surface reaction appears to be applicable for the As and the Se, since they exhibit a $1/d_p$ composition dependence. The antimony results are inconclusive. The gas + MSS results (not shown here) also indicate that there is a $1/d_p$ dependence for arsenic and selenium. For the "coal only" tests, no apparent particle size dependence is observed which is consistent with previous results for this coal from more extensive testing (5). The important implication is that the partitioning mechanism for As and Se is changed when cofiring MSS with coal compared to that from coal alone. MSS appears to allow As and Se to undergo surface reaction, whereas the Ohio bituminous coal alone utilized here does not.

Arsenic, selenium, and antimony volatilized during coal combustion are believed to form oxyanions, predominantly of the forms As_2O_3 , SeO_2 , and Sb_2O_3 (5, 14, 18–19). To explore the possibility of these vapor-phase oxyanions having reacted with cations, such as Ca or Fe, the differential size segregated distribution of the trace elements can be compared to the differential size segregated distributions of the cations to see if a correlations exists. In this work, polynomials were fit

FIGURE 6. Cross correlation of arsenic vs calcium and iron generated from the differential distributions shown in Figure 4 for the coal + MSS case.

both to the differential size segregated composition PSDs shown on Figure 4 and to absolute size segregated composition PSDs, with different polynomials for the submicron and supramicron regions in each case. An example of the correlations developed from the differential distributions of As vs Ca and Fe is shown in Figure 6 for the coal + MSS case.

Since these methods infer speciation, rather than measure it directly, only qualitative rather than quantitative conclusions are presented here.

Results from this methodology revealed that As and Se appeared to track calcium in the MSS + coal case but not in the coal only case. This suggests that the addition of MSS to coal can help scavenge As and Se to form calcium arsenates and selenates, which might be somewhat more environmentally benign than say the oxides. This argument is consistent with other work on this Ohio bituminous coal alone (5, 25–27) which has shown that the As and Se are scavenged by neither Ca, which is not sufficiently plentiful to provide sufficient active sites, nor by Fe, which appears to be deactivated by the high sulfur level. Hence much of these trace metals escape as the oxides for the Ohio bituminous coal without MSS. Hence addition of MSS to coal can change the speciation of the arsenic and selenium compounds emitted. Thus there is a tradeoff, environmentally, for MSS co-combustion with coal. More fine particles are generated, but these particles may decrease the emission of these semivolatile trace elements as vapor and/or ultrafine particulate matter.

For the “gas + MSS” profile, the cation (Si, Ca, and Fe) distribution curves are very similar. All three trace element distributions appear to follow these profiles reasonably closely in the supermicron region and less closely in the submicron region. Thus, no definitive conclusions can be drawn.

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