

Endocrine disrupting chemical emissions from combustion sources: diesel particulate emissions and domestic waste open burn emissions

Sukh Sidhu^{a,*}, Brian Gullett^b, Richard Striebich^a, Joy Klosterman^a,
Jesse Contreras^a, Michael DeVito^c

^a*Environmental Engineering, University of Dayton, 300 College Park, Dayton, OH 45469, USA*

^b*National Risk Management Research Laboratory (E305-01), US Environmental Protection Agency, Research Triangle Park, NC 27711, USA*

^c*National Health and Environmental Effects Research Laboratory (MD-74), US Environmental Protection Agency, Research Triangle Park, NC 27711, USA*

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Abstract

Emissions of endocrine disrupting chemicals (EDCs) from combustion sources are poorly characterized due to the large number of compounds present in the emissions, the complexity of the analytical separations required, and the uncertainty regarding identification of chemicals with endocrine effects. In this work, multidimensional gas chromatographic-mass spectrometry (MDGC-MS) was used to characterize emissions from both controlled (diesel engine) and uncontrolled (open burning of domestic waste) combustion sources. The results of this study suggest that, by using MDGC-MS, one can resolve a much greater percentage of the chromatogram and identify about 84% of these resolved compounds. This increase in resolution helped to identify and quantify various classes of polycyclic aromatic hydrocarbons (PAHs) in the combustion emissions that had not been identified previously. Significant emissions (when compared to industrial sources) of known EDCs, dioctyl phthalate (over $\sim 2,500,000$ kg year⁻¹) and bisphenol A (over $\sim 75,000$ kg year⁻¹) were estimated from uncontrolled domestic waste burning. Emissions of several suspected EDCs (oxygenated PAHs) were observed in both diesel soot and the uncontrolled domestic waste burn samples. The emission rates of known and suspected EDCs estimated in this study suggest that combustion emissions need to be characterized for EDCs to further assess its importance as a source of EDC exposure.

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1. Introduction

In recent years, several studies have been conducted to investigate the potential threat of endocrine disrupting

chemicals (EDCs) to wildlife and humans (Colborn and Clement, 1992; Colborn et al., 1993; Guillette et al., 1995). It is now known that several natural and man-made EDCs act as mimics or antagonists of endogenous hormones (Crews et al., 1995; Cummings and Metcalf, 1995; Soto et al., 1995). These concerns have led to the initiation of a significant number of in vitro and in vivo studies of the toxicology of EDCs, including the

*Corresponding author. Tel.: +1 937 229 3605; fax: +1 937 229 2503.

E-mail address: sidhu@udri.udayton.edu (S. Sidhu).

development of a comprehensive screening procedure to detect potential EDCs (Arnolds et al., 1996; Routledge and Sumpter, 1996; Zacharewski, 1997). Despite these investigations, the extent to which EDCs can negatively impact human and animal health is a subject of debate (Stone, 1994). Most of these EDC studies have thus far focused on commercial chemicals that may be “widely” distributed in the environment (Crews et al., 1995; Cummings and Metcalf, 1995; Harris et al., 1997; Soto et al., 1994, 1995) primarily via aquatic media. Little attention has been directed toward identifying or characterizing other potentially important sources of EDC emissions.

Given the large variety of complex emissions from combustion processes, it is reasonable to expect presence of EDCs as combustion byproducts. For the past two decades, numerous studies have been conducted to investigate the origin and formation mechanism of toxic combustion by-products (Dellinger et al., 1991; Fiedler, 1993; Gullett et al., 1994; Konduri and Altwicker, 1994; Lenoir et al., 2001; Oppelt, 1986; Sidhu et al., 1995). These studies have shown that known EDCs such as polychlorinated dibenzodioxins and furans (PCDDs/Fs), alkylphenols, phthalates, styrenes, polychlorinated biphenyls (PCBs), and polybrominated biphenyls (PBBs) are present in emissions from various combustion processes (Fiedler, 1993; Oppelt, 1986). Along with these known EDCs, several hundred other chemicals are also present in combustion emissions (Ryan et al., 1996). The complexity of the combustion emissions has resulted in the identification of only a small percentage of chemicals emitted, even though considerable effort has been expended to characterize emissions from some combustion sources (Rogge et al., 1993; Schauer et al., 1999).

An examination of Fig. 1, a high-resolution chromatogram of the semi-volatile (C_6 to $\sim C_{16}$) fraction of the emissions collected from a hazardous waste incinerator shows that $\sim 1\%$ of chemicals can be identified because only 10–15% of the chemicals are chromatographically resolvable (Ryan et al., 1996). The exact pattern of the chromatogram may vary among combustion sources, but the general pattern, complexity, and lack of resolution are similar. Initial results suggest that, by using multidimensional gas chromatographic-mass spectrometry (MDGC-MS), a greater percentage of the chromatogram can be resolved and identified (Rubey et al., 1999; Striebich et al., 2002). The majority of the chemicals resolved and identified with MDGC-MS were semi-polar oxygenates that have many structural similarities with known EDCs. The existence of these chemicals in combustion emissions was previously unknown; however, an understanding of combustion processes suggests that all thermal and combustion processes may possess the appropriate conditions and reactants to produce these chemicals (Lenoir et al.,

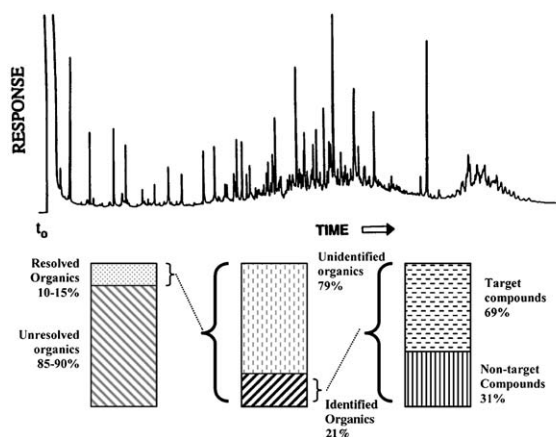


Fig. 1. Chromatogram of the semi-volatile fraction of the effluent from a hazardous waste incinerator collected with a MM-5 sampling train. This sample was subjected to methylene chloride extraction and HRGC-MS analysis using a $50\text{ m} \times 0.2\text{ mm} \times 0.3\text{ }\mu\text{m}$ dimethylsiloxane open tubular column.

2001). In fact, these chemicals are probably formed in the post-flame and cool zone of combustors by a surface-catalyzed mechanism similar to that responsible for the formation of PCDDs/Fs in combustion systems (Lenoir et al., 1998; Sidhu, 1999). Thus, these oxygenated and halogenated EDCs may be emitted from essentially any combustion or thermal process.

For this study, samples were collected and analyzed from both controlled and uncontrolled combustion sources. For controlled combustion emissions, diesel particulate samples were selected because of their known environmental and health impact (Harrison et al., 1997; Venkataraman and Friedlander, 1994). In a recent study, Taneda et al. (2002) showed that both estrogenic and antiestrogenic compounds exist in diesel exhaust particles. Also, Watanabe and Oonuki (1999) recently reported that exposure to diesel exhaust can increase the serum testosterone and estradiol levels. Diesel exhaust has also been found to affect the regulation of testicular function in male rats (Tsukue et al., 2001). The results of these studies indicate the need to identify compounds on diesel particulates that are causing estrogenic and antiestrogenic activity. Mori et al. (2002) identified 4,6-dimethyldibenzothiophene (isolated in the neutral portion of a hexane extract) as one of the compounds responsible for estrogenic activity. It seems likely that more than one class of compounds is responsible for the endocrine disrupting behavior of diesel particles, since hexane, benzene, dichloromethane, and methanol extracts of diesel particles showed estrogenic and antiestrogenic activity (Mori et al., 2002; Taneda et al., 2002; Tsukue et al., 2001; Watanabe and Oonuki, 1999). However, other potential classes of EDC compounds in diesel particle extract have yet to be identified.

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