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POLYCHLORINATED DIBENZO-P-DIOXINS AND DIBENZOFURANS: REMOVAL FROM FLUE GAS AND DISTRIBUTION IN ASH/RESIDUE OF A REFUSE-DERIVED FUEL COMBUSTOR

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ABSTRACT

In early 1989, a joint Environment Canada/U.S. Environmental Protection Agency program investigated the effect of changing combustion and flue gas cleaning system variables on the performance of these systems. Using information from earlier characterization tests at the same test site (Mid-Connecticut facility in Hartford), performance data on a refuse-derived fuel combustor and its lime spray dryer absorber/fabric filter (flue gas cleaning) system were obtained under good, intermediate, and poor combustor operation and high, normal, and low sulfur dioxide (SO_2) control by the flue gas cleaning system. The independent combustion system variables included steam load, air supply rate, and its distribution. For the flue gas cleaning system, the outlet gas temperature from the spray dryer absorber served as an easily measurable surrogate for the approach to saturation temperature, while SO_2 concentration in the flue gas at the fabric filter outlet continuously represented lime stoichiometry.

Test data included: acid gas, trace organic, trace metal, and particulate matter concentrations as well as material collection for the determination of ash/residue composition and production rates. In addition, process data and refuse-derived fuel feed and ash/residue generation rates were obtained.

Correlations between combustion conditions and furnace emission of organic pollutants are presented. The removal of polychlorinated dibenzo-p-dioxins and dibenzofurans, other organics, and particulate matter from flue gas is reported. The transfer of organics to ash/residue is detailed, and the close relationship between particulate removal and organics removal is noted. The results of input/output analyses of the organics across the flue gas cleaning system are discussed.

INTRODUCTION

The Mid-Connecticut project was undertaken jointly by Environment Canada (EC) and the U.S. Environmental Protection Agency (EPA) to aid the development of regulations for municipal waste combustion in both countries. Pollutants measured in the two-phased test program included polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF), chlorobenzenes (CB), chlorophenols (CP), polychlorinated biphenyls (PCB), polycyclic aromatic hydrocarbons (PAH), trace metals, acid gases, combustion gases, and particulate matter (PM). During the first phase (characterization, January 1989), test conditions and methodologies were developed or defined.[1,2] In the second phase (performance, February-March 1989), 13 valid tests were performed while varying combustion and flue gas cleaning process conditions.[3,4] The 13 valid performance tests were grouped according to major combustion conditions (steam load and combustion efficiency) and lime spray dryer absorber (SDA)/fabric filter (FF) operating conditions (FF inlet temperature and stack sulfur dioxide (SO_2) concentrations).

In addition to flue gas sampling for organics to determine concentrations, an input/output analysis of their mass rates in the various flow streams was made. This effort entailed determining mass rates of the refuse-derived fuel (RDF) feed, dry bottom ash, grate siftings, economizer ash, FF residue, and flue gas, both uncleaned (i.e., into the SDA) and cleaned (i.e., leaving the FF and entering the stack). Samples from these streams were also analyzed for organics contents. The tests were conducted on Unit 11 (605 tonnes/day) of the Mid-Connecticut facility in Hartford [5](see Figure 1) using EPA test methods or EC/EPA-approved methods when standard methods were unavailable. Data quality assurance and control were conducted by the test contractors as well as an independent EC/EPA team.

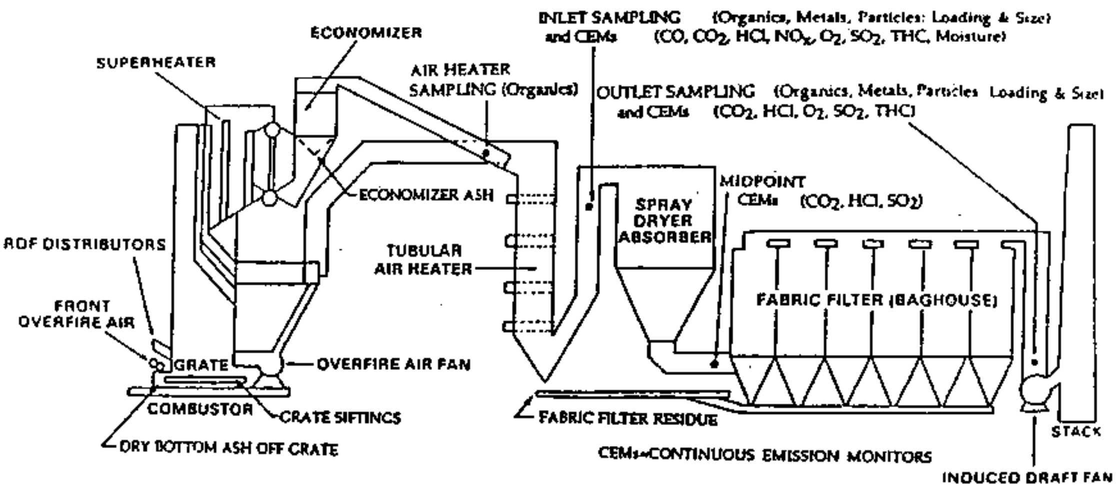

Figure 1. RDF Spreader Stoker with Spray Dryer Absorber and Fabric Filter

This paper focuses on the results of organics measurements made during the performance tests. The distribution of organics in the input and output streams is summarized, the effects of combustion efficiency on the concentration of organics contained in the combustor flue gases are discussed, the efficiency of the flue gas cleaning equipment in removing organics from the combustor flue gases is summarized, and the relative concentration of organics in the combustor and flue gas cleaning residues is presented.

RESULTS

It has been postulated that carbon monoxide (CO) concentrations in flue gas can be used as a measure to characterize the efficiency of combustion in destroying the organics in flue gases. During the Mid-Connecticut performance tests, combustion operating conditions were varied to provide test average flue gas concentrations of CO (measured at the SDA inlet) ranging from less than 100 to more than 900 ppm. To facilitate the evaluation of test results presented in this paper, combustion conditions are defined as "good" when the test average flue gas concentration of CO is 100 ppm or less, "intermediate" when CO concentrations are between 100 and 250 ppm, and "poor" when they are greater than 250 ppm.

The distribution of organic compounds (based on the average of all tests) in the RDF, combustor ash (dry bottom ash plus economizer ash), flyash (fabric filter residue), and stack gas emissions is shown in Figure 2. Comparison of the organics in the RDF feed with all organics leaving the facility in the combustor ash, collected flyash and stack emissions shows a net reduction in all classes of all measured organic compounds except PCDF. Comparison of the total measured organics in the RDF feed with the sum of the organics in all output streams indicates that combustion results in a net reduction in total organics which exceeds 90%.

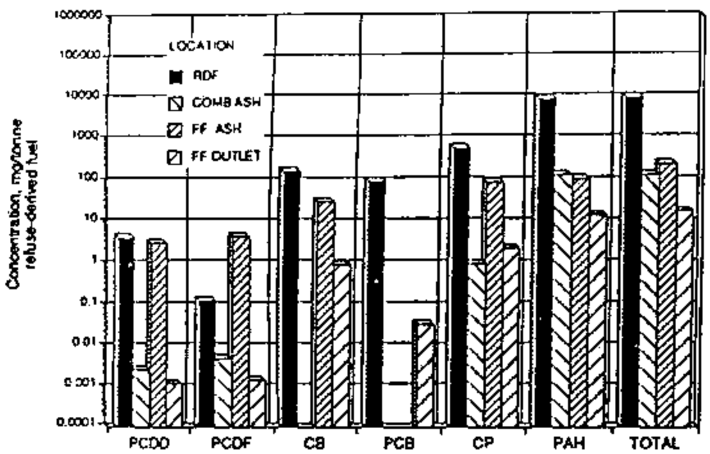

Figure 2: Distribution of Organics

Figures 3,4, and 5 indicate the concentrations of organics leaving the combustor; i.e., organics at the SDA inlet. PCDD/PCDF and total organics in the combustor flue gas tend to increase as combustion becomes less efficient (Figures 3 and 4). Regression analyses using all major parameters characterizing combustion conditions indicated that either CO or THC is the best single parameter for predicting flue gas concentrations and hence destruction of PCDD, PCDF, CB, CP, and PAH. Flue gas concentrations of these organic compounds increase with deteriorating combustion conditions as characterized by CO concentrations (Figure 5). While the PCB did not follow this trend, this apparent anomaly is probably associated with the low level and high variability of these concentrations. There was no significant correlation between the concentration of organics in flue gas and steam load, flyash concentration, or flyash carbon content.

The removal of organics in the flue gas cleaning system exceeded 95% for all measured semivolatile organic compounds and over 99% for PCDD and PCDF. As seen in Figure 6, concentrations of CB and CP in the stack gas decreased as combustion improved. Stack concentrations of PCDD, PCDF, PCB and PAH varied only slightly and inconsistently with combustion conditions. Stack concentrations of semivolatile organics were lower for intermediate and good combustion conditions than for poor combustion conditions. There was no apparent correlation between flue gas cleaning operating variables and stack concentrations of organics. This lack of correlation was probably due to the high collection efficiency of organics noted for all tests.

Concentrations of CB and PCB in the combustor bottom ash and economizer ash were below the detection limits for all samples analyzed (Figure 7). PCDD and PCDF were found at detectable levels in only 2 of 12 bottom ash samples analyzed. Both were from tests with good combustion conditions. PCDD was detected at low concentrations in 4 of 12 samples, and PCDF was detected at significantly higher concentrations in 10 of 12 samples. The amounts of CP and PAH in the two combustor ash streams were 100 to 1000 times higher than the amounts of PCDD and PCDF. The higher average concentrations of PCDD and PCDF noted during the good combustion tests result primarily from the concentration of these organics found in the two bottom ash samples containing detectable quantities of PCDD and PCDF.

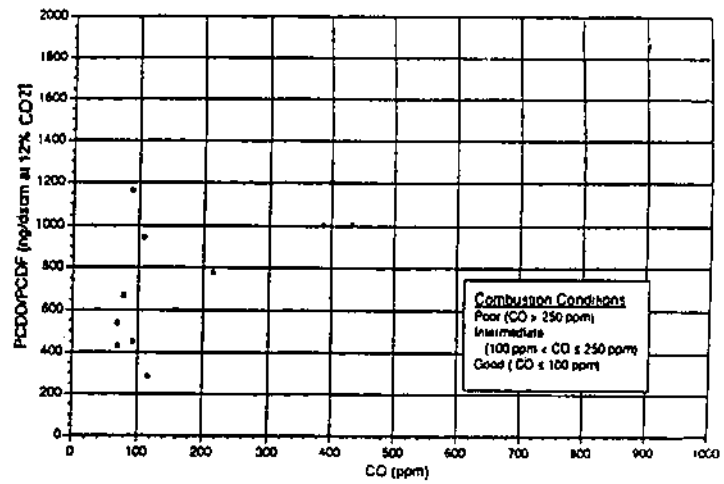

Figure 3. PCDD/PCDF vs. CO in Flue Gas at SDA Inlet

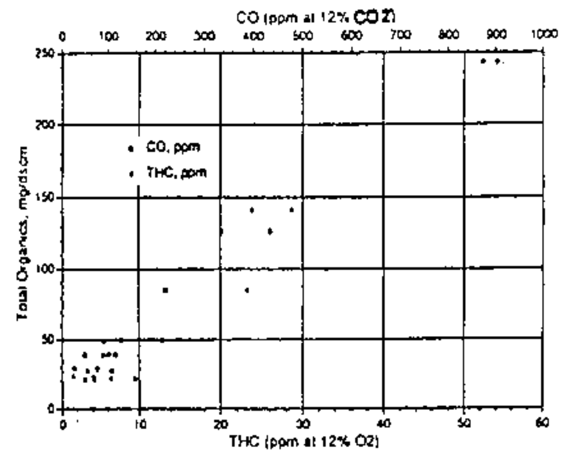

Figure 4. Total Organics vs. THC and CO

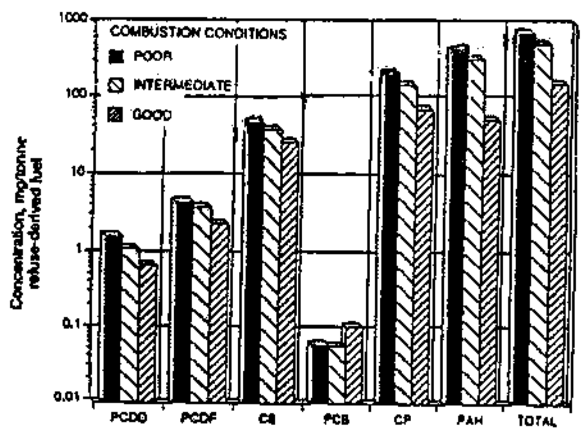

Figure 5. Organics Concentrations at SDA Inlet

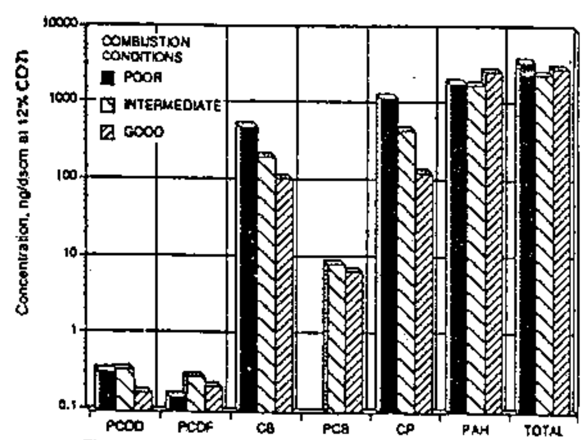

Figure 6. Organics Concentrations at Fabric Filter Outlet

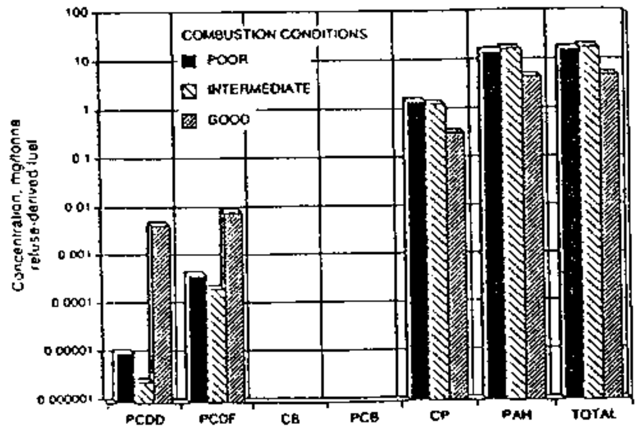

Figure 7. Organics Concentrations in Combustor Ash

Figure 8 shows that the major fraction of all semivolatile organic compounds (except PCB) leaving the combustor was in the FF ash. The relative amounts of all organic compounds in the FF ash increased with worsening combustion conditions (increasing CO).

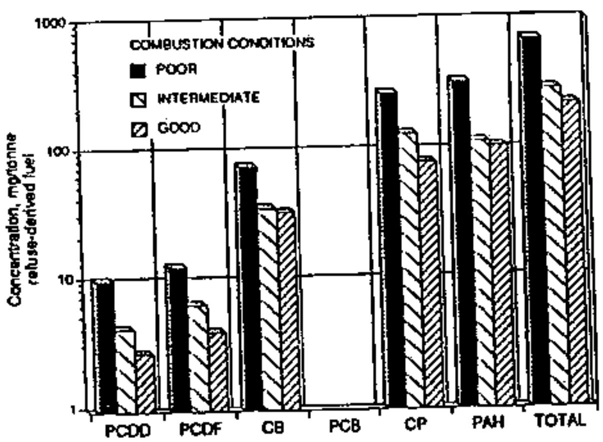

Figure 8. Organics Concentrations in Fabric Filter Ash

CONCLUSIONS

•The concentrations of semivolatile organic compounds (PCDD, PCDF, CB, CP, PAH, and PCB) in the furnace flue gas depended on combustion conditions. Net destruction efficiencies for these organics ranged from approximately 90% for poor combustion to 96% for good combustion.

•Average CO or THC concentration in the flue gas was the best single indicator for the furnace destruction of organics.

•Average concentrations of PCDD, PCDF, CB, CP, and PAH in the flue gas tended to increase as combustion deteriorated (i.e., CO increased).

•The concentrations of semivolatile organics in flue gas in decreasing order were: PAH, CP, CB, PCDF, PCDD, and PCB.

•PCDD/PCDF removals in the SDA/FF system exceeded 99.9%, while PM removal exceeded 99%. Removals of all organics classes (PCDD, PCDF, CP, CB, PAH, and PCB) were over 95%.

•Organic compound removals in the SDA/FF system appeared to be independent of flue gas cleaning system operation, which generally kept hydrogen chloride (HCl) removals above 90%. Total organic emissions in flue gas ranged from 300 mg/hr for good combustion to 1440 mg/hr for poor combustion.

•PAH and CP were the major trace organics in the RDF. Both were also the main components in the stack gas but with values substantially below those in the RDF.

•The major fractions of all organics leaving the combustor (PCDD, PCDF, CB, CP, and PAH) were collected and discharged with the FF residue. PCB was below the detection limit in all the ash/residue streams.

•Generally, the organics concentrations in the combustor ash (bottom ash plus economizer ash) were considerably less than in the FF residue. With the exceptions of CP and PAH in bottom ash, the relative amounts of organics in the combustor ash were lower than in the stack gas.

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