USF Tampa Graduate Theses and Dissertations

USF Graduate Theses and Dissertations

11-18-2005

Occupational Exposure to Ultrafine Particles and Polycyclic Aromatic Hydrocarbons from Candle Emissions

David J. Silver
University of South Florida

Follow this and additional works at: https://digitalcommons.usf.edu/etd

Part of the American Studies Commons

Occupational Exposure to Ultrafine Particles and Polycyclic Aromatic

Hydrocarbons from Candle Emissions

David J. Silver, C.I.H., M.Sc.

A dissertation submitted in partial fulfillment
of the requirements for the degree of
Doctor of Philosophy
Department of Environmental & Occupational Health
College of Public Health
University of South Florida

Major Professor: Raymond D. Harbison, Ph.D.
Thomas Bernard, Ph.D.
Yehia Hammad, Sc.D.
Joette Giovinco, M.D.

Date of Approval:
November 18, 2005

Keywords: Condensation Nuclei, Nanometer, Particle Distribution, Lifetime
Average Daily Dose, Optical Particle Counter

DEDICATION

I dedicate this Dissertation to my wife, Lydia and my son Joshua. They provide support, encouragement and kindness when I need it most. I want to inspire my son, Joshua to live up to his full potential and find true happiness in whatever direction he chooses.

ACKNOWLEDGMENTS

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

OCCUPATIONAL EXPOSURE TO ULTRAFINE PARTICLES AND
POLYCYCLIC AROMATIC HYDROCARBONS FROM CANDLE EMISSIONS

David J. Silver, C.I.H., M.Sc.

ABSTRACT

Ultrafine particles (UFPs) are present in the ambient atmosphere and are generated from atmospheric gases, pollution sources and combustion. Candles emit carbonaceous soot particles similar to UFPs present in the ambient atmosphere. With the exception of lead, airborne concentrations of candle emissions have not been shown capable of causing cancer or cardiopulmonary disease during normal use. The purpose of this research is to determine the occupational risk associated with candle emissions.

Candle studies employ chambers to measure candle emission exposures and assess public health risk. Chambers used in previous studies did not match normal room conditions. They were affected by turbulence and high temperature, which affected particle distribution and constituent concentrations, while making it difficult to extrapolate the results.

The chamber designed for this study sought to avoid the problems noted above. This study also employed a room constructed to closely simulate a normal work environment. Candle suppliers and users were surveyed to determine occupational candle use and settings. Scented, unscented, and church candles were measured in both ventilated and unventilated environments. A condensation nuclei counter was used to measure UFPs from candle emissions.

Relative to previous chamber designs, results indicated a reduction in candle soot generation, no significant airborne concentrations of metals, and airborne concentrations of polycyclic aromatic hydrocarbons (PAHs), below occupational limits. Scented candles generated more soot than unscented candles.

UFP studies have demonstrated only weak associations between ambient UFP exposures and cardiopulmonary disease. However, ambient UFP exposures were used as a benchmark for candle soot exposures. The lifetime average daily dose (LADD) was calculated from the candle soot measurement data and ambient UFP data. Candle soot generated inside the test room ranged from 5.73×10^9 to 1.86×10^{11} number of candle soot particles inhaled daily

compared to the 3.25×10^{11} to 2.45×10^{12} soot particles inhaled in the ambient environment. The calculated candle soot dose was nearly an order of magnitude less than the calculated ambient dose.   The conclusion is that candle emissions do not pose a health risk under normal occupational use.

CHAPTER ONE

INTRODUCTION

Ultrafine particles (UFPs) are present in the ambient atmosphere and are generated from atmospheric gases, pollution sources and combustion. Candles emit carbonaceous soot particles similar to UFPs present in the ambient atmosphere. With the exception of lead, airborne concentrations of candle emissions have not been shown capable of causing cancer or cardiopulmonary disease during normal use. The purpose of this research is to determine the occupational risk associated with candle emissions.

Public and occupational health concerns about the health risk associated with candle emissions are a result of several studies linking candle emissions to cancer risks (Lau et al., 1997; Fine et al; 1999, Krause, 1999). Candles are a two billion dollar a year business in the United States, so the public health impact of producing hazardous emissions from candle emissions could be enormous. The economic impact and worldwide use necessitate an improved risk assessment methodology for the evaluation of candle soot exposure.

Several studies have found harmful emissions from candles. Public anxiety over candle emissions has been ignited by claims of polycyclic aromatic hydrocarbon (PAH) exposure (Lau et al., 1997), dioxin exposure (Malisch, 1994; Lau et al., 1997; Schwind et al., 1994), and neurotoxicity from lead wicks (van Alphen, 1999; Krause, 1999; Sobel et al., 2000). Krause's 1999 "scented candles study" reports that candle soot has the same morphological features and PAH content as diesel soot and should be considered equal in carcinogenicity. Krause also claims that scented candles, having a greater soot generation rate than unscented candles, also pose greater carcinogenic risk.

As it is later shown, these studies were flawed; still, the reported findings depressed market demand for candles, particularly for scented candles. Candle studies from the United States, Germany, Great Britain, and Belgium reported exaggerated candle emissions and that implied significant risk of cancer and cardiopulmonary disease. The National Candle Association (NCA) was prompted to publicly advise end users to trim wicks and provide adequate ventilation.

Reported candle emissions exposure data derived from chamber testing are a result of atypical candle use and atypical production of PAHs, dioxins, and benzene. Significant cancer risks associated with candle emissions, though the

main trigger of public concern, have not been demonstrated using relevant occupational or residential conditions. More broadly, there has been a lack of reliable research on occupational health risks associated with exposure to candle emissions.

To put the previous studies in proper perspective, this study seeks to determine risks associated with candle emissions in occupational settings. To characterize candle soot's toxicology and health effects, the literature was reviewed. Carcinogenic candle emission constituents were measured in the improved chamber and in the test room. The validity of diesel soot carcinogenicity being used as a surrogate for candle soot was explored. Test room measurement of ultrafine particle concentrations provided ultrafine particle counts that previous chamber and test room mass concentration studies lacked. Size-specific particle count data generated from candle emissions in a test room was compared to the size distribution cited in a number of ambient ultrafine particle studies. The test room ultrafine particle measurement of candle soot provided more accurate exposure and dose data than do models extrapolated from chamber data.

Main Research Goals

This study seeks to accurately determine the occupational health risk from exposure to candle soot by accomplishing the following goals:

Improve the chamber design to produce data that are more reliable. Chamber factors that are known to affect candle soot generation include turbulence.   Turbulence is a fluid motion in which velocity, pressure, and other flow quantities fluctuate irregularly in time and space. Turbulence was addressed in the improved chamber, which was designed to simulate normal room conditions.

Test the validity of previous chamber studies regarding the differences between soot emission rates of scented and unscented candles.

Provide a more accurate assessment of candle soot exposure. Since candle soot has many of the same characteristics as soot found in the natural ambient environment and from combustion sources, ambient ultrafine particles provide the only reasonable benchmark for candle soot exposure. Ultrafine particle studies examining the link between exposure and cardiopulmonary disease have either resulted in weak associations or been inconclusive.

Determine the occupational health risk produced by candle emission exposure. Occupational health risks from ultrafine particle exposure to a workplace specific number of candles were calculated using a lifetime average daily dose (LADD) model.   LADD model inputs included EPA human exposure factors, test room data, ventilation data, and occupation-specific candle type and

2

number of candles. Occupational LADD of candle soot was compared to the ambient environment LADD for ultrafine particles.

Study Limitations

Measurements were restricted to a single size range of particles detected by a condensation nuclei counter. Test room specifications and candle use information were based on a single survey. Comparative data was restricted to the Clearwater, Florida area. Human factor data was for the average working man or woman.

The study restricts the ultrafine particle size measurement to the particle size range of a condensation nuclei counter, 20 nanometers - 1 micrometer. Although the CNC does not provide particle size distribution, optical particle counters were used to classify size from 0.1 to 10 micrometers.

Particle formation component studies demonstrate the need for hazard analysis of aging particles. Health risks associated with particle age distribution and age specific intake in relation to distance from the candle source is not part of our study. We assume that the toxicology is the same for all proximities of persons to candles.

The test room specifications were based on observations, survey data and assumptions. Candle use data and occupation specific information were limited to our survey rather than the extensive national candle marketing report.   Test room furnishings (particle sinks) were selected from survey information and observations in the Clearwater, Florida area.   The test room was single, isolated, with no air exchange between other rooms. The unventilated environment was completely sealed to prevent infiltration and exfiltration. The ventilated environment was similar with the exception of not sealing the door and window.

The test room was assumed a well-mixed room.   The smaller a particle is, the more it behaves like an ideal gas. An ideal gas will fill a volume entirely, whereas particles are subject to thermal effects, cloud formation, boundary layers, unequal spatial distribution and settling. When sampling particles, variation in the quantity sampled can occur. No corrections were made for factors affecting particle concentration. Ultrafine particles such as soot approach molecular size, so the variation in particle measurement is expected to be low. The condensation particle size counter measured within the size range of the instrument. Particles smaller than 20 nanometers were outside the range of the instrument. Proximity was always three feet from the candles; however, the positioning of the counter was randomly varied. The variation in placement minimized spatial variation bias to one side of the room.

Measurements of ambient ultrafine particles were conducted in a single urban location, Clearwater, Florida.   The location reflected particle apportionment in a small urban location, and is not entirely representative of other locations. The single location data was supplemented with particle counts cited in ultrafine particle studies.

An average working man or woman was assumed.   Respiration rates and lifespan were based on the EPA human factors study. Workers in various occupations differ by age, sex and health status. We did not consider the effects of age, sex or health status in this study.

Conclusive comparisons to referent doses of ultrafine particles were not provided in our risk estimation. Morbidity studies provided only weak associations, poor specificity, low sensitivity and were riddled with confounders. Lacking conclusive evidence of specific health effects associated with ultrafine particles, a comparative risk analysis was done. Comparative risk analysis can be quantitative, qualitative or both. Typically, initial analyses are quantitative and focus on selected issue areas. However, the final rankings are invariably qualitative. Judgments about the priority assigned to candle emission associated health effects reflect a variety of qualitative factors, such as the degree of public concern, ambient exposure acceptability and acceptable risk. Final rankings are categorized as low, medium or high risk and by definition involve qualitative judgments about the relative importance of different candle emission issues. Candle soot ultrafine particles are carbonaceous core particles similar to those found in the ambient atmosphere. Combustion particles or soot found in the atmosphere is the primary referent standard used in this study.

Candle Materials

Candles are made of two components, the solid fuel source and the wick. The fuel is a soft solid substance at room temperature and melts when heated. The candle wax frequently has dye and fragrance additives. The candle material is flame combusted and generates thermal breakdown products.   Table 1 lists common candle materials.

Table 1

Materials and Chemicals used in Candle Construction

Candle Materials
Candle body
Paraffin, Stearine, Gelled mineral oil, Beeswax, Tallow
Wick
Paper fibers, Lead (banned), Zinc, Tin
Colorants
Organic synthetic, Aniline
Fragrances
Oils, Extracts

Candle wax consists of derivatives from petroleum (paraffin), animals (tallow) or insects (beeswax). A common candle material is refined paraffin, a mixture of n-paraffin, isoparaffins, and cycloparaffins (naphthenes). Paraffin contains crude oil, which mostly consists of crystalline hydrocarbons, typically C_{22}–C_{28} hydrocarbons. Paraffin's chemical and physical properties depend on the crude oil and the refining process. The melting point for refined paraffin is typically between 52 and 56°C. In tea lights, paraffin has a low carbon number (C_{18}) and low melting point. Paraffin with a higher melting point than the internal wax covers the external layers of candles to ensure the candles do not bend when exposed to sunlight. Candles typically melt at 70 to 75°C. Some paraffin waxes contain residues of light aliphatic and aromatic hydrocarbons. Opalescent or white candles are pure compounds or mixtures of stearine. Stearine is a mixture of stearic acid (C_{18} aliphatic acid) and palmitic acid (C_{16} aliphatic acid), with a melting point typically between 60 and 62°C. Gelled mineral oil (gel wax) consists of crude oil and mostly undefined hydrocarbons and is in a semi-liquid gelled state. Gelled candles contain light aliphatic and aromatic hydrocarbons. According to the NCA, candle wax does not contain lead.

Wicks are generally made from cotton, and vary in thickness and weave. The wick controls the melting, evaporation, and burning of the candle. Capillary action transports the liquid wax from the melting area to the burning zone. The weaving of waxes and paper fibers into the wick, keeps the wick stiff and upright. In some candles, the wick stiffener is embedded metal. The types of metals released from a candle depend on the wick composition, typically lead, zinc, or tin. A longer wick produces a larger flame, brighter candle and increased soot generation rate. Trimmed wicks generate less soot than untrimmed wicks (NCA, Candle Safety Tips, 2000). Lead and other metals are typically found in the wick, not in the wax.

Properties and Emissions of Candle Materials

The thermal breakdown of candle wax and wick generates airborne polycyclic aromatic hydrocarbons (PAHs), organic compounds, and dioxins (Guo et al., 1999). The organic characteristics of candle emissions vary with the type of paraffin, stearine, or mineral oil burned. Some of these organic hydrocarbons are OSHA regulated, and safe exposure limits might be exceeded with an atypically large number of candles burned. Several hundred candles burning in a small room will produce an atypical concentration of soot, PAHs and organic compounds. The heat from the candles and the carbon monoxide generated would be more of an immediate health hazard than the concentrations of organic compounds.   This type of candle use is atypical of residential and occupational use.

Some candles contain lead. Lead in candles has been shown to be harmful. Of the U.S.-made candles containing metal wicks, these are typically zinc or tin. The United States National Candle Association (NCA) membership manufactures 90 percent of all candles used in the U.S. and do not employ lead wicks, and most candle wicks are made of 100% cotton or cotton-paper combinations.   The NCA membership voluntarily agreed to cease production of lead-containing candles in 1974, once it was shown that burning lead-wick candles result in increased lead concentrations in indoor air (Sobel et al., 2000). The economic impact from public concern over lead in candles has led to decreased candle sales and increased costs for informing the public about the lead candle ban. Despite the voluntary ban, lead wick candles can still be found on the market. In a recent consumer survey, 9 of 285 (3%) candles had 33- to 85-weight percentage lead in the wicks (Public Citizen's Health Research Group, 2000). Some candle manufacturers dodge import restrictions or are uncontrolled cottage industries.   Uncontrolled hazardous substances in candles demonstrate the need for public information regarding candle materials.

Colorants are organic synthetic substances, aniline colorants, or organic pigments that are soluble in wax. Some candles have a dyed external layer of micro paraffin, which has a higher melting point than the internal wax, to reduce bending of the candle when exposed to sunlight. The over dipping layer includes decorative varnishes or lacquers with colorants. Purple candles have been investigated for dioxin emissions (Malisch, 1994). Lau et al. (1997) investigated candle emissions for various organics that included dioxin emissions.

Scents are added to candles primarily for aromatherapy and relaxation. Spas often use fragrant aromatherapy candles containing essential oils and extracts from plants dissolved in organic solvents. Restaurants, however, are more likely to use unscented candles because of patron sensitivities. Scented

candles are claimed to generate more soot than unscented candles (Krause, 1999).

Hazards Posed from Candle Use

Candles used in church vigils and around icons are purchased by individuals and may be dyed, scented, or unscented; however, unscented dyed candles are burned more often. The greater use of scented candles in spas may justify exposure analysis for dioxins and furans in scented aromatherapy candle emissions.

Candle materials, candle type, number, and use have an effect on concentration and type of airborne hazardous emission constituents   Potential occupational risks of these hazardous constituents include nervous system disease associated with lead, cancer with PAH laden soot and cardiopulmonary disease with ultrafine particles. In previous candle studies, untrimmed wicks and scented candles were associated with increased soot production (Krause, 1999). Although in comparison to other combustion sources, there are relatively few emissions studies for candles, information obtained has included experimental methods and emission data.   Current advancements in analytical technology have permitted researchers to provide more sensitive measurements of trace organics and metals in the air. Dioxins, benzene, lead, zinc, and PAHs have been measured in candle emissions inside chambers and rooms in many of these studies. Candle emissions were reported to contain lead and zinc emissions from metal wicked candles (van Alphen, 1999). Candle colorants and fragrances have been associated with the release of dioxins and furans (Lau et al., 1997; Malisch, 1994). Table 2 lists common candle emission constituents identified in previous candles studies.

Table 2

Candle emission constituents identified in previous studies

Common Candle Emission Constituents	
Dibutyl phthalate	Styrene
Diethyl phthalate	Formaldehyde
Acetaldehyde	Acrolein
Benzaldehyde	MEK
Naphthalene	Ethanol
Polycyclic Aromatic Hydrocarbons	Soot
Dioxin	Lead
Benzene	Zinc
Toluene	Tin
Ethyl benzene	Other trace metals

Lead Emission Studies

Studies on the effects of lead in children have demonstrated a relationship between exposure to lead and a variety of adverse health effects. These health effects include impaired mental and physical development, decreased heme biosynthesis, elevated hearing threshold, and decreased serum levels of Vitamin D. The neurotoxicity of lead is of particular concern, because evidence from prospective longitudinal studies has shown that neurobehavioral effects, such as impaired academic performance and deficits in motor skills, may persist even after blood lead levels have returned to normal (Needleman, 1990). Although no threshold level for these effects has been established, the available evidence suggests that lead toxicity may occur at blood lead levels of 10-15 µg/dl or possibly lower (Agency for Toxic Substances and Disease Registry (ATSDR), 1988).

Van Alphen (1999) identified hazardous lead emissions from lead wicked candles as a health risk to children. Airborne concentrations of lead were measured from candles burning inside a chamber. Modeling was used to extrapolate lead dose based on airborne concentrations expected in a normal room from candle emissions. Children's lead intake was modeled from exposures using a biokinetic model. Van Alphen reported that blood lead levels of children have the potential to rise above 10 µg/dL during candle use when typical background exposures are present. Van Alphen measured lead from chamber-generated mass concentrations of candle soot. Although van Alphen's chamber study measured lead from mass concentration samples and did not elucidate the particle size distribution, deposited soot on surfaces can still pose a risk to children due to hand-to-mouth activity.

Wasson et al. (2002) tested the emissions of lead candles purchased in the United States. The wicks in the candles Wasson tested were 39-74% lead (the remainder was fabric or paper) and the lead cores, approximately 100% lead, had linear wick densities of 13-27 mg per centimeter of wick material. Mass concentration of soot was collected and analyzed from burning candles in a chamber. Individual candles emitted airborne lead that ranged from 100 to 1700 µg/h. Exposure modeling was used to estimate room concentrations and inhalation exposure of children. Wasson suggested that airborne concentrations have the potential to exceed EPA 1.5 $\mu g/m^3$ and OSHA 50 $\mu g/m^3$ guidelines.

Nriagu et al. (2000) assessed the amount of lead released from 14 different brands of candles with metal-core wicks, sold in Michigan. The six candles made in the United States released 1.1-66.0 µg of lead per hour, the five Mexican candles released 0.5-5.9 µg per hour, and the four Chinese candles released 1.8-327.0 µg per hour. The concentration of lead that would accumulate in a closed bedroom measuring about 12 feet by 15 feet by 10 feet, or 50 m^3, after burning each candle for 2 hours was estimated at 0.04-13.1 $\mu g/m^3$, in some

cases far exceeding the U.S. Environmental Protection Agency's (EPA) ambient air quality standard of 1.5 µg/m^3. This EPA standard is based on a 24 hour, 365 day per year residential exposure, whereas, the OSHA PEL of 50 µg/m^3 is based on workplace exposures in a 40 hour week.

<center>Organic Hydrocarbon Emission Studies</center>

Studies reporting candle emissions including acetaldehyde, formaldehyde, dioxins, PAHs and organic hydrocarbons did not demonstrate airborne concentrations above OSHA permissible exposure limits or EPA guidelines. Table 3 provides a summary of airborne concentrations of candle emission constituents as reported from candle studies.

Table 3

Reported Concentrations of Chemical Compounds Emitted from Candles

Author / Date	Study	Findings	Comparison Criteria
Lau (1997)	Acetaldehyde	0.834 µg /m^3	[a]OSHA PEL: 360 mg/m^3 RfC: 9 µg /m^3 Cancer: 0.5 µg /m^3
Lau (1997)	Formaldehyde	0.190 µg /m^3	[b]ACGIH TLV: .3 ppm (.37 mg/m^3) ceiling OSHA PEL: .75 ppm (.9 mg/m^3) [c]TWA; 2 ppm (2.5mg/m^3) 15 min [d]STEL
Lau (1997)	[e]PCDD/PCDF	0.635 µg/m^3	RfD: 13 ng/kg bw
Schwind (1994)	Formaldehyde	17 µg /m^3	Same as above
Schwind (1994)	PCDD/PCDF	0.38 [f]pg I-TEQ/m^3	RfD: 13 ng/kg bw
Lau (1997)	[g]BaP	0.002 /m^3	PEL: 200 /m^3
Schwind (1994)	Naphthalene	0.04 /m^3	PEL 50 mg/m^3
Malisch (1994)	Dioxins	0.5 ng I-TEQ/kg	RfD: 13 ng/kg

[a]OSHA PEL: Occupational Safety & Health Administration Permissible Exposure Limit
[b]ACGIH TLV: American Conference of Governmental Industrial Hygienists Threshold Limit Value
[c]TWA: Time Weighted Average
[d]STEL: Short Term Exposure Limit
[e]PCDD/PCDF: polychlorinated dibenzo-p-dioxin (PCDD) and polychlorinated dibenzofuran (PCDF)
[f]pg I-TEQ/m^3: picograms of dioxin toxic equivalent value
[g]BaP: Benzo[a]pyrene

Polycyclic Aromatic Hydrocarbons Candle Emission Studies

Polycyclic aromatic hydrocarbons (PAHs) have long been recognized as a component of soot generation.   Soot is produced from combustion of fuel sources such as gasoline in cars, during cooking and while burning candles. Some of the PAHs in soot are classified as EPA probable human carcinogens. Previous candle emission studies reported diverse types and quantities of organic constituents and PAHs. Many of these studies failed to clearly communicate that cancer risk from candle emissions is low.

Cancer risk may be increased for humans exposed to PAH-containing materials by inhalation or dermal contact for a long period. There is evidence of a dose-dependent relationship for some PAHs associated with skin contact on surfaces in animal and human studies (ATSDR 1995). The ATSDR has quantified specific PAH exposures that increase the human risk for cancer.

The EPA classifies seven PAHs as probable human carcinogens. Compounds for which animal data are sufficient to demonstrate a cause-and-effect relationship between exposure and cancer incidence (rate of occurrence) in animals, but where human data are inadequate or absent, are classified by the EPA as group B2 probable human carcinogens. These PAH B2 carcinogens include Benz[a]anthracene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[a]pyrene, Chrysene, Dibenz[a,h]anthracene, and Indeno[1,2,3-cd]pyrene. Several of these PAHs, including benz[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, chrysene, dibenz[a,h]anthracene, and indeno[1,2,3-c,d]pyrene, have caused tumors in laboratory animals through inhalation, ingestion and prolonged skin contact.

EPA lists diesel exhaust as a mobile source air toxic due to the cancer and noncancer health effects associated with exposure to whole diesel exhaust including PAHs, particulate matter and exhaust gases. EPA believes that exposure to whole diesel exhaust is best described, as many researchers have done over the years, by diesel particulate concentrations.

The benzene-soluble fraction of coal tar pitch volatiles and mineral oil mist, which contain several PAH compounds, are regulated by OSHA. The OSHA permissible exposure limit for CTPV is 0.2 mg/m^3, time weighted average (TWA). The American Conference of Governmental Industrial Hygienists (ACGIH) threshold limit value (TLV) is 0.2 mg/m^3, TWA. The ACGIH classifies CTPV as a confirmed human carcinogen.

Burning candles do not necessarily generate these specific PAHs or at airborne concentrations that can affect health. Table 4 lists compounds that are classified as probable human carcinogens by the EPA and OSHA.

Table 4

PAHs Classified as EPA and OSHA Human Carcinogens

EPA B2 Probable Human Carcinogens	OSHA Regulated CTPV PAHs
Benz[a]anthracene	Phenanthrene
Benzo[a]pyrene	Anthracene
Benzo[b]fluoranthene	Pyrene
Benzo[k]fluoranthene	Chrysene
Chrysene	Benzo[a]pyrene
Dibenz[a,h]anthracene	
Indeno[1,2,3-c,d]pyrene	

Although PAH-induced reproductive, developmental and autoimmune toxicity have not been observed in humans, these effects have been demonstrated in animal studies (ATSDR, 1995). In test animals, benzo[a]pyrene (BaP) has caused blood and liver damage when high doses were ingested, and immune system damage through contact with the skin. Mice fed high levels of BaP had decreased fertility, and damaged sperm and reproductive organs. Pregnant mice fed BaP had offspring with reduced viability, decreased birth weight, higher cancer incidence and reduced fertility.

Animal testing demonstrating development and reproductive toxic risks associated with PAH exposure have concerned childbearing women. Studies in animals have also shown that PAHs may cause harmful skin effects and alter the body's ability for fighting disease after both short- and long-term exposure (ATSDR, 1995). Persons affected with auto immune diseases could be affected by candle soot containing PAHs. BaP has caused respiratory tract tumors from inhalation, and stomach tumors, leukemia, and lung tumors from ingestion. Short-term exposure to high levels of BaP may cause red blood cell damage, leading to anemia and a suppressed immune system. Long-term exposure to BaP has resulted in skin rashes, sensitivity to sunlight, eye irritation and cataracts. Morbidity and mortality studies have found an increase in lung cancer rates in people exposed to coke oven emissions, roofing tar emissions and tobacco smoke; all of these compounds contain BaP and other PAHs. Cancers of the larynx and the scrotum may also be associated with PAH exposure.

Watson et al.(2001) demonstrated that pressure, airflow, temperature, fuel stoichiometry and fuel type affect the formation of chemical constituents and soot particle shape in carbon black and combustion generated soot. (Stoichiometry refers to the amount of air needed for complete combustion of a particular fuel.) Both soot and carbon black are similar with regard to formation and structure, however, Watson revealed that soot contains more organic compounds and PAHs than does carbon black. Watson also showed that organic hydrocarbon

constituents and graphite content varied in concentration among newly formed and aged particles.

Hebgen et al. (2001) demonstrated that pressure, temperature, fuel type, and residence time affect the relative amounts of curved and planar PAHs formed, the yield of fullerene molecules, and the relative amounts of fullerenic and graphitic carbon in soot particles. Fullerenes are fuel combustion evaporation products consisting of closed spherical shells comprised only of carbon atoms. Fullerenes play a direct role in the formation of PAHs. Fullerenic development of PAHs can also be formed in low-pressure fuel-rich flames of certain hydrocarbons, the highest yields being obtained under conditions of substantial soot formation.

Hamins (1993) characterized the chemical composition of fuel soot at various distances from the flame. Scanning electron microscopy analysis showed that newly formed soot particles were fuel specific, but mature particles were similar for some fuels. Newly formed particles had different chemical constituents from those of mature particles. The age of the particle and related chemical constituents, have specific effects on PAH quantity and carcinogenicity. The finding of age dependent particle constituents demonstrates the need for analyzing the hazards of aged particles.

Marvin et al. (2004) found that toxicity and carcinogenicity varied among ambient carbonaceous particles that were physically and chemically similar. Diesel fuel-, gasoline-, wood- and paraffin-created soot have unique morphologies, PAH constituents and carbon content. Marvin pointed out that the carcinogenicity of diesel emissions might be due to sulfates, nitrogen oxides, carbon monoxide and various hydrocarbons, rather than to soot. Risk assessments of diesel soot particulate are influenced by particle composition and confounding exhaust factors.

The statement in Krause's U.S. Scented Candle study that candle soot and diesel soot have the same cancer potential and cancer risk slope factor is invalid. Soot formed by different fuels and under different conditions, produces unique PAH constituents. The EPA attributes diesel emissions risk primarily to the PAHs attached to the diesel soot particulate. Since the candle soot particulate may have a composition different from that of diesel soot, the two cannot be presumed to pose the same cancer risk.

Huynh (1991) investigated candle sooted surfaces inside a church, for PAHs. Analytical results revealed 882-µg benzo[ghi]perylene per gram of candle soot and 163 µg BaP per gram of candle soot. However, airborne concentrations of contaminants do not correlate well with surface levels, due to environmental factors that affect airborne concentrations in an open space (Nazaroff, 2004). Therefore, the above results cannot be reliably used to derive inhalation doses.

The above studies reported PAH emissions that varied with the composition and number of candles. The reported airborne concentrations of human carcinogenic PAHs were too far below EPA and OSHA regulatory limits and guidelines to be of concern.

Schwind et al. (1994) identified airborne quantities of formaldehyde, naphthalene and dioxins from 30 candles burning for 4 hours in a 50 m^3 room.

Lau et al. (1997) identified acetaldehyde, formaldehyde and BaP from 30 candles burning for 3 hours in a 40 m^3 room with realistic air flow conditions. Each study burned more candles per volume of air than is typical in residential or occupational settings.

Fine et al. (1999) werenot able to measure significant levels of PAHs from paraffin and beeswax candles burning in an air chamber volume of 0.64 m^3.

Wallace (2000) presumed that citronella candles were a source of PAHs in a real time measurement study. Wallace pointed out that he did not quantify or verify his findings.

Dioxin Emission Studies

Recent investigations have measured airborne dioxin concentrations from emissions of burning candles. Agencies throughout the world do not agree on carcinogenic potency of dioxin in humans, classifying dioxin as non-cancerous, suspect, probable or likely. In 1976, a chemical facility explosion in Seveso, Italy exposed a large population to relatively high levels of dioxin. Since the spill, claims have been made regarding dioxin's ability to induce cancer in humans. The media has long promoted the myth that dioxin is the strongest human carcinogen known to man. The current EPA reassessment (2003) considers dioxin a "likely" human carcinogen.

Sources

Dioxins are formed primarily as unintentional by-products of incomplete combustion and various chemical processes. Although forest fires and possibly other natural sources may produce dioxins, these sources are negligible compared with anthropogenic sources. Dioxins are produced in small quantities during the combustion of fossil fuels, wood, and municipal and industrial waste. Bleaching processes that were used in pulp and paper production produced dioxins, and they occur as contaminants during the production of some chlorinated organic chemicals, such as chlorinated phenols. Currently, the major environmental source of dioxins is incineration.

Dioxin Toxicology

Animal health effects. In some animal species, 2,3,7,8-Tetrachlorodibenzo-p-Dioxin (2,3,7,8,-TCDD) is especially detrimental and can cause death after a single exposure (ATSDR, 1999). Exposure to levels lower than lethal concentrations can cause a variety of effects in animals, such as weight loss, liver damage, disruption of the endocrine system, and weakening of the immune system. Some animal species exposed to TCDDs during pregnancy had miscarriages, and the offspring of animals exposed to 2,3,7,8-TCDD during pregnancy often had severe birth defects including skeletal deformities, kidney defects, and weakened immune responses.

Chronic exposure of animals to dioxins has resulted in several types of cancer. No information is available on the carcinogenic effects of 2,3,7,8-TCDD in animals following inhalation exposure (ATSDR, 1998). Animal studies have reported tumors of the liver, lung, tongue, thyroid, and nasal turbinates from oral exposure to 2,3,7,8-TCDD. TCDD was evaluated by the International Agency for Research on Cancer (IARC) in 1997. Based on human epidemiological data, dioxin was categorized by IARC as a "known human carcinogen." However, TCDD does not affect genetic material, and exposures from environmental and occupational sources do not reach cancer risk levels (World Health Organization, 1999).

Human health effects. In humans, short-term exposure to high levels of dioxins may result in skin lesions, such as chloracne and patchy darkening of the skin, and altered liver function. Long-term exposure is linked to impairment of the immune system, the developing nervous system, the endocrine system and reproductive functions.

Individuals who may be exposed to higher than average levels of dioxins include those who ingest food containing higher concentrations of dioxins than are found in the commercial food supply. These groups include recreational and subsistence fishers who routinely consume large amounts of locally caught fish, subsistence hunters who routinely consume the meat and organ tissues of marine mammals, and subsistence farmers living in a contaminated area who consume farm-raised beef and dairy products. Persons who live near industrial or municipal incinerators, and persons who live near hazardous waste sites contaminated with dioxins could be exposed to higher levels of dioxins than the general population.

The EPA (2003) reassessment finds that dioxins are potent animal toxicants with potential to produce a broad spectrum of adverse effects in humans. Dioxins adversely affect reproduction and development, the immune system, chloracne (a severe acne-like condition that sometimes persists for many years), and cancer. Human studies, primarily of workers occupationally

exposed to 2,3,7,8-TCDD by inhalation, have found an association between 2,3,7,8-TCDD and lung cancer, soft-tissue sarcomas, lymphomas, and stomach carcinomas, although for malignant lymphomas, the increase in risk is not consistent (ATSDR, 1998). The EPA characterizes the complex mixtures of dioxin to which people are exposed as a "likely human carcinogen." This is because individual components of this mixture could be characterized as "human carcinogens" or "likely human carcinogens" under EPA's draft cancer risk assessment guidelines (1996, 1999). In particular, TCDD, the most toxic of the dioxins, can be identified as a "human carcinogen" under the Agency's draft guidelines, based on the weight of the animal and human evidence, and the other dioxins as "likely human carcinogens."

According to the EPA, current evidence suggests that both receptor binding and most early biochemical events such as enzyme induction are likely to demonstrate low-dose linearity. The mechanistic relationship of these early events to the complex process of carcinogenesis remains to be established. If these findings imply low-dose linearity in biologically based cancer models under development, then the probability of cancer risk will be linearly related to exposure to TCDD at low doses. Until the mechanistic relationship between early cellular responses and the parameters in biologically based cancer models is better understood, the shape of the dose-response curve for cancer below the range of observation can only be inferred with uncertainty. Associations between exposure to dioxin and certain types of cancer have been noted in occupational cohorts with average body burdens of TCDD approximately 1-3 orders of magnitude (10 to 1,000 times) higher than average TCDD body burdens in the general population. In terms of total TEQ, the average body burden in these occupational cohorts level is within 1-2 orders of magnitude (10-100 times) of average background body burdens in the general population.

Dioxin Levels Generated from Candles do not Reach Criteria Levels

To express the relative toxicity of the various dioxin compounds, the concept of Toxic Equivalent Quotient has been applied to the dioxin family. Toxic equivalent (I-TEQ) is a method facilitating a mutual comparison of substances belonging to the same chemical group eliciting various toxic effects and to present them at a comparable level in relation to the most toxic one of the group (e.g., TCDD in this report). The TEQ weights the entire mixture on 2,3,7,8 TCDD, the most toxic of all of the dioxin compounds. The TEQ is calculated from toxic equivalency factors and the absolute concentration of each chemical component. Tests of individual contaminants are used to establish the potency of each compared to TCDD. PCDDs and PCDFs can contain from 4 to 8 chlorine atoms; both the number and position of the chlorine atoms determines the overall toxicity of each congener. 2,3,7,8-tetrachlorodibenzodioxin is the most potent congener and is assigned a toxic equivalency of 1. The relative toxicity of the remaining congeners is expressed as a fraction of 1. Therefore,

the total weight of the toxic equivalents is the measurement used when an environmental mixture of several PCDD/PCDF congeners is being evaluated.

Greene et al. (2003) critically reviewed 5000 scientific papers on TCDD toxicology. The identification of a no-observed-adverse-effects level (NOAEL) of a 13 ng / kg maternal body burden was the most relevant for deriving a reference dose (RfD) for humans. The studies were consistent in reporting more than a dozen different dioxin induced adverse effects in humans over the past 25 years. The most consistent clinically important adverse effect following human exposure is chloracne. Chloracne is a severe skin condition caused by skin contact with chlorinated hydrocarbons and is characterized by pustules and skin lesions. Following a review of all published studies, the best estimate of a NOAEL for production of chloracne is around 160 ng / kg body weight.

The EPA contends that the epidemiological data alone are not yet deemed sufficient to characterize the cancer hazard of TCDD as being a "human carcinogen." However, combining consistent, suggestive evidence from epidemiological studies with the strong positive associations in animal studies and inferences drawn from mechanistic data supports the characterization of complex mixtures of dioxin and related compounds as "likely" cancer hazards. While major uncertainties remain, efforts of this reassessment to bring more data into the evaluation of cancer potency have resulted in an estimate of 1×10^{-3} per pgTEQ/kgBW/day. This slope factor and resulting risk specific dose estimate represents a plausible upper bound on risk based on evaluation of human and animal data within the range of observation and at a minimally detectable response level. With an upper bound risk estimate of 0.001 cancer cases, $20m^3$ inhalation volume and 70 kg, the calculated EPA unit risk per mg/m^3 is 2.85×10^5.

In the last 10 years, the enforcement of stricter emission standards for dioxins and furans by many countries significantly reduced the release of these substances into the environment. Initiatives on municipal and medical waste burning are regulated by Europe and the United States (Table 5).

Table 5

Waste Management. Global Waste Emission Standards for Dioxins

Country	Municipal Waste ng TEQ/m^3	Medical Waste ng TEQ/m^3
European Union	0.1	0.1
Canada	0.080	0.080
Hungary	0.1	0.5
USA	13 (total mass)	2.3 (total dioxins + furans)
New Zealand	0.1	0.1

Dioxins Produced from Candles

In December 1992, the German national press caused public anxiety by the report, "High Levels of Dioxins in Purple Candles." German candle sales declined since the public's perceived health threat. In response to these allegations, Malisch (1994) measured airborne dioxin from candle emissions with pigment "violet 23." Malisch tested candle emissions produced from the purple colored layers of 24 candles. Even though visual observations indicated the same color intensity of all 24 candles, the candles contained different mixtures of dyes. The three candles with the highest amounts of chlorinated dioxins in their bulk wax contained 1.8, 1.4 and 0.8 ng I-TEQ/kg wax. Eighty three percent of all samples were below 0.5 ng I-TEQ/kg and 57% below 0.2 ng I-TEQ/kg. The amount of dioxins in the bulk wax is significantly lower than EPA's reference dose of 13 ng I-TEQ/kg when ingested. This implies that when burned the amount of dioxins inhaled, as a percent of room air will be significantly lower than amount of dioxins in the wax. This report does not measure candle emissions, but estimated that burning a candle "highly contaminated" with dioxin TEQ could lead to an air concentration of 40 pg TEQ/m^3. Using the calculated EPA unit risk of 2.85 x 10^5, the cancer risk is 1 in 100, exceeding the upper bound risk by a factor of 10. Because dioxins were not measured from candle emissions in this study, it is not possible to accurately predict airborne concentrations and cancer risk.

Schwind et al. (1994) measured dioxin exposures generated from candle emissions. Schwind theorized that the thermal breakdown of aniline dyes and oils in dyed and aromatic candles are responsible for dioxin generation, but his results indicated nonsignificant airborne levels of dioxins. From airborne modelling, they calculated an airborne concentration of 0.038 pg I-TEQ/m^3. Using the calculated EPA unit risk of 2.85 x 10^5, the cancer risk is 1 in 100,000.

Lau et al. (1997) evaluated emissions of burning candles for the presence of PCDD/PCDF. They reported dioxin from burning candles as an emission factor adjusted to the amount of contaminant in 1 g of wax. PCDD/PCDFs were detected in all types of candles tested, with emission factors ranging from 0.004 to 0.047 pg TEQ/g. Assuming a worst-case scenario, the highest emission factors were used to estimate the chemical air concentrations that would result from burning 30 candles for 3 hours in a room 40 m^3 (a total of 540 g wax burned). Resulting PCDD/PCDF was 0.635 (pg TEQ/m^3). Using the calculated EPA unit risk of 2.85 x 10^5, the cancer risk is 2 in 10,000.

Considering that the EPA estimates that the upper bound cancer risk of PCDD/PCDF for the general population is 10^{-3}, the risk posed by PCDD/PCDF in candle emissions is insignificant.

Ultrafine Particle Emission Studies

Ultrafine particles are classified by the National Institutes of Health (NIH) and National Institute for Occupational Safety and Health (NIOSH) as airborne particles with diameters less than 100 nm.   These comprise soot, and particles formed from gases (secondary particulate matter) and inorganic materials. Soot and fuel combustion particulate are terms describing the same particulate and can be used interchangeably. The main chemical constituents of atmospheric secondary particulate matter in urban locations include sulfuric acid, ammonium sulfate, ammonium and organic compounds. The relative distribution of particle types in the atmosphere is described by apportionment, a term used by the EPA and the research community. Particle monitoring of the atmosphere throughout urban areas in the U.S. shows that fuel combustion particulate accounts for 20-65 percent of the total mass (U.S. EPA 1998-2001).

The ambient atmosphere particle distribution includes size ranges from a few nanometers to micrometers. Coarse mode particles are large particles generated by mechanical processes and include windblown dust, salt particles from windblown sea spray, and mechanically formed particles such as from construction sites. These large particles settle out rather quickly and have lifetimes in the atmosphere limited to hours. Fine particles (such as candle soot) and coarse particles have different chemical compositions, sources, and lifetimes in the atmosphere, and there is little mass exchanged between the particles in these two modes.

Soot formation is a two phase process. In the first phase, soot consists of smaller nuclei mode particles that form from the vapor phase of burning organic hydrocarbon fuel. This vapor consists of various molecular weight hydrocarbons that transform quickly into a solid phase (nucleation). Large aromatic (benzene-based) hydrocarbons grow into primary nuclei particles to about 0.5 to 2 nm in diameter. Continued growth occurs in the secondary accumulation phase when particles group together to form larger sized masses (coagulation). Candle soot is created with the same soot formation process. Li et al. (1993) describe candle soot formation as initial particle size of 4 nm with continued growth to 20-50 nm and slower growth of chains and clusters to larger sizes reaching 2,500 nm. Their description of particle coagulation is consistent with coagulation times (Hinds, 1999). Chemical constituents of soot depend on candle fuel type, candle additives and environmental conditions during combustion. Ambient atmospheric particles are formed from many different sources including gas stoves, gasoline, propane, paraffin candles, motor vehicles, and forest fires. Carbonaceous soot particles make up a fraction of the atmospheric particulate, the other fraction being sulfates, nitrates and metals. Particle formation following particle emissions produces new particles with unique elemental composition, inorganic ions and carbonaceous compounds (organic and elemental carbon). Based on these complexities, it is difficult, if not impossible to classify the chemical makeup of ambient soot particles.

Carbonaceous agglomerates formed from the accumulation phase of soot formation are present throughout the world: in urban settings, rural areas and remote locations. Morawska et al. (2003) reported that soot particles have been collected from the atmosphere above Phoenix, Arizona and from the atmosphere at an altitude of 2.1 km above the Southern Ocean, off Tasmania.   The soot particles from both locations are likely of anthropogenic origin and consist of individual soot globules that are typically 20 to 50 nm across, attaching to one another in a chain. Land-based Arizona soot probably originated from an industrial source; that from the Tasmanian ocean probably originated from aircraft or ship emissions.

Gasoline and diesel exhaust contribute the majority of ultrafine particles in ambient air of a typical city (Junker et al., 2000; Molnar et al., 2002). Miguel et al. (1998) discovered that heavy-duty trucks emit several hundred times more carbonaceous particulate than light duty trucks. Morawska et al. (1998) and Ristovski et al. (1998) demonstrated that a significant fraction of diesel emission particles have diameters smaller than 0.1 μm. Gasoline combustion particles are mostly agglomerates ranging from 0.01 - 0.08 μm. Ristovski et al. (2000) reported that particles from natural gas emissions are smaller than from diesel or even gasoline emissions and range from 0.01-0.07 μm, with the majority being between 0.020 and 0.060 μm. The World Health Organization reported that most particles emitted from vegitative burning, which includes controlled burning and uncontrolled fires, are ultrafine, with only a small fraction in the larger size range, and with most of the mass present in particles less than 2.5 μm in aerodynamic diameter. Ambient soot particles generated by combustion sources range in size from a few nanometers to several hundred nanometers (Figure 1).

Particle Size Distribution of Combustion Sources
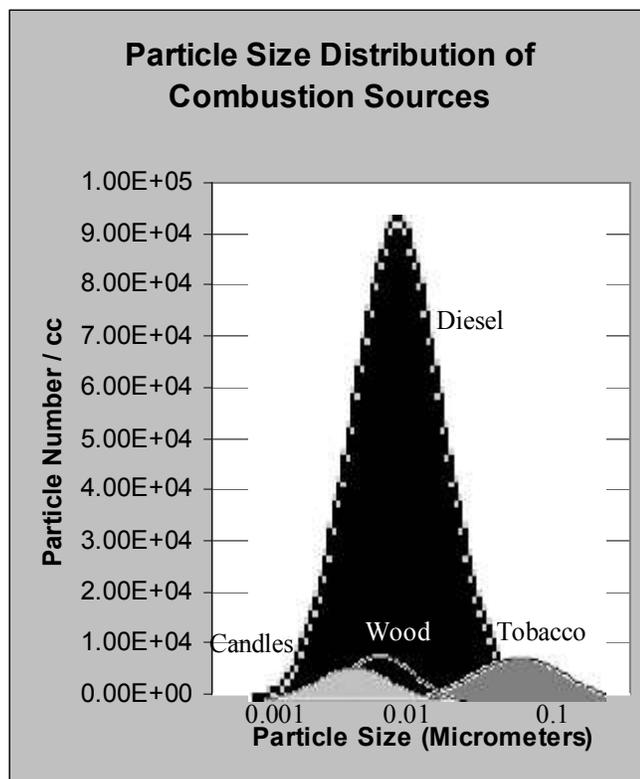
Figure 1. Example of particle size distributions. Particles are formed from various combustion sources including vehicles running on diesel (tallest peak) and gasoline, nucleation phase candle soot (Li et al., 1993), forest fire (wood burning) and environmental tobacco smoke (far right peak). (Morawska et al., 1998; Ristovski et al., 2000)

Particles inhaled indoors are comprised of direct indoor sources of incense, candles, and cooking, and outdoor sources of vehicles and wood burning. Concentrations of ultrafine particles vary by geographical location, physical location and types of nearby sources. Outdoor sources add to the indoor particle concentration through infiltration pathways. Several studies demonstrate the impact that outdoor sources have on indoor environments. Wallace (2000) reported that indoor particle concentrations range from one to two times outdoor concentrations.   Monitoring station measurements of a Boston neighborhood environment reveal background particles as numbering from a few thousand particles/cm^3 to about 2×10^4 particles/cm^3. In indoor microenvironments averaged across sample days in Boston, mean ultrafine particle concentrations ranged from 3800 to 140,000 particles/cm^3, with 7-200 µg/m^3 of PM$_{2.5}$ and 5-12 ng/m^3 of particle-bound PAH. PM$_{2.5}$ indoor-outdoor ratios exceeded 1.0 in settings with high levels of human activity, with lower ratios for ultrafine particles. Levy et al. (2002) showed that cooking contributed significantly to increased levels of indoor pollutants. Reponen et al. (2003) investigated the exposure gradient in ultrafine particle concentrations for people living near interstate highways in Cincinnati, Ohio. In addition, optical particle concentrations of sizes ranging from 0.3-20 µm and mass concentration with

20

cutoff diameter of 2.5 μm were taken. Ultrafine particle concentrations closer to roads or in tunnels, where car traffic contributes the most significant urban sources of particle numbers, are ten times higher or more and can reach or exceed levels of 10^5 particles/cm^3. PM_{10} and $PM_{2.5}$ concentrations are no more than 25 – 30% above background level at roads (calculated as the difference between the maximum at the road and the background levels). Ultrafine particle concentrations measured at specific locations in the United States and Europe reflect the ultrafine particle count variability (Table 6).

Table 6

Urban Ultrafine Particle Number Concentration Levels.

Author	Location	Range particle number concentrations (particles/cm^3)
Osunsanya et al. (2001)	Aberdeen, UK	Average: 10,241, range: 740 – 60,636
von Klot et al. (2002)	Erfurt, Germany	Average: 17,300, range: 3,272 – 46,195
Bloch, et al. (2002)	Santa Monica, CA (Near Airport)	Average: 56,104, range: 4,940 – 300,000 Average: 41,276, range: 10,800 – 428,000
Wichmann et al. (2000)	Erfurt, Germany	Average: 15,773 ± 10,321
Pekkanen et al. (2002)	Kuopio, Finland	Average: 44,300
Penttinen, et al. (2001)	Helsinki, Finland	Average: 14,500
Ayers, et al. (1998)	Six City Study	Range: 10,000-50,000
Tiittanen et al. (1999)	Kuopio, Finland	Range: 6,980 – 40,200
Reponen et al. (2003)	Cincinnati	Range: 11,000 – 32,000
Levy et al. (2002)	Boston, Mass	Range: 3,800 – 140,000
Molnar et al. (2002)	Gothnenburg, DK	Range: 4,944 – 105,114
Hussein et al. (2004)	Helsinki, Finland	Range: 3,700 – 46,500
Pekkanen et al. (2002)	Helsinki, Finland	Maximum: 50,310
Dennekamp, et al. (2001a)	Aberdeen, UK	Maximum: 100,000

Ultrafine particles enter the human body from inhalation. In the lungs, these particles are free to react with biological tissue. Penetration into deeper structures depends on particle size and inhalation rate. Health effects of the inhaled particles rely on both the particle size and the way in which they react with the pulmonary tissue. Particles larger than 5 µm are less damaging and are trapped in the upper respiratory tract, nose and trachea. Particles just under 0.5 µm normally remain suspended in the lung and are expelled when exhaling. However, particles in the ultrafine range, i.e., less than 100 nm, can deposit into the deepest portion of the lung, the alveoli, where gas exchange takes place. The International Commission on Radiation Protection's (ICRP) 1994 model for regional particle deposition for light exercise predicts 50% alveolar deposition for 0.01-0.02 µm particles. For particles less than 0.1 µm, total deposition increases from 20% (0.1 µm) to 80% (0.01 µm) or even 95% (0.001µm)

The airborne concentration of smaller nuclei and accumulation mode particles resident in air space is dependent on density and proximity to other particles. Newly formed soot particles are less than 0.05 µm and deposit more deeply in the lung, as noted above. Nuclei particles quickly attract each other by van der Waals forces, reducing the ultrafine particle population and increasing the population of larger particle sizes. The newly formed soot nucleus grows into an elementary soot particle 4 nm in diameter and then clusters into small chains. The chains merge to produce primary particles, 20 to 50 nm in diameter, which coagulate to form larger soot aggregates of up to a few hundred nm.

Candle emission risk estimates have been using mass concentration rather than particle number to measure exposure. Mass concentration does not account for the dynamic nature of particle size distribution during generation. Removal mechanisms, such as settling, deposition to surfaces, or attachment to rain droplets, are weak for larger particles.

Particle surface area is larger for a higher number of smaller particles than for the same mass of larger particles. Most ambient airborne particles are in the ultrafine range. When interpreting a normal distribution of particle mass concentration by size, the total mass of the ultrafine particles is insignificant in comparison with the mass of the larger particles, thus masking the health effect of the smaller particles.

$PM_{2.5}$ and PM_{10} fractions are particle mass concentration terms found in the EPA regulatory language and literature to describe ambient air quality standards, both indoor and outdoor. $PM_{2.5}$ (fine particles) is the mass concentration of particles with aerodynamic diameters smaller than 2.5 µm. PM_{10} is the mass concentration of particles with aerodynamic diameters smaller than 10 µm. PM_1 or $PM_{0.1}$ fractions refer to mass concentrations of particles smaller than 1 and 0.1 µm, respectively. Previous mass concentration studies of $PM_{2.5}$ and PM_1 do not provide information on the particle size distribution. PM studies only describe the mass concentration of particles below the cutoff point or PM

number. Optical particle counters have the capability of enumerating particle counts across multiple size ranges, thus defining the particle distribution accurately and repeatably. Particles smaller than one micrometer are typically measured with a condensation nucleus counter. Ultrafine particle number and number size distribution data can be monitored in real time, whereas mass, mass size distribution or morphology require sample collection and electron microscope analysis. Size specific data is more useful than mass concentration because the narrow size range of the ultrafine particles can be measured and compared to current ultrafineparticle studies. In our study, ultrafine particle counts are monitored in real time to provide reliable and repeatable exposure estimates.

Figures 2a and 2b illustrate the differences between particle counts and mass concentration. The data used to show these differences are fictitious, however, the higher number of small particles in Figure 2a compared to the low mass in Figure 2b, drives home the point.   The size distribution for particles between 0.1 and 25 µm is heavily weighted toward the smaller particles (Figure 2a). However, the larger particles account for the bulk of the total mass (Figure 2b).
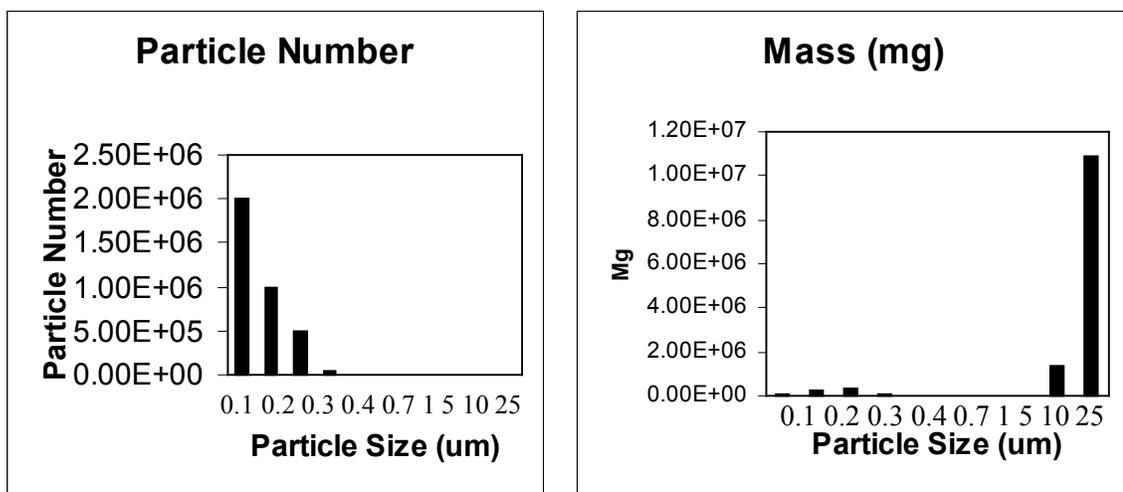
Figures 2a and 2b. Typical particle distributions with respect to number and total mass.

Animal and human studies of ultrafine particles have attempted to demonstrate an association between cardiac and respiratory disease. Urban sources of ultrafine particles are claimed to be associated with cardiopulmonary morbidity and increased rates of asthma, hospital admissions and medication usage. Donaldson et al. (2000) reported that some ambient particle concentrations are associated with pulmonary and cardiac adverse health effects in asthmatics. Seaton et al. (1995) reported that ultrafine particles may cause biological effects because their relatively large surface area and small size allow them to penetrate deep in the lung and react with epithelial and inflammatory cells. Ferin et al. (1992) demonstrated that rats exposed to ultrafine particles

have significantly more pulmonary inflammation than rats exposed to the same particle mass with larger diameters.

Candles generate ultrafine particles similar to many combustion processes found in ambient sources. Therefore, candle soot is classified as an ultrafine particle produced from a combustion process, along with vehicle emissions, cooking sources and natural processes including forest fires.

Deeper penetration of smaller particles into the lungs and changes in surface chemistry have a direct effect on biological cellular systems. Animal investigations have shown that pulmonary cellular inflammation, hyperplasia and neoplasia occur in rats exposed to ultrafine particles. Biokinetic mechanisms for these cellular changes have been proposed. Oberdorster (2000) reported cellular changes in animals exposed to ultrafine particles of titanium dioxide. Similar studies reported similar results with carbon black and styrene particles. Even though cancer risk from inhaling candle soot particles is uncertain, emerging studies suggest that the small size of the particle is responsible for cardiopulmonary health risks.

Pulmonary inflammation from exposure to ultrafine particle exposure may be caused by glutathione sulfhydride (GSH) depletion.   Glutathione is considered to be the most powerful, most versatile, and most important of the body's self-generated antioxidants (Kidd et al. 1997). Glutathione is found in almost all living cells. The liver, spleen, kidneys, pancreas, and the lens and cornea, have the highest concentrations in the body. GSH availability down-regulates the pro-inflammatory potential of leukotrienes and other eicosanoids. It is a powerful antioxidant and thus neutralizes free radicals and prevents their formation. GSH plays an important role in immune function via white blood cell production. Li et al. (1996) provided evidence that greater inflammation and depletion of GSH occurs in a rat lung containing ultrafine carbon black than containing fine carbon black.   Stone et al. (1998) showed that epithelial cells in culture exposed to fine or ultrafine carbon black (CB) exhibit depletion of GSH.

Oxidative stress mechanisms may be responsible for ultrafine particle-induced pulmonary inflammation. Donaldson et al. (2000) found that the antioxidant nacystelin instilled into the rat lung along with ultrafine particles, produced up to 60% less inflammation than ultrafine particles alone. Wilson et al. (2002), using several cell-free tests for the generation of free radicals, showed that ultrafine carbon black produced more oxidation than fine carbon black. Donaldson et al. (2002) demonstrated increased pulmonary inflammation in rats exposed to low-solubility, low-toxicity ultrafine particles from the same material. The pulmonary inflammation is related to the larger surface area of the smaller particles and associated oxidative stress. The authors suggest that contact with ultrafine particles and resulting oxidative stress will result in increased influx of Ca(++) into macrophages, via the membrane Ca(++) channels. Donaldson et al. (2003) chemically assayed free radicals at particle surfaces of ultrafine carbon

24

black particles. The free radicals cause oxidative stress to cells and ultimately, pulmonary inflammation. Donaldson speculated several pathways for cardiovascular effects including autonomic nervous system induced heart rhythm changes and ischemic events including increased clotting, homeostasis, and athermanous plaque (narrow artery buildup) rupture.

Some epidemiological studies look at associations between morbidity, mortality and adverse health effects, and mass concentrations; others make the comparison with ultrafine particles. Mass concentration data hides the particle distribution and the contribution of a particular particle size fraction to the mass. Ultrafine particles studies employ techniques that emphasize the impact of a particular particle size and health effect. Pope et al. (2000) estimated that for lifelong residents in the world's most polluted locations, the average life span decreased between 1 and 3 years due to fine particulate air pollution exposure. Penttinen et al. (2000) reported that the association of acute respiratory effects with ultrafine particle number was stronger than with fine particle mass. Janssen et al. (2001) showed that increased exposure to traffic is associated with increased hospital admissions for cardiovascular disease. The association implies that locations with greater urban traffic have a higher risk of cardiovascular disease.

Peters et al. (1997) measured the association between fine and ultrafine particles in a panel study of 27 nonsmoking adult asthmatics at different times. Peters provided evidence that a decrease in peak expiratory flow (PEF) and an increase in cough and feeling ill were associated with elevated concentrations of ultrafine particles, independently from fine particles.   Health effects from the 5 day mean of the number of ultrafine particles were larger than the mass of fine particles. The association between ultrafine particle number and decreased PEF was stronger than for particulate matter smaller than 10 μm (PM_{10}). The panel study revealed both net and gross changes in pulmonary function, with changes in concentrations of ultrafine and fine particles.   The panel approach was beneficial in revealing shifting patterns of PEF with particle size, that might go unnoticed with other research approaches. Differentiating between fine and ultrafine size particle counts, more accurately determined the size fraction responsible for decreased PEF.

Pekkanen et al. (2002) conducted a panel study among 45 adults with coronary heart disease in Helsinki, Finland during a 6 month period in 1998-1999. Electrocardiograms, recorded symptoms and exercise-induced ST depressions were recorded biweekly. Exposure monitoring for ultrafine particle number concentration and mass concentration was conducted within a 5 km radius of the study population's residences.   Particle number concentrations for particle diameter sizes 0.01-0.1 μm were most strongly associated with ST-segment depressions. The associations were strengthened for persons with the exclusions of left bundle-branch block, left ventricular hypertrophy or anterolateral infarction. Increased odds ratios ranged from 1.03 to 3.29 with 95%

confidence intervals ranging from 0.54 to 6.32.   This study demonstrated independent associations between both fine and ultrafine particles and the likelihood of specific electrocardiogram changes used as an indicator of myocardial ischemia (ST segment depression).

Wichmann et al. (2000) showed that mortality data associate fine particles with immediate health effects, and ultrafine particles, with more delayed effects. Immediate effects are attributable to respiratory disease mortality, whereas delayed effects show an increase in cardiovascular disease mortality. These results suggest particle size may affect symptom timing as well as symptom type.

That study also found that both fine particles (represented by particle mass) and ultrafine particles (represented by particle number) showed independent effects on pulmonary and cardiovascular mortality. The urban environment in Erfurt, Germany was characterized into gases and particles in two size categories. The groups consisted of ultrafine particles and fine particles with diameters between 0.1 and 2.5 µm. The fine particle group was further subdivided into three size ranges. The daily average total number concentration was 18,000 particles/cm^3 with 88% of particles below 0.1 µm and 58% below 0.03 µm in diameter. A time series approach was used to determine short-term changes in particle concentrations and concurrent changes in mortality of 200,000 persons. Relative risk was used to determine the relation of increase in death, to particle size range. Timing of effect was evaluated by examining pollutant levels from 0-5 lag days.   A lag period is defined as the period between exposure and effect by the best single day or over a multiple of days. Mortality increased in association with ambient air pollution, after adjustment for season, influenza epidemics, day of week, and weather. In the sensitivity analysis, the results proved stable against changes of the confounder model. Results pointed to comparable associations for ultrafine and fine particles in a distributed lag model and contribution of the previous 4 to 5 days. The data suggest a more delayed association of ultrafine particles than of fine particles if there are single-day lags. Comparable associations for gaseous pollutants are artifacts of carbon monoxide linearity with particles from the same sources. The data also suggests that fine particles cannot be used as an indicator of ultrafine particles. No clear associations or patterns were observed for immediate or delayed effects.

Von Klot et al. (2002) revealed that acute effects of ultrafine particles on respiratory health of asthmatics are more severe for adults than for children. Pulmonary inflammation develops over a period ranging from hours to days. Cumulative effects over 5 days are stronger than same-day effects.

Osunsanya et al. (2001) investigated the association of PM_{10} and ultrafine particle number with PEF and respiratory symptoms (cough and shortness of breath) in 44 adults having either chronic airflow obstruction, or asthma and older than 50 years.   Bronchodialator use, PEF rate and scored symptoms were measured daily for 3 months. Central site monitoring in Aberdeen, Scotland

consisted of an R&P 1400a mass concentration monitor and a TSI condensation counter for ultrafine particle measurement. Twenty-four hour mean, minimum and maximum particle number counts and particle mass concentration were recorded daily. Time series and regression analysis were used to statistically determine the association between particle concentrations and respiratory health effects. Correlation was good (> 0.5) for indoor and outdoor particle concentrations; however, correlation was poor (< 0.5) between PEF and particle measurements. Odds ratios for mean temperature, mean wind speed and mean humidity were nonsignificant for 10% decrement in day time peak flow rate, high cough score and high breathlessness score. No significant associations between respiratory symptoms and ultrafine particles were found. Odds ratios between PM_{10} and respiratory symptoms were 1.284 with 10% decrement in daytime peak flow rate, 1.47 with high cough score and 1.214 with breathlessness score. The study suggests that the effects of ultrafine particles are indistinguishable among PM_{10}, black smoke and the particle count. The effects of different but intercorrelated particle measures and confounding weather patterns added to overall errors in particle measurements. The strongest associations were with high scores of shortness of breath and same day PM_{10}, and high scores of cough and 3-day PM_{10}. With increases of PM_{10} from 10 to 20 $\mu g/m^3$, there was a 19% increase in the rate of 10% decrements in daytime PEF. Stronger associations among coughing, shortness of breath and PM_{10} imply that the coarse particles contribute more to these symptoms than ultrafine particles.

Penttinen (2001a) explored ultrafine particle and self administered PEF testing with spirometry confirmation. Of 78 adult asthmatics recruited for the study, PEF rate was measured on 57 acceptable non-smoking adult participants, and self-administered biweekly spirometry on 54 non-smoking adults located less than two kilometers from the monitoring site. The spirometry testing improved the accuracy of PEF tests. Ambient particle concentrations taken at a central monitoring site in Helsinki, Finland included PM concentrations and particle number counts. The PM cutoff sizes were 10, 2.5 and 1 μm particle diameter, while particle number size ranges were classified into 0.01-0.1 and 0.1-1 μm diameter. The median mode for ultrafine particles was 14,500 particles/cm^3, and 800 particles/cm^3 in accumulation mode. Ambient nitrogen dioxide and carbon monoxide were measured concurrently with the particle measurements. PEF testing occurred in the morning, after work and before bedtime. Biweekly spirometric testing for six months found that forced vital capacity (FVC), forced expiratory volume in one second (FEV1) and pulmonary expiratory flow residual (PEFR) had inverse but mostly insignificant associations with particle numbers on previous days. Standard errors were large and the only significant association was with the accumulation mode. No associations were found for the coarse mode. PEFR results were inversely associated with PM_1 and $PM_{2.5}$ concentrations. PEFR was most strongly associated with particle number concentrations of particle sizes from 0.1 to1.0 μm. Mean particle number concentrations were negatively associated with daily PEFs. Comparatively, particle mass concentrations had less effect than ultrafine particles. Nitrogen

dioxide and carbon monoxide had little effect on respiratory symptoms and bronchodialator use.

Von Klot et al. (2002) conducted a panel study of 53 adult asthmatics in Erfurt, Germany during the winter of 1996/1997.   The effects of ultrafine particles, particle mass concentration and ambient gases (NO_2, CO, SO_2) were measured against participant-inhaled short acting B2 agonists, inhaled corticosteroids, wheezing percent, shortness of breath, problematic breathing while sleeping, phlegm and cough.   Concurrent measurements of ultrafine particle number concentrations 0.01-0.1 micrometer in diameter ($NC_{0.01-0.1}$) with a mean of 17,300/cm^3, and mass concentrations of fine particles 0.01-2.5 µm in diameter ($MC_{0.01-2.5}$) with a mean of 30.3 µg/m^3, did not correlate well (r=0.45). Corticosteroid use was associated with cumulative exposures over 5 days (OR=1.22, 1.23, 1.28) and 14 days (OR=1.45, 1.51, 1.44) of ultrafine and fine particles. Beta2-agonist use with 5-day mean ultrafine particle number concentration was associated with ultrafine particles raging from 0.01-1 µm ($NC_{0.01-0.1)}$ (OR=1.11, 1.01-1.21) and $PM_{2.5}$ ($MC_{0.01-2.5}$) (OR=1.10, 1.01-1.20). Asthma medication use and respiratory symptoms are associated with ambient particle concentration and ambient nitrogen dioxide. The study population was not uniform with regard to degree of asthma and cigarette smoking. The assumption of personal exposure across the city with one central monitoring site provides a source of misclassification of exposures.

Reviews of statistical findings on particle number concentration and health effects show an inherent weakness in many morbidity studies. Relative Risk and Odds Ratios showed relative strength in a particular study; however, overall strength was close to the null hypothesis, 1.0.   Published studies in this area do not satisfy Hill's criteria for cause and effect. Figure 3 displays a summary of odds ratios in several epidemiological studies.
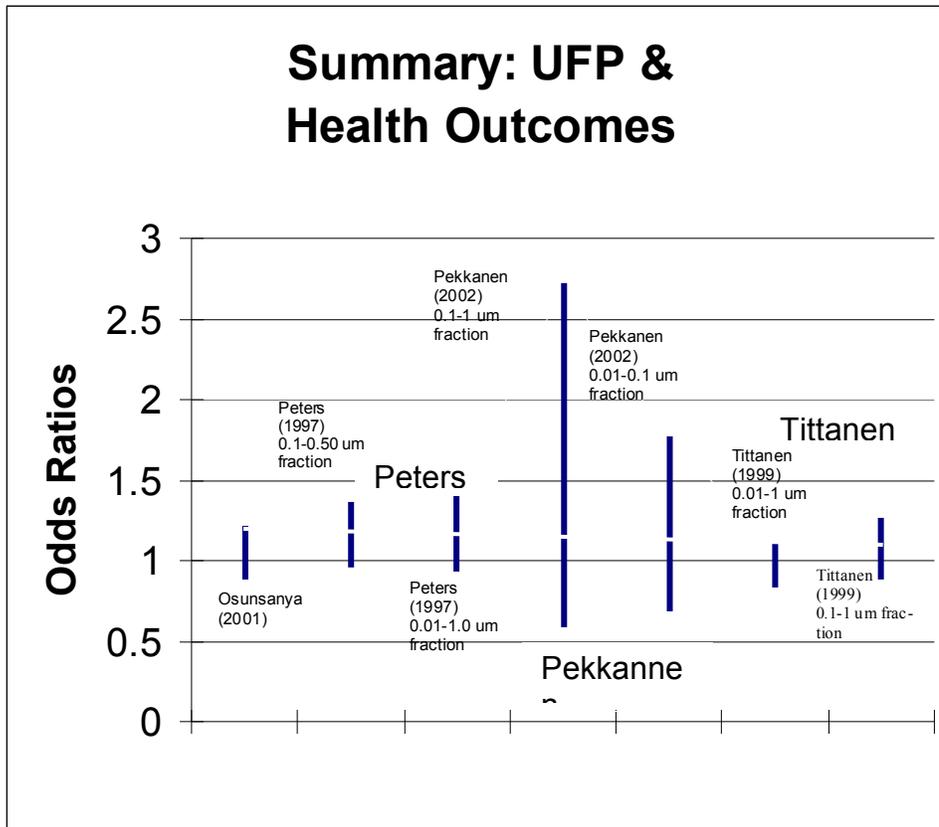
Figure 3. Summary of the results of the epidemiological studies on association of ultrafine particles with health outcomes: odds ratios and 95% confidence interval (bars) for ultrafine particles by study.

The relative risk of developing cardiopulmonary disease from candle soot particle exposure is additive to ambient carbonaceous ultrafine particle exposure in the ambient atmosphere. However, many studies associating ambient ultrafine particle exposure to cardiopulmonary disease do not appropriately control for confounding.   Rather than singling out the diesel particulate, contributory factors such as the diesel exhaust factors may be additive cancer and cardiovascular risks (ATSDR, 1999). Pope and Donaldson have shown an increased in morbidity and mortality in their six-city study, however, the study lacked particle distribution below mass cutoff points. There is growing evidence of cardiovascular and pulmonary effects with ultrafine particle number concentration. Bioassay tests have supported ultrafine particle association theories based on cardiovascular cytotoxicity and effects on immunologic pathways in cells. Clinical studies of asthmatics challenged with ultrafine particles have verified results that show changes in FEVs. Even though specific studies of candle soot and health effects do not exist, the size effects of the candle soot and ultrafine particles are known. Current studies include the increase in asthmatic responses and cancer mortality. Mirrored studies involving particle number led to uncertainty because of differences in contributing factors from cities. Comparisons of candle soot with diesel particulate, and mass

concentration or ultrafine particle mortality studies, encountered confounding difficulties. Animal testing involving controlled ultrafine particle characteristics and testing environments have resulted in repeatable measurement data for cellular, pulmonary and cardiovascular changes.

Reliability of Previous Chamber Studies

Previous studies characterized candle emissions by chamber-generated particles are believed to provide unreliable exposure data (Krause, 1999; Van Alphen, 1999; Lau et al., 1997; Nriagu et al., 2000). These chamber studies employed designs that may have led to an overestimation of soot exposure and health risk, due to excessive chamber turbulence, temperature and unequal pressure. The data could be unreliable because environmental conditions inside the chamber are not the same as in a normal room. Chamber conditions may have affected the amount of soot generated and PAH content. Watson et al. (2001) showed that fuel types and environmental variables (pressure, temperature, stoichiometry and the presence of oxidants) produce distinct carbon particles with a unique solvent extractable fraction (SOF). Soot has more SOF and PAHs than carbon black. The type and quantity of PAHs are unique to soot formation conditions. Many absorbed organic compounds contained within soot are formed during combustion. Shaddix et al. (1994) demonstrated that flickering flames emit significantly greater amounts of smoke for a given fuel flow rate than non-smoking steady flames. Flame flickering increases candle soot generation rate. Morgan et al. (2003) showed that increasing the temperature of a burning fuel changes the chemical composition of the emission.

There are also issues with what data were collected and how they were used. Chamber data measurements reported mass concentration and assumed a single monodisperse diameter for particle size. Measurements of mass concentration did not account for the dynamic nature of the size distribution in an occupied room. Candle emission exposures were estimated from chamber data and uncertain models. The inaccurate chamber data, model uncertainties and reliance on mass concentration data suggested a need for improved chamber testing. We find that improved chamber test conditions invalidate previous test chamber data.

Mass Concentration Measurement of Candle Soot

Another shortcoming of previous chamber studies is that they measured mass concentration and not the candle soot size fraction most responsible for alveolar deposition (< 100 nm). Studies that reported cardiopulmonary morbidity and mortality associated with mass concentration size fractions lacked particle size distribution data. (Spengler et al., 1985; Siegmann, 1990; Dockery et al., 1992; Seaton et al., 1995). Studies that employ mass concentration measurement, collect all particles below a specific diameter cutoff point, and do not take note of particle size distribution. Study replication and comparisons of

health effects are difficult when the size fraction most responsible for the effects is hidden in the mass concentration.

Investigators of chamber candle emissions used exposure estimates to model mass concentration data from chambers (Fine et al., 1999; Krause, 1999; Malisch, 1994; van Alphen, 1999). Corrections were neither made for particle distribution dynamics that took place inside the chamber, nor contemplated for the indoor air quality model. True particle size dynamic data are required for accurate regional pulmonary dose determinations.

Li et al. (1993) revealed that prior to agglomeration forces, candle soot particles are less than 10 nm. Consistent with the Aerosol Technology handbook by Hinds (1999), particle agglomeration increases with particle concentration and time. The particle distribution inside a space is a function of the time that the nuclei particles are first generated and the time that they coagulate into larger particle sizes. The agglomeration rate inside a chamber and a room result in unique spatial and temporal particle size distributions. Size specific particle count data is essential for the evaluation of specific particle size ranges and health risks. Particle deposition in specific regions of the pulmonary system is based on particle size, sedimentation, impaction and particle shape. Ultrafine particles deposit in the alveolar region where gas exchange with capillaries takes place.

Osunsanya et al. (2001) explained that particle size fractions in particle count studies were advantageous over mass concentration studies, as the particle sizes most responsible for lung deposition or a size-related health effect can be specified. The lack of particle count data in mortality and morbidity studies demonstrated the need for particle size distribution data specific to health effects.

Pekkanen et al. (2002) evaluated the health effects produced by fine-particulate air pollution ($PM_{2.5}$, aerodynamic diameter < 2.5 μm). Health effects were associated with a high number of ultrafine particles in urban air. Pekkanen established that specific particle size distribution data is useful for source determination, spatial variability and deposition dynamics. Central site monitoring of mass concentration in $PM_{2.5}$ studies was an improper measure of ultrafine particle exposure.

Reliance of Candle Emission Risk Assessments on Chamber Data

Studies that reported increased health risks from candle emissions inputted mass concentration chamber data into indoor air quality models (Fine et al., 1999; Krause, 1999; van Alphen, 1999). Such models may consider pressure relations between rooms, infiltration, exfiltration, ventilation rate exchanges, particle sinks, thermal gradients, particle agglomeration, temperature, temporal spatial distribution, and filtration. Models employ probabilistic mathematics and can be simple (few variables) or complex (many variables). Simple models are

easier to calculate, however, factors and terms are often missing from these equations. Complex models are mostly probabilistic in nature, dependent on the quality and types of equations, and often subject to aggregation errors caused by lumping variability factors as a single value.   Both categories of models depend on input from the user. Model uncertainty with chamber data is best reduced by using a simulated test room environment.

Reliability Analysis of Indoor Air Quality Models

Furtaw et al. (1996) measured particle concentration in rooms containing point sources of particle emissions. Furtaw found that particle concentrations tend to be higher in close source proximity than when they are farther away. Models vary in their predictive ability to resolve spatial particle concentration for lone particles or particle clouds. The assumption in deterministic models that particles are homogenous and evenly dispersed is unreasonable. Variation due to proximity gave an explanation for "personal air" monitors often yielding higher concentrations than nearby micro environmental monitors. Proximity errors were corrected by using a two-compartment model with the source located in a small virtual compartment within the room compartment linked with a stochastic air transfer rate parameter. This method provided a more reliable simulation of exposure concentrations than the well-mixed model for assessing exposure to emissions from active sources.

Nicas (2000) evaluated the accuracy of indoor air quality models and discovered that some predictive models for particle dispersion in indoor air were deterministic. The models did not account for the probabilistic nature of the pollutant concentration at a given room position and time. Nicas demonstrated that this variability must be considered when estimating concentrations involving small numbers of contaminant particles. Deterministic models specified fixed deposition, sinks and air exchanges. Stochastic models presented probabilistic determinations that were more likely to occur with particle exposures. Particle concentrations in space were dynamically influenced by eddy currents, turbulence, cloud formation and varied deposition rates. Probabilistic models permit forecasting of spatial variation in concentration. Nicas developed two models based on Markov chains to account for a portion of this variability and to describe indoor air contaminant dispersion by turbulent diffusion and advection, and removal by exhaust airflow. Turbulent eddy diffusion models portray a continuous concentration gradient with distance from an in-room contaminant emission source. Nicas modeled exposure with a Drivas model that is equivalent to the Markov mode.   The Drivas model represented particle spatial distribution with wall surfaces and particle removal by exhaust air.

Guo et al. (2000) critically evaluated indoor source emission models. Of 50 models, he found that only a few were accurately predictive of risk.   He showed that component models of risk were source emission and exposure modeling. The complexities of the two components made predictions difficult.

Mathematical probability equations were calculated using factor input that incorporated mass concentration, sinks, coagulation, particle size, infiltration, exfiltration, air changes, filtration, particle counts, turbulence, and eddy currents. Guo confirmed that for a determination of concentration and particle size distribution to be reliable, it must reflect actual exposure. As research on particle dynamics continues, new features were added to these models to account for uncertainties in the model. Many of the models Guo studied have become obsolete.

Stephen et al. (2002) advanced the risk analysis of cancer risk from benzene in candle emissions using Monte Carlo Simulation statistical methods. Benzene emission data was attained from previous chamber studies. The results of Monte Carlo simulation more accurately approximate the true value based on deterministic and probabilistic inputs. Unfortunately, large variance may be produced by the simulation. Input included exposure sum assessments over a 24-hour period, emission rate, building volume, exchange of the building air, candle burning time, and post-extinguished candle time. Latin hyper cube sampling and benzene exposure were estimated in the 50th and 95th percentiles and factored into the risk calculation. (The statistical method Latin hypercube sampling was created by Ronald L. Iman, J. C. Helton, and J. E. Campbell, 1981, to generate a distribution of likely series of parameters from a multifaceted distribution.) The U.S. Environmental Protection Agency (EPA) Exposure Factors Handbook and inhalation unit risk values from the U.S. EPA Integrated Risk Information System (IRIS) provided points of reference for the risk calculation. Point estimates were calculated at the 95% upper confidence limits of emission rate. Stephen reasoned that the health risks associated with candle use, of 2×10^{-7} and 3×10^{-6} (excess cancer per individuals) are acceptable for protecting public health. The accuracy of the chamber data is questionable because benzene concentrations generated under chamber conditions may vary when compared to benzene generated from candles in a normal room.

Levy et al. (2003) completed an investigation of air pollution control cost-benefit. Levy found it challenging to determine the accuracy of dispersion model estimates to predict population exposures. Not many models can capture the necessary spatial and temporal domains with adequate sophistication. Model validation with field measurements was not feasible for marginal concentration changes. Levy applied the notion of intake fraction (the fraction of a pollutant or its precursor emitted that is eventually inhaled) to provide insight about population exposures and model performance. Relative concentrations of ammonia, sulfate, and nitrate were modeled to determine the influence on ammonium nitrate intake fractions. Model input included past intake comparisons, dispersion models and applied source-receptor matrices. These findings provided a framework for investigating factors that influence population exposures to particulate matter.

Nazaroff (2004) investigated estimation uncertainties for ultrafine particle deposition rates. Infiltration, exfiltration, coagulation, and airflow mixing were measurable, however, turbulence, eddy currents, and boundary layers were predicted by probabilistic modeling. Lack of uniform mixing, spatial distributions and concentrations near boundary layers introduced significant error in determining accurate particle concentrations. First order rate assumption was inadequate for the model, and transport rates through boundary layers depended on near-surface airflow conditions. Following candle soot generation, particle removal occurred by exfiltration through air change, filtration, and deposition. Exfiltration losses are straightforward to quantify and apply to all sized particles. Filtration and deposition losses rely on particle size, shape, composition, concentration, room air rate, room surface characteristics and ductwork volumetric airflow. Coagulation is a significant factor for determining particle concentration when submicrometer particles are greater than 8000 particles/cm^3. Fewer particles are counted because the combining of particles reduces the particle number concentration. Deposition rates and room surface-to-volume ratio varied largely. Deposition rates for ultrafine particles range from 2 to 50 µm/second. For a room surface-to-volume ratio of 3 m^2/m^3, the equivalent deposition rates range from 7.2 to 1,800 µm/hr.

The literature appears to show that candle emissions contain soot, dioxin, lead from lead wicked candles, PAHs and organic compounds. The reporting of these emissions may alarm the public, however, the quantities and types of emissions are either too low or are inconclusive. Lead emissions are a danger only to children, and only from lead wicked candles, which are rare in today's marketplace. Chamber candle studies demonstrating hazardous candle emissions are flawed due to excessive turbulence, pressure, and temperature extremes not found in real rooms. Krause's scented candles study incorrectly assumes a similar toxicology for candle soot and diesel soot and does not take into account that PAH content are different for fuel types and environmental factors. However, chamber improvement will lead to a more accurate candle emission generation of soot, PAHs and metals. The improved chamber will confirm that previous chambers produced misleading high emissions. To make unnecessary the inappropriate use of mass concentrations to predict health outcomes, a simulated occupational environment will provide more appropriate ultrafine particle count measurement and PAH data. Because soot is the same as carbonaceous particles in the ambient environment, soot particle counts can be compared to the ambient environment.   We predict that occupational exposure to candle soot is low compared to the ambient environment ultrafine particles.

CHAPTER TWO

EXPERIMENTAL METHODS

Previous candle studies have reported specific organic and inorganic candle emission constituents.   Of these constituents reported, the types most acknowledged to be a potential health risk are lead, PAHs, ultrafine particles, dioxin, formaldehyde and benzene.   Candles are used in homes and some occupations. This study sheds light on the quality of previous candle studies and emphasizes the occupational risks for some workers exposed to candle emissions.

Candle studies have been either chamber studies or test room studies reporting particle mass concentrations.   The chamber studies were flawed because interior chamber conditions do not match conditions inside a real room. Chamber conditions were turbulent and had different fuel oxygen ratios; therefore, they produced unreliable data. An improved chamber simulating real room conditions may provide different soot generation rates and emit different organic species.

Expected results of an improved chamber are lower soot generation rates and a more accurate assessment of health risk.   Measurement data includes PAHs, lead and zinc emitted from the same types of candles burned in a previous chamber study for comparison. Comparison of average emission rates and chemical concentrations will show that non-metal wicked candles are not a public health hazard for lead or zinc emissions. The data will show that the mass concentration data of scented candles in Krause's scented candles study is affected by the interior chamber conditions.

Ultrafine Particle Exposure Data inside a Test Room

Chamber studies lack conditions normally found in a room such as surfaces, ventilation and space.   These conditions affect particle growth, deposition, particle size distribution and filtration. In the effort to control for environmental variables and to provide a more accurate measurement method, a simulated occupational environment test room was built to provide more realistic data from candle emissions. The test room provided more reliable data than chamber model extrapolation because real conditions can be applied directly rather than having to be assumed or modeled. A real test room improves the accuracy of exposure estimates and provides a more reliable estimation of candle emission risk. Another major benefit of using a test room is the ability to

collect accurate particle count concentrations rather than mass concentration data. This allows the actual size fraction, ultrafine particle counts to be measured and compared to current studies exploring the association of ultrafine particles and cardiopulmonary disease. Mass concentration data is limited to data under a cutoff point and hides the fine particulate fraction that may be responsible for specific diseases.

Chamber data did not provide enough evidence for associating normal candle use with disease. Based on cited references associating ultrafine particles with cardiopulmonary disease, the test room allowed for measurement of candle soot ultrafine particles. Additional testing of PAHs was done to determine if PAHs may be formed differently under room conditions as opposed to chamber conditions.

To determine the occupational risk from candle emission exposures, candle use and occupational settings were determined prior to constructing the test room. The candle market was surveyed to determine the three most predominant occupations using candles, candle types, number of candles, exposure duration, ventilation and furnishings. These factors influenced the design of the test room and of the candle burns. The measured particle count data were used to calculate Lifetime Average Daily Dose (LADD) for workers in candle use occupations, based on EPA human factor data. The particles comprising candle soot are in the ultrafine size range that is cited in some ambient particle pollution morbidity studies. The associations of cardiopulmonary disease and the ultrafine range are estimated as quantified risks in these studies and may be possible to use as a reference. The comparative risk analysis of candle soot dose to ambient ultrafine particle dose provides a perspective on occupational candle soot exposure acceptability.

Our test room measurement data will show that PAHs are not a threat. The UFP particle number data will be used to show that ambient UFPs provide a greater human dose. The exposure data and EPA human factor data was incorporated into the lifetime average daily dose calculations for specific occupations.

Experimental Scenarios for the Improved Chamber and Test Room

Upon chamber redesign, candles are burned inside the chambers and emissions are drawn into sampling media. Temperature and pressure are controlled to best simulate a normal room during the test run. Pre-tared filters are analyzed for particle mass, PAHs, lead and tin.   To compare soot generation rates between the scented candles study and scented candles burned in the improved test chamber, 20 test runs of individual candles will provide sufficient data for statistical analysis.

The test room is constructed to closely simulate average occupational environments for the three most predominant candle use occupations. A typical test room run includes selected candle types and numbers based on occupation, burning under ventilated and non-ventilated conditions. Measurements were taken of particle distribution using standard optical particle counters, ultrafine particle concentration using a condensation nuclei counter and PAHs using OSHA Method 58.

Design of the improved chamber was influenced by designs described in previous chamber studies (van Alphen, 1999; Krause, 1999). The improved chamber environment included two 45-liter stainless steel chambers and a background airbox (Blake and Silver, 2000). A centrifugal fan drove ambient air through the background airbox, within which a system of filters effectively divided the airbox into two disjoint regions. The air in the filtered region could enter each of two identical chambers that contained the burning candles.

The filtering was done by a Honeywell model 22500 circular HEPA filter, surrounded by a Honeywell model 38002 blanket style prefilter with activated carbon. These were tightly mounted just inside the background airbox, providing a positive seal that precluded leakage between prefiltered and filtered regions.

Each chamber was made of 0.059 in. stainless steel, with 45 l capacity, 10 in. internal diameter, TIG-welded seam, end caps of 1 and 0.5 in. acrylic plate, and four baffles (determined to be necessary through preliminary testing) to deflect inlet airflow from the candle(s) until nonflickering was achieved. This reduction in flame flickering was a considerable improvement over Krauss's chamber.

The background airbox included a hinged lid resting on foam rubber seals, to allow direct access to services. To achieve 40 l/min air flow into the chambers, four inlet holes were drilled into the base of each chamber. The inlet holes, two with 1/8 in. diameter and two with 3/32 in. diameter, were spaced midway between the center and outer shell of each chamber at the four compass points. A tube inserted through the chamber pressure tap hole aided in blowing out candle flames. The inlet holes and matched airflow served to maintain constant temperatures and equalize pressure in the chamber.
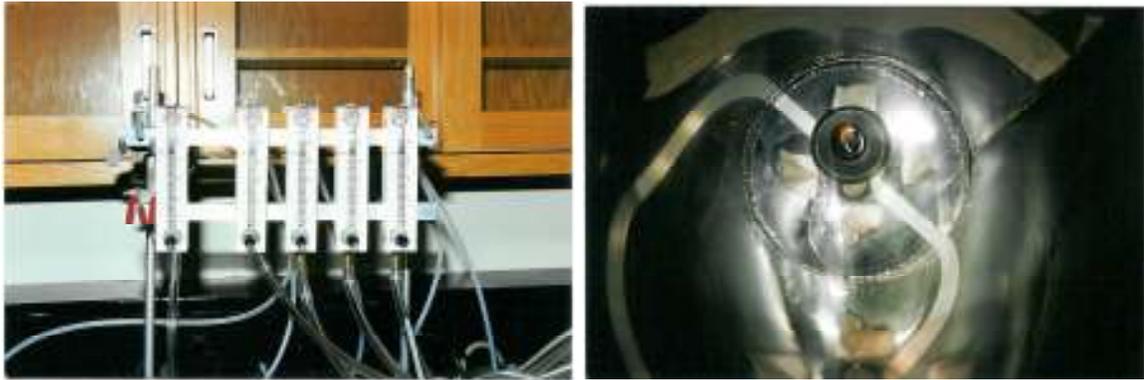
Figure 4. Blake, Silver chamber used to test candle emissions. Left: flow controllers over chamber. Right: non-flickering candle inside cylindrical chamber during testing (bird's eye view).
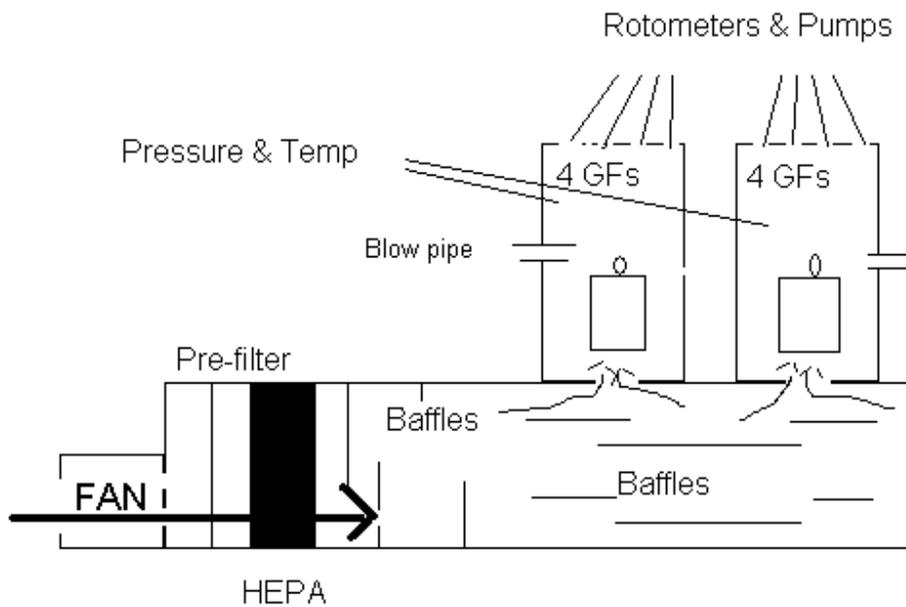
Figure 5. Schematic Design of Candle Emissions Collections Chambers with Baffles.

Candle Emission Measurement in the Improved Chamber

Chamber pressure (0.01 in. of water) was monitored with a Dwyer gauge, interior chamber temperature monitored with a thermometer and air speed

maintained with an adjustable rheostat during all segments of the test runs. Rotometers calibrated with 37 mm glass fiber filters inline allowed for constant checking of airflow. A stopwatch timer allowed for accurate timing of the mass concentration sample tests. Mass concentration data was collected and analyzed for each scented candle.

Scented candles similar to the candles in Krause's "U.S. Scented Candle Study" (1999) were selected to match generation rates. Candle emissions were collected on four air-sampling cassettes arranged in line over the chamber. Activated carbon pre-filter and high efficiency particulate air (HEPA) filter inside the air box provided a positive seal and assurance against outside air contamination entering the box. Similar sampling and analytical procedures for mass concentration and PAH analyses were the same as in previous candle chamber studies. In accordance with OSHA Method 58, GFE 37 mm filters were used to collect mass concentration for gravimetric and organic analysis.

Test Room Design, Construction and Use

The experimental layout consisted of a 40 m^3 simulated workplace environment test room, candles at 3-foot height and aerosol measurement equipment. A single door and closed single pane window provided the only openings to the test room. Furnishings and ventilation were matched to the specified occupation. The air conditioning system to the room consisted of a supply vent and return vent filtered with a lightly loaded matt filter. Sets of 1, 5, 10 and 20 unscented, scented and church candles were placed on a tray near the center of the room. Eight runs of each candle set were measured under ventilated and nonventilated conditions.

To determine occupational health risk from candle emission exposure, specific candle use information was required. A list of questions facilitated gathering information and included consensus of types of institutions, candle types, length of candle burning and number of candles used. Initially, candle suppliers throughout the U.S. were called to determine the types of businesses using candles the most. Restaurants, spas and churches were phoned over a three-month period to gather information on candle demographics. Directly asking the end user businesses regarding their use of candles provided answers to specific use questions that suppliers cannot provide. A tally sheet was employed to record results on a particular spa, restaurant or church. The respondent was asked to provide his or her full name and asked the following questions.

1. How many candles do you burn in a room?
2. What are the types of candles burned in the room?
   - Candle stick: paraffin, beeswax or gel?
   - Shape: votive, tapered, gel in glass or other?
   - Color: white or other than white?

- Scented or unscented?
- Do you trim the wicks prior to lighting?
- What types of furnishings are present in your restaurant, church or spa?

3. What is the duration of time that a worker may be near a burning candle during an average workday?

Questions were clarified upon request. Participants were told that their name or business would not be used in the survey. Unnecessary conversations were avoided with the respondent. The respondent was thanked for participating in the study.

Survey data was scrutinized to determine candle selection and numbers burned. Candle demographic data was analyzed to determine the majority candle types for a particular occupation. Information regarding furnishings allowed for realistic materials in the test room. Types of surfaces of materials (furnishings) influence the particle dynamics and exposure concentrations. The test room with representative furnishings made it possible to eliminate sink rate modelling, a source of particle concentration uncertainty. At last, dose is calculated with exposure duration and particle count data.

Test Room Design Variables

Candle types, exposure duration, and furnishings in occupational settings were specified from survey information.

Candle type

Survey data identified three types of candles in three predominant candle-using occupations. Restaurants typically burn long-duration unscented candles during the dinner hour. Churches burn candles in a variety of circumstances that include lighting candles by icons, ceremonial candle lighting and church vigils. Because vigils use the most candles, long tapered candles typical of vigils were selected. Aromatherapy candles are typically burned in spas. Scented candles emit fragrant aromas that relax and sooth clients during message therapy and a number of scented candles surround a spa bath.

Vanilla scented candles are widespread, however, to test the differences of soot generation rate between unscented and scented candles, a more strongly scented Yankee candle was burned.

Exposure Duration

Restaurants typically burn long duration nonscented candles during the dinner hour. Churches vary in use-time of candles. With massage therapy, massage workers are exposed to candle emissions during the length of the

massages. Exposure duration is short when preparing a spa bath because the spa worker lights the candles and leaves.

Furnishings

Furnishing selections in the test room were based on a consensus of materials used in representative settings that would be common to all three occupations. The choice of common furnishings was essential to generate accurate occupation specific data because material surfaces have distinct sink rates that influence particle reentry into the air stream. The intensity of particle attraction to a room object varies with the type of material. Particles attach strongly to some materials and weakly to others. If the attraction is weak, particles can reenter the air stream when disturbed. Common to all three are windows, commercial carpeting, gypsum wall, ceiling tiles and either tables or pews. Desks represented restaurant tables or church pews in our test room because the wood surface is a typical material used in these institutions. Corrections were not made for some materials in institutions such as tablecloths in restaurants, or water and Formica tops in spas. Churches span many architectural styles, some in shopping centers, some under elaborate vaulted ceilings. The shopping center style of architecture was used due to testing limitations.   The shopping center style has less room volume than a grand church or synagogue, however, this provided an environment for greater particle density and a "worst case" scenario. Because we did not simulate the most typical church environment and used the shopping center model, some error is assumed in the test room model for churches.

Candle Emission Measurement in the Test Room

Candles were lit with a long butane lighter.   Measurements of ultrafine particles were positioned off center to the candle burn, 3 feet away.   Candle emission particle counts were measured under two environmental conditions, unventilated and ventilated.   The ventilated environment was approximately 2 air changes per hour, the lower end of most commercial establishments that are generally between 2-12 air changes per hour.

Measurement Techniques

During candle burning, three particle counters measured three overlapping size ranges of particles, 0.1-1 µm, 0.5-10 µm, and 0.02-1 µm.

Two laser optical particle counters (OPC) were employed to measure particle distribution during selected test runs. The OPCs allowed observation of particle growth dynamics. After the soot is generated, growth in particle size occurs with agglomeration of particles. Two OPCs were used, the MetOne 237H with particle size range from 0.1 to 1 µm and the LASAIR 510 with a range of 0.5 to 10 µm. Both instruments were calibrated according to American Standards

Technology Method (ASTM) using Duke Scientific latex spheres. The latex spheres were generated to a comparison chamber with a Royco 236 nebulizer and reference optical particle counter. Particle size concentration data included the ambient background, peak, extinguishing the candles, and decay time.

The particle distribution data was used to gain knowledge of particle size distribution dynamics in the unventilated test room. The ultrafine particle concentration behavior of the rapid rise to peak concentration and sudden decline seemed odd compared to the ventilated room. The new question of a healthier particle environment in the unventilated test room had to be evaluated. Perhaps ultrafine particles diminish to near background levels due to the effects of coagulation.

UFP concentrations from candles typically used in occupations were measured in the test room. Multiple test runs of the specified candle type and candle number provide statistical strength and a reduction in uncertainty. Test room UFP data allow calculation of lifetime average daily dose (LADD) for workers in specific occupations. Ambient Clearwater UFP data and ambient UFP data cited in studies allows for calculation of a benchmark to be used to compare occupational dose to ambient dose.

A condensation nuclei counter (CNC) was chosen to measure ultrafine particles emitted from candle emissions. The TSI 8525 P-Trak is comparable to laboratory CNC counters and provides the appropriate range of measurement as needed for the study. Particle counters convey particle size specific data rather than a single cutoff point as in mass concentration measurements. CNCs are optical particle counters that grow ultrafine seed particles to a detectable size by condensing alcohol vapor around the particle. The seed particle surface can be a small cluster of vapor molecules, an ion, or a solid particle (Molnar et. al 2002). The CNC is an appropriate measurement device for the range of candle generated soot particle counts from the individual soot particle (20-50 nm to the accumulation mode (50-2,500 nm). The CNC automatically data logged particle concentrations during candle emission testing. Recording of temperature and relative humidity occurred with each burn session. Specific information included background levels, peak concentrations and decay rates.

Non-condensation optical particle counters were not selected for the candle soot ultrafine particle measurements because of optical measurement errors called Rayleigh scattering (Hinds, 1999) often observed for particles less than 100 nm in diameter. Optical particle counters count particles by light scattering and collecting this energy on semiconductors. Most of the light extinction caused by aerosols is due to scattering. Aerosols both absorb and scatter solar radiation. Particles in the 0.1 - 1.0 μm size range scatter light efficiently, as their radii are comparable to the wavelengths of visible solar radiation. Scattering of light in this size interval (Mie scattering) is characterized by the Mie theory, which states that particles interact with radiation as a function

of their surface. Aerosols smaller than 0.1 µm are called optically small particles. They scatter solar radiation by Rayleigh scattering. Rayleigh scattering is inversely proportional to the fourth exponent of the wavelength of the radiation.

Measurement of Scented and Unscented Candle UFPs

Ultrafine particle count data measurements between scented and unscented candles were done to determine extent of agreement with Krause's scented candles study.

PAH Measurements

Rather than relying on chamber study results, for measurement of PAH, OSHA Coal Tar Pitch Volatile (CTPV) PAH carcinogens were measured in the test room. PAH measurement during a test run provided a more accurate assessment of PAHs that may form differently in a test room compared to a chamber. Soot and PAH formation are affected by turbulence, heat and pressure inside a chamber. Airborne PAHs were collected and analyzed for a typical candle burn. The test room with ten candles provided a maximum environment for PAH concentrations from candles. Placement of the candles was in the center of the room and the sampling system opposite the candles. Pre-calibrated high volume Medo pumps equipped with glass fiber filters ran for several hours of candle burn.   Analysis of samples by the OSHA gravimetric method (Method 58) was done by Schneider Laboratories, Inc. of Richmond, Virginia, an AIHA accredited laboratory. Because of expected lower concentrations, adjustments to Method 58 allowed for more sensitivity in the mass concentration weighing of samples. Increasing the recommended flow rate from 2 to 7 l/min raised the total sample volume from 960 liters to about 2000 liters. Increasing the flow rate accommodated a complete burn of the candles and allowed for increased sensitivity of the method, by lowering the limit of analytical detection.   Analytical results of our PAH data were compared to OSHA permissible exposure limits for compliance determination.

CNC and Unventilated Conditions

Test Room Conditions and the effect of Door Movement

Prior to measuring candle soot in the sealed unventilated test room, particle counts were logged to observe the effect of door movement on the counts. The impact of door movement on measurement results was important to the study because experimenter access was required during the extinguishments of candles, twice causing the door to be quickly opened and shut. Manually recorded data from the TSI 8525 portable CNC occurred at one-minute intervals. Opening and closing the door occurs within a 2 second time frame to extinguish the flames and to leave the room. Testing was done to determine the extent and significance of the measurement error. Skewed data resulting from opening the

door would mean that the tester would not be able to leave the room.   To make this determination, testing was done without leaving the room. Measurements took place during the entire candle burn and post extinguishments. Manual measurements of a sample set of 10 candles occurred at one-minute intervals in the closed test room with a stopwatch. Graphs from candle testing in which the door was open and shut for extinguishments were compared to the data collected without opening the door. The resulting graphs were observed for rise, peak, decline, second extinguishments peak and slope angle. The second post-extinguishments peak and the subsequent decreasing slope were observed to be similar regardless of the door being opened and shut briefly to extinguish the flame.

Unventilated Workplace Test Room Conditions

The unventilated test room was sealed to the outside and to the inside of the room. The airborne concentration of soot particles did not have to be corrected for the exchange of particles inside of the room and outside of the building. Each test occurred with single, five, and ten candle sets in the sealed unventilated room. Measurement with the particle counters occurred during the burn, during extinguishment and for 30-60 minutes beyond the extinguishments. Sealing the room occurred after lighting candles and running the particle counter. Maximum peak particle concentrations generated within the sealed unventilated room provided data expected to simulate conditions in an occupational workplace. Using the highest expected particle concentration when comparing candle soot particles to referent ambient concentrations allows for a qualitative risk comparison. If the maximum concentration results in a lower risk from candle soot exposure compared to ambient particle exposure, then concern over the candle soot risk is diminished.

Background particle count data was collected to adjust the peak particle count concentration for each candle test. Comparisons were made between candle types and candle numbers using one-way analysis of variance. The linearity of particle concentration with number and type were analyzed to determine the predictability of exposure concentrations. Lifetime average daily dose of a specific occupation was calculated from the test room data. Risk assessment was qualitatively assessed as a comparison to ambient exposures.

Ventilated Workplace Test Room Conditions

Ultrafine particle count real time measurements in the ventilated room illustrated the effect of the air handler and filtration on peak concentrations. The ventilated test room was prepared to reflect a typically ventilated room with infiltration and exfiltration. The door was closed during testing, however, the doors and windows were not sealed as they were with the unventilated test room condition.   To our surprise, the data produced an initial peak, however, with the air exchanges, the peaks and valleys duplicated themselves over the air

44

exchange rate. The steady state average was calculated from the initial burn time to flame extinguishment. Sets of one, five, ten, and twenty candles were allowed to burn over 2-3 hours. Eight runs for each candle set were measured with the CNC.

Particle count data from the ventilated room condition were not adjusted with background concentrations. Differences were observed for particle dynamics in the ventilated and unventilated test room conditions. The steady state ultrafine particle concentrations in the ventilated test room cannot be added to the background concentrations, but require complex particle distribution modeling.   Comparisons were made between candle types and candle numbers using one-way analysis of variance. Steady state particle concentrations measured from candle types and numbers reflecting an occupation were used for the dose estimation and comparative risk assessment.

<p align="center">Lifetime Average Daily Dose Model</p>

The test room exposure data and EPA human factor data were inputted into the Lifetime Average Daily Dose (LADD) model for workers in specific candle use occupations. Model variables to calculate dose included inhalation rate, exposure duration, particle concentration based on occupation specific candle types, number of candles used and burn duration.

The dose and ambient ultrafine particle concentration data were inputted into the dose model. LADD for candle soot and ultrafine particle exposures were calculated using a 70 year factor in the averaging time. Noncancer risks use the the 30 year factor for occupational exposures.   The LADD was calculated using equation 1, where C= airborne contaminant concentration (particles/m^3); IR = inhalation rate (m^3/day); ED = exposure duration, or the total time a person is exposed to the soot or ultrafine particles (days); AT = averaging time (days), equal to 70 years risk assessment, or 25,550 days. Averaging time for occupational exposure is 30 years or 10,950 days.

$$\text{LADD} = (C \times IR \times ED) / (AT \times 1000) \tag{1}$$

Averaging and standard deviation were used to clearly show the variability of the data and its reliability. The occupational dose was compared to the ambient dose.

<p align="center">*Rationale for the LADD Comparison*</p>

A comparison of occupational candle soot exposure to ambient and secondary source soot exposure provides a qualitative (semi-quantitative) estimate of risk. Concrete morbidity and mortality rates for persons exposed to ultrafine particles are lacking in the literature. Difficulties with confounders and

lag times of symptom onset have plagued numerous studies that show only weak associations. No regulations or mandates exist for reducing the concentration of ultrafine particles at this time. The public perceives the risk of ultrafine particle exposure to ambient levels of airborne soot from traffic, kitchen stoves and combustion sources as acceptable.   The strength of this design is that exposure concentrations and dose can be predicted for specific workplaces.

This type of occupational study has never been done. The comparison of candle soot with ambient soot sheds light on dose apportionment and on whether candle soot is necessary to control.   Many times, the occupation is blamed when people are exposed to the same things outside the workplace.

LADD Model Input

Input data in the occupational dose model includes the number of candles used, type, and burn duration. Particle concentration data was collected previously from the unventilated and ventilated test room setups. The soot concentrations collected from the unventilated test room were used to determine occupational risk in workplaces that may occasionally be unventilated. In locations with moderate climates, extreme climates with moderate temperature months, and during power outages, ventilation may not be operational. The soot samples collected from the ventilated test room represent occupations in temperature extremes where the temperatures are controlled by ventilation.

Input for ambient dose model allowed for a comparison of candle soot dose to ambient soot dose. Ambient input included soot concentrations of urban locations, secondary indoor sources and results of ultrafine particle studies worldwide. Ultrafine particle measurements were taken in the Clearwater area at restaurants, churches, spas, parking lots, roadways, commercial stores and a mall. Candle soot particulate was measured in a number of area restaurants to validate readings in the simulated test environment. Additional input data includes secondary sources such as gas stoves in restaurants. Secondary sources were used in determining the contribution and impact to occupational exposure. Literature data that cited ultrafine particle concentrations in urban environments was validated by CNC measurements taken by roadways, by kitchens and inside stores.

The candle soot comparative risk assessment was qualitative and relied on a comparison of two exposure sources. Quantitative risk assessment measures the probability of developing a disease based on established dose response curves. Reference ultrafine particle dose associated with cardiopulmonary disease is unreliable due to the weak associations and confounding affects of other pollutants. Due to the lack of a reliable reference dose, occupational candle soot dose is compared to ambient ultrafine particle dose.   Breathing ambient ultrafine particles is an acceptable risk.

CHAPTER THREE

RESULTS

To show that previous chamber studies produced unreliable data, factors that affect candle soot generation were identified. This included a flickering flame caused by turbulent conditions and associated fuel oxygen ratio. Chamber conditions that do not resemble actual room conditions were eliminated. The improved chamber resulted in soot reduction.

Chamber Mass Concentration Results

Previous chamber data was compared to improved chamber data by calculating averages, variance and mean comparisons using the t-test. The data sets should show that the increased soot generation in the previous chamber designs exaggerated exposures and overestimated the risk from candle emissions.

Soot generation rate was calculated by dividing the soot mass by the burn time in minutes for each candle run. Comparison of previous chamber study data revealed a lower soot generation in the improved chamber data (Table 7). Figures 6a and 6b are graphical representations of the candle soot data from each chamber. T-test results of the two chambers confirm that the means are statistically different, t= 2.47, 95% C.L. (Table 8).

Table 7

Mass Concentration Generation rate of Chamber Studies

Scented Candles Study (Krause) Chamber Data	Improved Silver-Blake Chamber Data
Heavy sooter: 1650 µg/min Scented: 188 µg/min All scented: 393 µg/min Non-scented: 54 µg/min	Scented candles: 63 µg/min

Average candle soot generation rate was 63 ug/min, much lower than the mean 393 µg/min reported in a previous study.

Original Chamber - Scented Candles

Average = 393 ug/m

Average = 393 ug/minute
Geometric mean = 2.26 ug/min

Log ug/minute

Individual Scented Candles

Scented Candles - Improved

Average = 63 ug/m

Average = 63 ug/minute
Geometric mean = 1.59 ug/minute

Log ug/minute

Individual Scented Candles
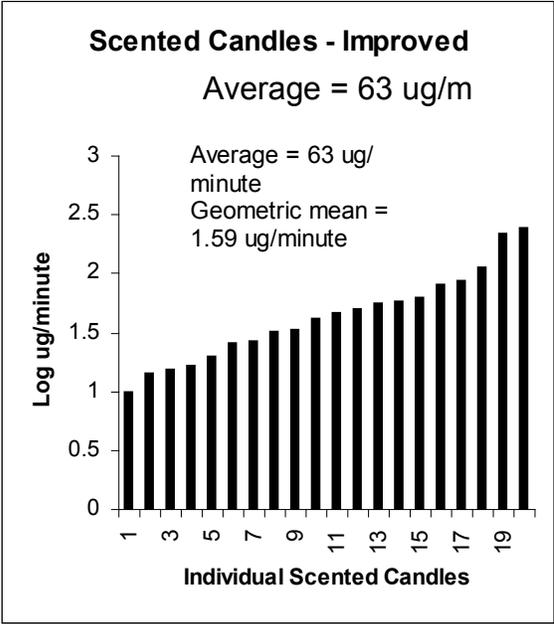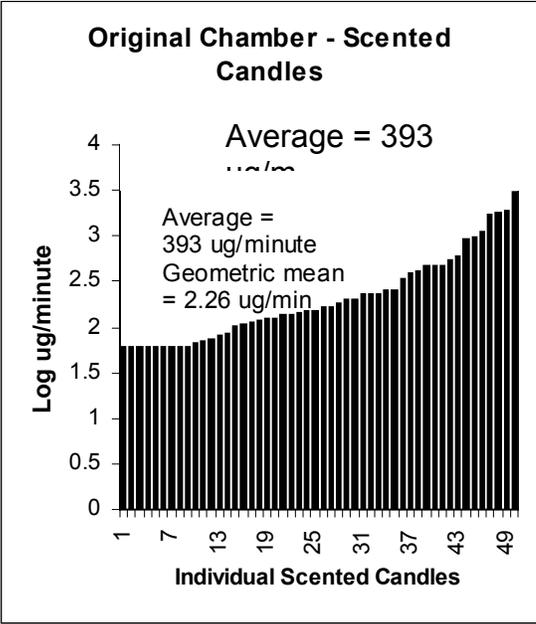
Figures 6a and 6b. Soot generation rates of scented candles in the two types of chambers.

Table 8

t-test results of the unimproved and improved chambers.

Parameter	Chambers	
	Krause Chamber N=50	Silver-Blake Chamber N=20
Mean	393	62.8
Mean $_{geometric}$	2.26	1.59
95% confidence interval for Mean	250.4 thru 535.5	-162.6 thru 288.3
Standard Deviation	594	64.4
High/Low	3.090×10^3/60.0	249/9.87
Median	159	44.5
Average Absolute Deviation from Median	300	39.3
t*	2.47	

Note. *p=0.016

      Sixteen candles were tested for lead and zinc inside the improved chamber. Particle mass concentration averaged 62.8 mg/m^3. Resulting lead concentrations inside the chamber were less than the analytical limit of detection (1 μg) and airborne concentrations inside the chamber averaged 1.14 μg/m^3. It is safe to assume that modeled lead exposures are negligible. Zinc from metal wicks resulted in an average concentration of 0.053 mg/m^3 with a standard deviation of 0.124 mg/m^3. The OSHA permissible exposure limit (PEL) for zinc oxide is 5 mg/m^3.

      OSHA Method 58 PAH analysis of anthracene, benzo[a]pyrene, chrysene, phenanthrane and pyrene resulted in PAH mass on the filters that were less than the limit of detection (3 μg). Airborne concentrations inside the chamber averaged 1.14 μg /m^3. That the levels of PAH in the chamber escaped detection is consistent with other chamber candle emission studies.

Test Room Survey Data

To determine the occupational health risk from candle emission exposures, exposure to ultrafine particles are measured in our test room. The candle material and additives are associated with candle combustion products and soot generation rate. To accurately determine the dose for each specific workplace, candles were chosen based on our survey results. Candle selection was based on typical candles for the three occupations that use the majority of candles.

Candle suppliers provided information on what occupations use candles the most. Candle demographical information allowed the most representative candle types and numbers to use in the study. Maximum numbers of candles that might be used in a workplace were derived from the survey information.

Furnishings were selected that would typically be used in the specific occupational settings. Realistic sinks, airflow rates and room parameters remove conditions that would have to be accounted for with theoretical models. The actual type of furnishings and room conditions allowed for results that are more accurate.

Candle Use Demographics

Candle use data and data generated from the candle emission testing provided evidence that workers are not overexposed in most workplace environments. Only in workplace environments that use an atypical number of candles, does the possibility of a health risk exist. Data from spas, restaurants and churches provided accurate information to estimate ultrafine particle dose (Table 9). Candle number volume density was used to determine candle numbers that applied to an occupational setting. The frequency of exposure was useful in calculating the dose. The data applies to American businesses and churches. Unique situations exist in which a high number of candles may be used in a small room for an extended period such as candles around an icon in a chapel.

Survey data revealed that 72% of restaurant candles are unscented paraffin candles, 77% of spa candles are scented colored paraffin candles, and most of the candles used in churches are tapered paraffin candles either colored or white. The survey shows that each occupation has a strong preference for a particular candle type and allows a better assessment of candle emission risks. Exposure time for restaurants is 6.1 hours, spas; 2.3 hours and churches; 8.0 hours per one Sunday a week.

Table 9

Summary of Candle Use Survey Data

Parameter	Location		
	Restaurant	Spa	Church
Candle Type			
Paraffin	72%	77%	65%
Beeswax	0%	23%	12%
Alcohol	22%	0%	4%
Gel or other	6%	0%	19%
Candle Shape			
Votive / tea light	96%	46%	8%
Tapered	4%	15%	60%
Pillar	0%	38%	32%
Candle Color			
White	100%	15%	52%
Colored	0%	85%	48%
Candle Scent			
Scented	0%	92%	13%
Unscented	100%	8%	87%
Trimmed wicks	0%	15%	0%

Room Dimensions

A random survey of restaurants, churches and spas revealed occupation-specific room sizes and dimensions. Field notes were taken of various businesses and churches located in the Clearwater area. Spas using candles are small, typically consisting of a relaxation table or Jacuzzi surrounded by one or more aromatic candles. Restaurants can be small and have only a few tables, or cavernous with many tables. Restaurants showed the most consistency in table spacing and the use of a single candle on each table. Churches were usually large with high ceilings and, depending on the religion, used varying number of candles based upon the specific event that is taking place.

Occupation Specific Number of Candles

Data on numbers of candles used in a particular type of business or religious institution were based on interviews, general inquiry and observations. Survey results indicated the greatest number consistency with restaurants, some variability with spas, and the widest variability with churches. Church candles in ceremonies, vigils, holidays and religious practices, ranged from one to several hundred. Candle type, number and exposure duration data collected for this study was used to calculate the lifetime average daily dose of candle soot particles.

Most spas used a single candle, however, some used up to ten candles in one session.   With massage therapy, aroma candles are lit throughout day. This was the greatest exposure for a typical spa worker.

Candle types placed on tables inside dining rooms during the dinner hour were unscented paraffin. Table spacing in the room and the number of tables determined the density of candles and candle emissions inside a room. The number of candles lit on tables may be the entire number of candles on tables or only during seating of a customer. Observations of various dining establishments revealed that candle density varied from one to five per hundred square feet of dining space.

The widest variation in candle type and number were with religious institutions. Candles are placed by altars, part of marriage ceremonies, lit to commemorate a special event or burned by the hundreds in a candle vigil (Table 10).

Table 10

Candle number and activities as practiced by various religious institutions.

Religious Institution	Activity	Number of candles
All Churches	There are no strict rules about the number of candles or definite placement.	1-100
All Churches	Candle Vigils	10-1000
Church	Priests place votive or prayer candles near a statue or shrine of a Saint or near the Reserved Sacrament.	1-100
Church	Following in procession, churchgoers light small taper candles off the large one, which eventually lights up the church. Church workers use this candle at baptisms and funerals.	1 per congregant
Synagogue	Lighting of Chanukah candles	1-9
Synagogue	Lighting of Sabbath candles	2-7

Candle Exposure Duration

Candle burn exposure duration depends on the occupation and application. Spa workers light candles in a relaxation room and leave. They return at the end of the session to extinguish the candles. Exposure is limited to a few minutes in these cases. Relaxation sessions last for one to three hours. Exposure to candles while massaging clients may last all day if clients repeatedly desire aromatherapy candles. Dinners in restaurants typically last from 6:00 p.m. to 10:00 p.m. and catered events last up to six hours. Church workers may be

exposed to candles for an entire Sunday or intermittently during the week, depending on the occasion.

Particle Distribution Dynamics Analyzed with Optical Particle Counters in the Unventilated Test Room

The particle size distribution dynamics observed from measurements in the unventilated test room were lacking information on particle size dynamics beyond the limit of the condensation nuclei counter. Two size ranged optical particle counters in conjunction with the condensation nuclei ultrafine particle measurements during candle burns allowed us to observe the particle size dynamics in real time.

The MetOne particle counter particle size ranges between 0.1 to 1 μm, whereas the Lasair ranged from 0.5 to 10 μm. OPC testing occurred before candles were lit (background) and at peak CNC concentrations (before extinguishing). See Figures 8-13.

Test results were limited by the inherent problems with the optical particles measuring non-spherical particles. Soot nuclei grow into chain aggregates that take on a very irregular structure with branching chains. Andersen impactors, scanning electron microscopy, microscopy-sizing methods provide more accurate measurements of agglomerate particle size concentrations. OPC data did provide some useful information on the ambient environment. The ambient background particle size distribution is mostly those particles less than 0.1 μm. Roughly, 90% of the particles are in the 0.1-0.2 μm range. Particles sized 0.3 to 1 μm, contribute little to the distribution (Figure 7).
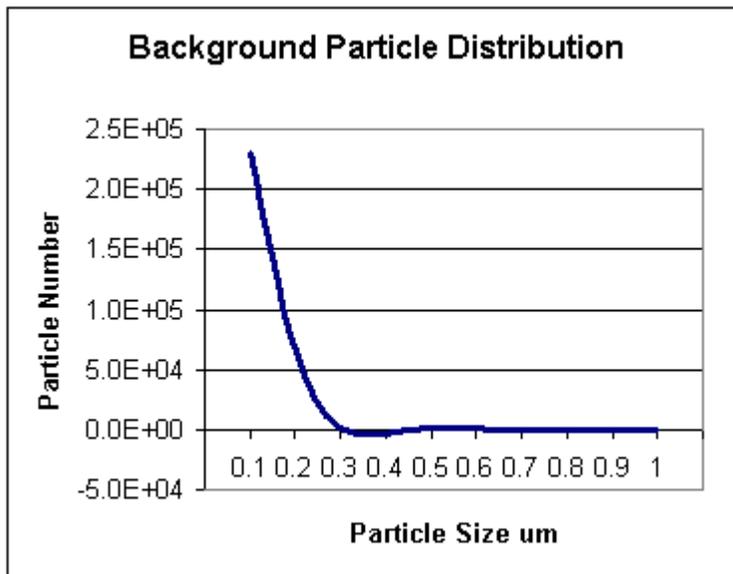
Figure 7. Background Particle Size Distribution in the Clearwater area.

53

Simultaneous measurement of candle particle emissions were conducted in the unventilated test room environment using the following instrumentation:

- Condensation nuclei counter (CNC) capable of measuring between 20 nm to 1 μm sized particles.
- MetOne 237H optical particle counter capable of measuring between 0.1 μm to 1 μm sized particles.
- Lasair 510 optical particle counter capable of measuring between 0.5 μm to 10 μm sized particles.

When the peak reading from the CNC was taken, readings from the MetOne 237H and Lasair 510 were recorded. Readings were compared to the initial background concentration. Observations indicated a decrease in sizes ranging from 0.1-1 μm and an increase from 1-10 μm. During peak CNC particle concentrations, nucleation particles dominate. Gas to particle conversion explains the generation of smaller particles not seen by the MetOne 237H, however as the particles undergo condensation and ultimately coagulation, increases can be observed for particles greater than 1 μm.

After the candle has peaked and the CNC readings diminish by more than half, the candle is still generating nucleation particles, however, the overall concentration of particles in the room reflects a faster coagulation time. The decrease from 0.1-0.2 μm reflects similar diminished CNC readings. The increases in particle size ranges greater than 0.3 μm reflects continued coagulation.
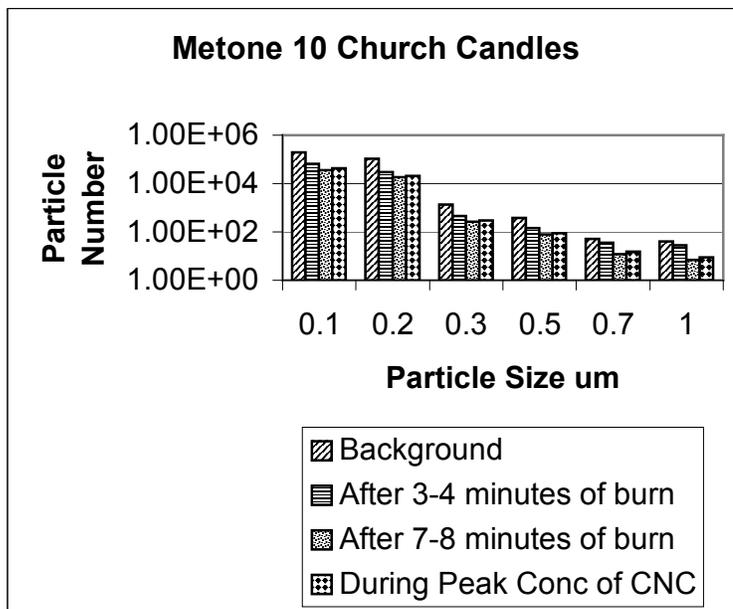
Figure 8. Ten-candle burn with MetOne analysis.

54

Lasair 10 Church Candles

Figure: Bar chart with y-axis "Particle Number" (logarithmic scale from 1.00E+00 to 1.00E+06) and x-axis "Particle Size um" (values 0.5, 1, 2, 3, 4, 5, 7, 10). Legend: Background, After 3-4 minutes of burn, After 7-8 minutes of burn, At Peak CNC Concentration.
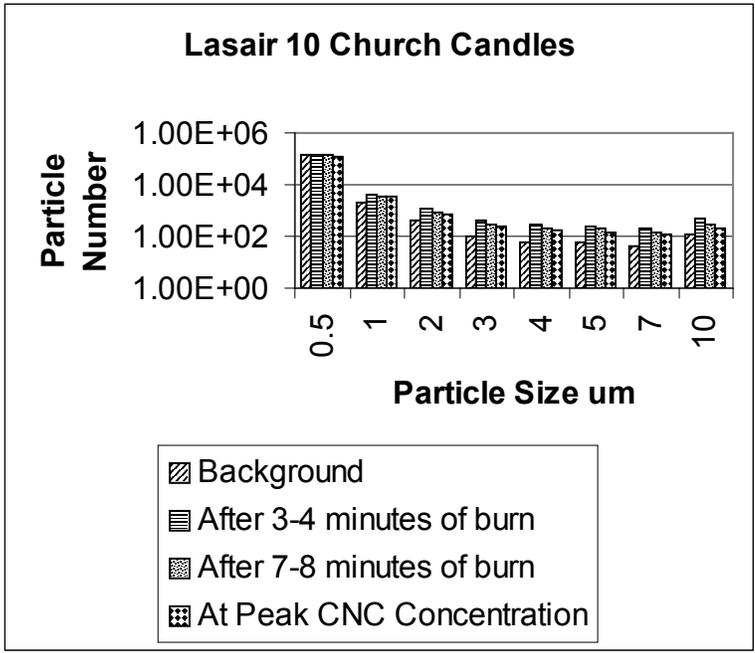
Figure 9. Ten-church candle burn with Lasair analysis.

Metone - 1 Church Candle

Figure: Bar chart with y-axis "Particle Number" (logarithmic scale from 1.00E+00 to 1.00E+06) and x-axis "Particle Size um" (values 0.1, 0.2, 0.3, 0.5, 0.7, 1). Legend: Background, Before Extinguishment.
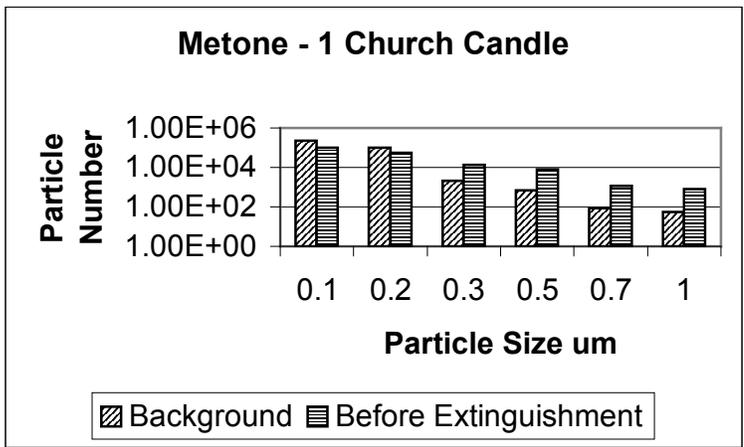
Figure 10. Single candle burn with MetOne analysis.

55
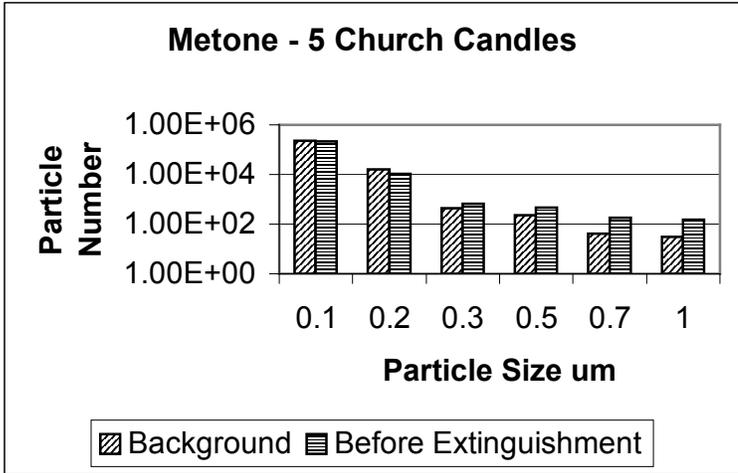
Figure 11. Five-candle burn with MetOne analysis.
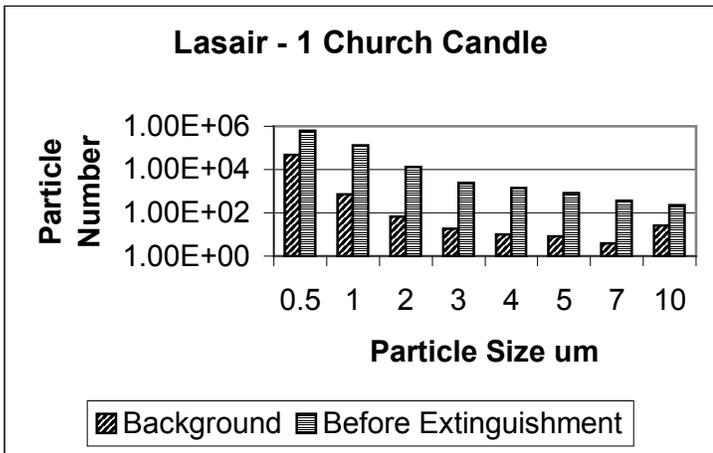
Figure 12. Single candle burn with Lasair analysis.
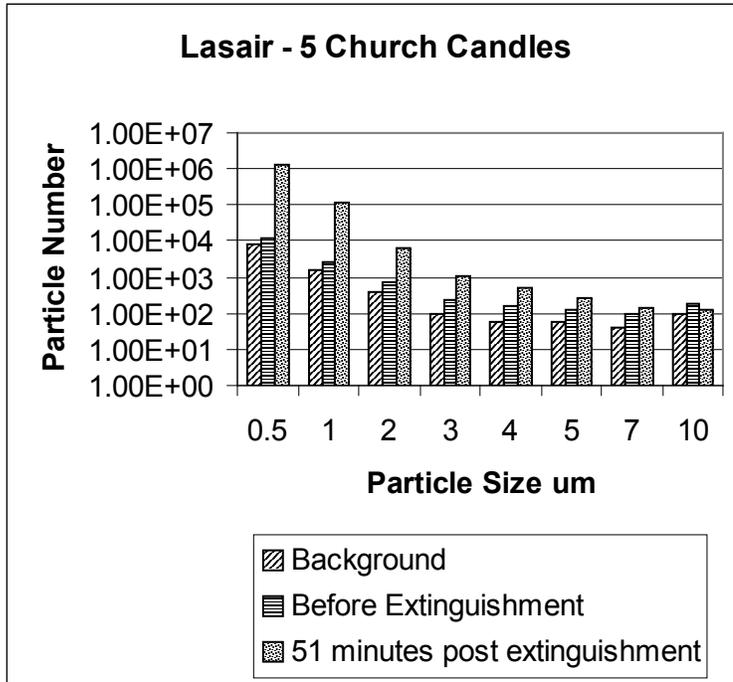
Figure 13. Five-candle burn with Lasair analysis.

Unventilated Test Room Particle Measurements

Simulated room particle data provided a more accurate estimation of particle exposure than a chamber. Accuracy is demonstrated by uncertainties in model extrapolation, unknown particle size distribution in chambers and simulating the actual occupational environment. Environmental parameters for the test room were a constant 24-26°C and 52-67% relative humidity.

Unventilated work environments occurring in locations where moderate temperatures are expected in some cases. In unventilated environments, filtration and air exchanges do not occur and the exchange of airborne materials with the outside by natural ventilation depends on building envelope openings. A "tight" room was assumed where the room is sealed off completely to the outside and other rooms in the building. Sealing the room allowed for control of natural ventilation variables. Gaseous pollutants rely on generation and purging to control the concentration at any given time. Without purging the room through exhaust or air changes, concentrations of gases can build continuously to saturate the air. In a sealed room, gas laws do not govern particle removal mechanisms. Gravitational settling, coagulation, diffusion, and thermophoresis serve to remove particle concentrations.

Ten church candles were burned in the sealed, unventilated test room. Particle count measurements were recorded in minute increments using the

57

CNC. At the beginning of the candle burn, the particle number concentration rose rapidly to a peak level and then decayed at a slightly slower rate. The ultrafine particle concentration continued to decline for several hours close to background levels. The peak occurred long before the end of the candle burn, yet had not been expected until the end.

The first phase of candle soot generation is the nucleation mode where these small particles buildup rapidly. The ultrafine particle concentration peaks as particle size growth occurs with agglomeration of the nuclei into larger sized chain aggregates. The nuclei particles are able to penetrate into the alveolar regions of the lung, whereas, the larger agglomerates are deposited in the upper branches based on size. Without the removal dynamics of filtration, emitted ultrafine particles quickly coagulate with surrounding particles and the ultrafine particle count diminishes with time. The larger sized particles are in the accumulation mode and are beyond the size range that can be detected by the CNC. Some nuclei particles attach to larger particles, the other fraction is lost by deposition and diffusion. When the candle flame was extinguished, a small second peak appeared and then returned to the original rate of decrease. Extinguishment caused a second nucleation mode to occur and this new generation of nuclei was swept up by coagulation forces (Figure 14).
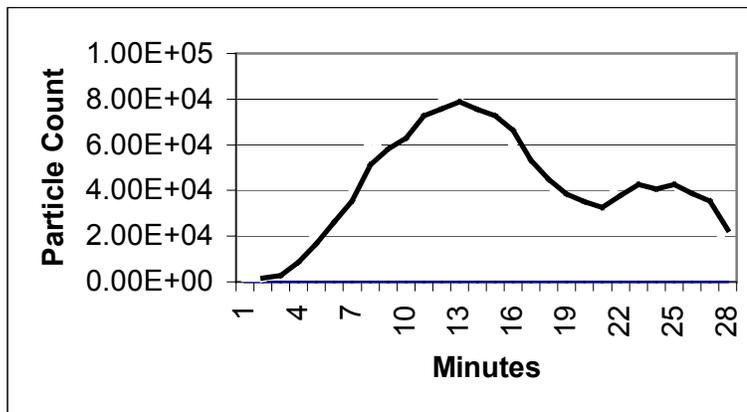
Figure 14. Graph of 10-candle burn inside non-ventilated test room.

The ten-candle burn demonstrated the dynamics of particle size distribution in real time. In subsequent testing, the effects of varying the number and types of candles were investigated. Logged particle concentration data of single, five and ten candle burns, proved the quick build up and eventual decay held true regardless of candle number (Figures 15, 16, and 17).
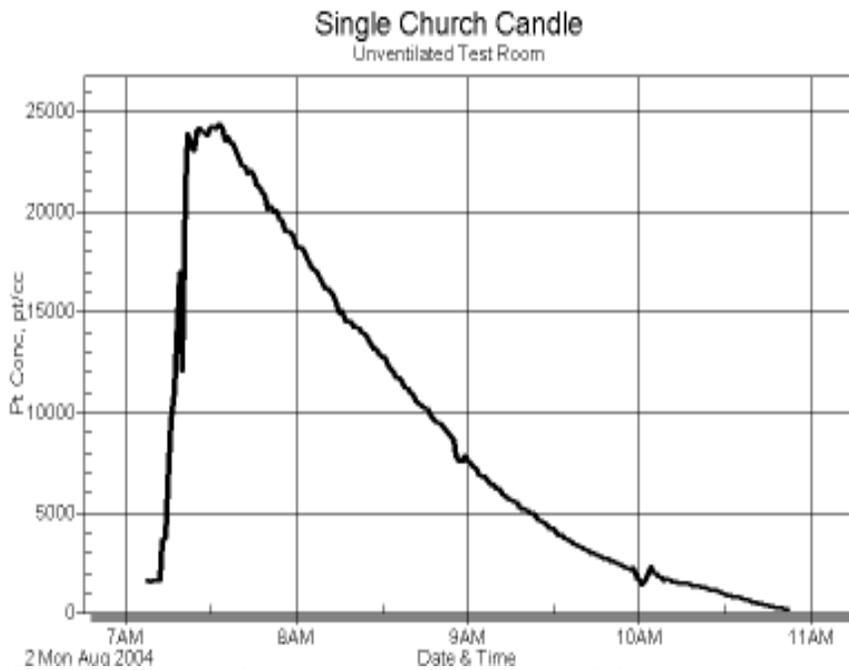
Figure 15. Graph of one-candle soot particle count generation rate, unventilated test conditions**.**
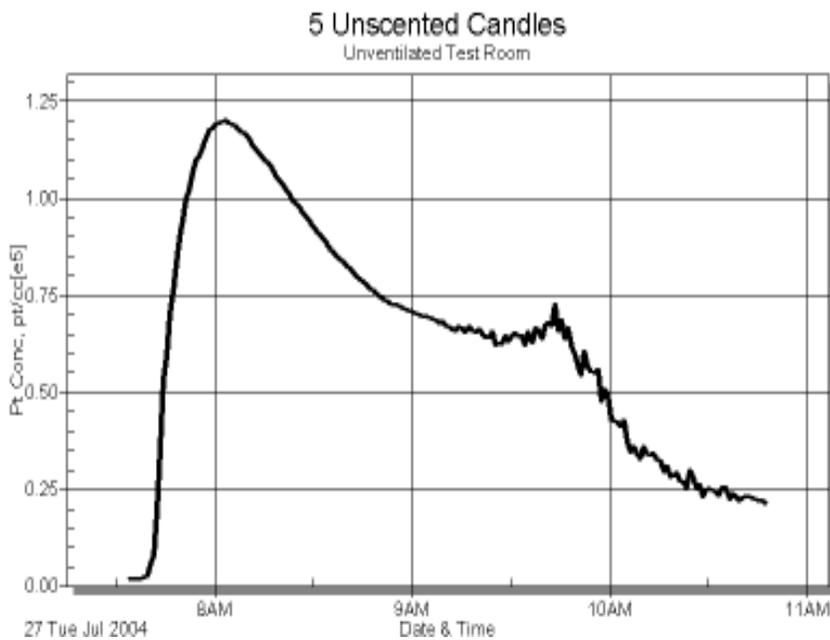
Figure 16. Graph of five-candle soot particle count generation rate, unventilated test conditions.
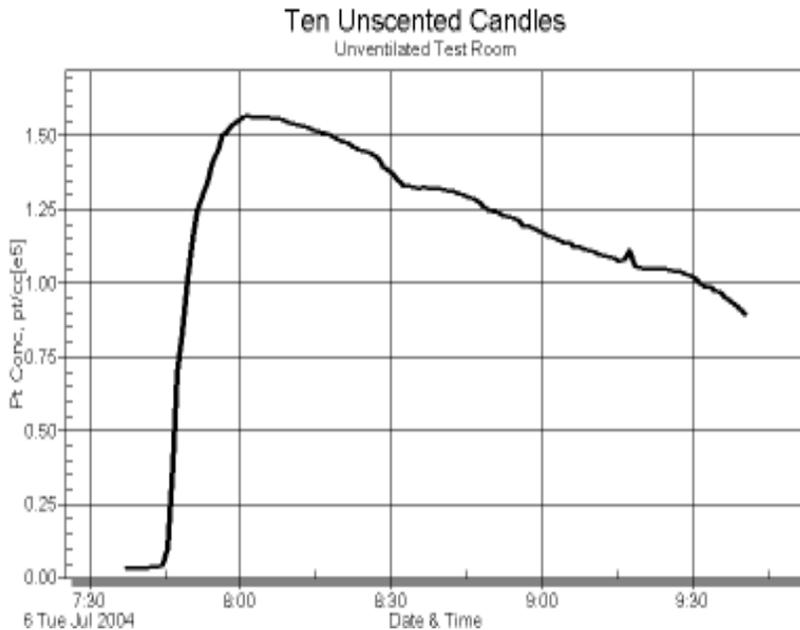
Figure 17. Graph of ten-candle soot particle count generation rate, unventilated test conditions.

Increasing the candle number in the unventilated sealed test room, increased the peak concentration of nuclei particles. With increasing number and different types of candles, a rapid peak maximum occurred followed by a slower decline and the second peak during extinguishments. During the testing, temperature, humidity and furnishings remained constant, the only variable to change was the candle number and type. This allowed for a more sensitive analysis of candle effect because furnishings, ventilation, temperature and relative humidity were a fixed constant.

Candle Soot Generation Rate Differences between Unscented, Scented and Church Candles

One-way ANOVA is used to test the hypothesis that particle concentration depends on candle types. For example, church candles may differ in the amount of soot generated than from scented candles. To test this hypothesis, our analysis includes three candle sets (1, 5 and 10 candles) of three different types of candles. Group *A* is the unscented candles, group *B* is the church candles; group *C* is the unscented candles. In detail, each group is probably different: has slightly different highs, lows, and hence it is likely that each group has a different average (mean) size. Can we take this difference in average size as evidence that the groups in fact are different (and perhaps that candle type causes that difference)? Note that even if there is *not* a "real" effect of candle type (the null hypothesis), the groups are likely to have different average particle concentrations. The likely range of variation of the averages if candle type-effect hypothesis is wrong, and the null hypothesis is correct, is given by the standard deviation of the estimated means.

60

Particle count data from same number sets of unscented, scented and church candles were tested for differences using 1-way analysis of variance. The F statistic was relatively small, indicating very little difference between candle sets and acceptance of the null hypothesis. One-way analysis of variance (ANOVA analysis) revealed no significant variation between types of candles and particle concentration (Table 11).

Table 11

ANOVA of Scented, Church and Unscented Candles

Source of Variation	Statistical Parameter			
	Sum of Squares	Degrees of freedom	Mean Squares	F
Single Candle – Unscented, Church, Scented				
Between	1.6694×10^8	2	8.3468×10^7	0.4717*
Error	3.7158×10^9	21	1.7694×10^8	
total	3.8827×10^9	23		
Five Candles – Unscented, Church, Scented				
Between	9.3318×10^8	2	4.6659×10^8	0.8942**
Error	1.0958×10^0	21	5.2179×10^8	
total	1.1891×10^0	23		
Ten Candles – Unscented, Church, Scented				
Between	3.1935×10^9	2	1.5967×10^9	1.086***
Error	3.0888×10^0	21	1.4709×10^9	
total	3.4082×10^0	23		

Note. *p=0.63, **p=0.42, ***p=0.36

Candle Soot Generation Rate Differences Between Candle Set Numbers

Particle count data from same number sets of unscented, scented and church candles were tested for differences using 1-way analysis of variance. The F statistic was much greater than one, indicating significant difference between candle sets. One-way analysis of variance (ANOVA analysis) revealed significant variation between numbers of candles and particle concentration (Table 12).

Table 12

Analysis of Variance (ANOVA) between candle types

| | Statistical Parameter | | | |
Source of Variation	Sum of Squares	Degrees of freedom	Mean Squares	F
Unscented Candles – Sets of 1, 5 and 10 candles				
Between	4.7035×10^0	2	2.3518×10^0	51.12*
Error	9.6604×10^9	21	4.6002×10^8	
total	5.6696×10^0	23		
Church Candles – Sets of 1, 5 and 10 candles				
Between	5.1729×10^0	2	2.5864×10^0	39.14**
Error	1.3879×10^0	21	6.6088×10^8	
total	6.5607×10^0	23		
Scented Candles – Sets of 1, 5 and 10 candles				
Between	6.8828×10^0	2	3.4414×10^0	32.82***
Error	2.2023×10^0	21	1.0487×10^9	
total	9.0851×10^0	23		

Note. Candle type and particle concentration based on the number of candles are statistically dependent. *$p<.0001$, **$p<.0001$, ***$p=0.36$

Linearity of Peak Concentrations with Varying Candle Number in a Fixed Volume Room

Linearity of peak concentrations with candle numbers was tested. Candle numbers 1, 5, 10, and 20, with scented, unscented, and church candles, were plotted against peak concentration (Figures 18-20). The relationship was linear and allowed for prediction of peak concentrations with candle number. With the three candle types, peak concentrations increased with candle number in a 2-3 minute time range, and then the concentrations decreased to background levels in less than 30 minutes. The exposure to the ultrafine particles was minimized by the presence of larger agglomerated particles and their ability to quickly pick up the nuclei particles as they were generated. Our results imply that occupational exposure to ultrafine particle concentrations from candle sets in an unventilated test room is brief for the several minutes of peak exposures and approaches background levels over several hours. Continued averaging after the peak diminishes over time, minimizing exposure as more time passes after the peak. There is less exposure and less risk, the further time passes after the peak.
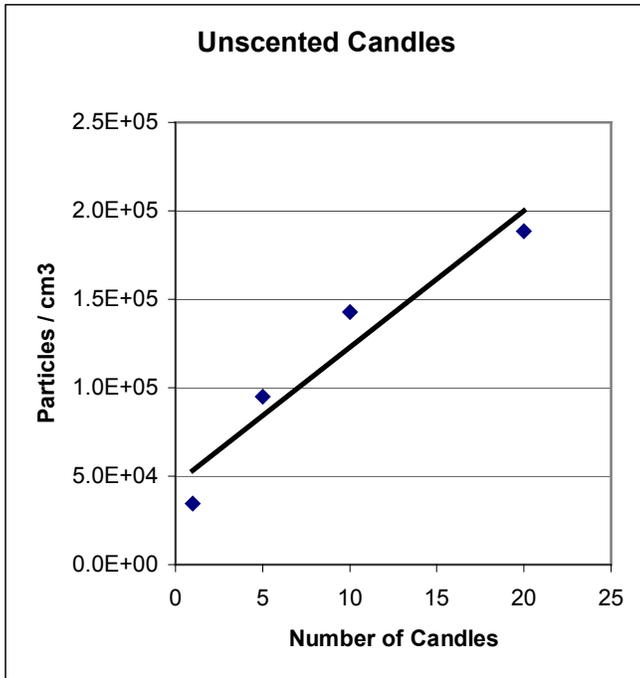
Figure 18. Regression analysis of unscented candles' particle number concentration with candle number, unventilated test conditions (R=0.96)
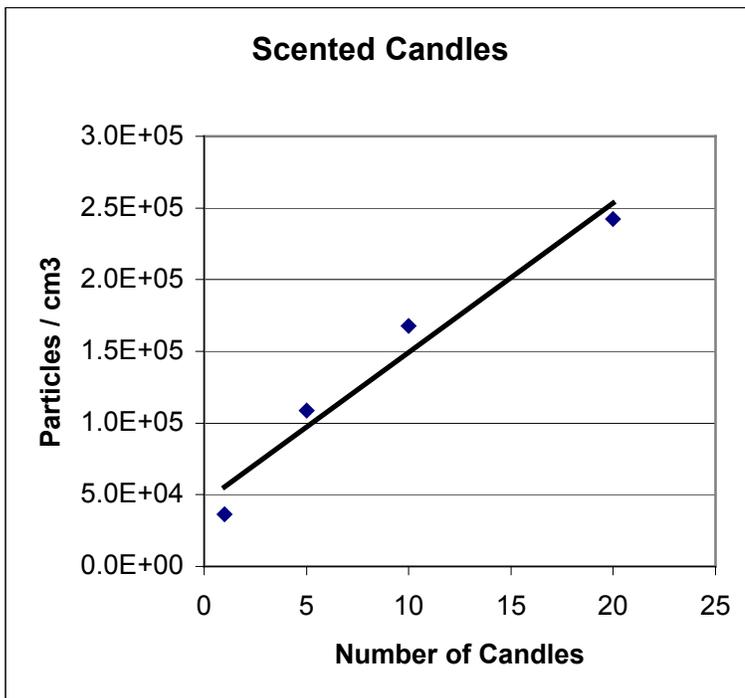
Figure 19. Regression analysis of scented candles particle number concentration with candle number, unventilated test conditions. (R=0.98)
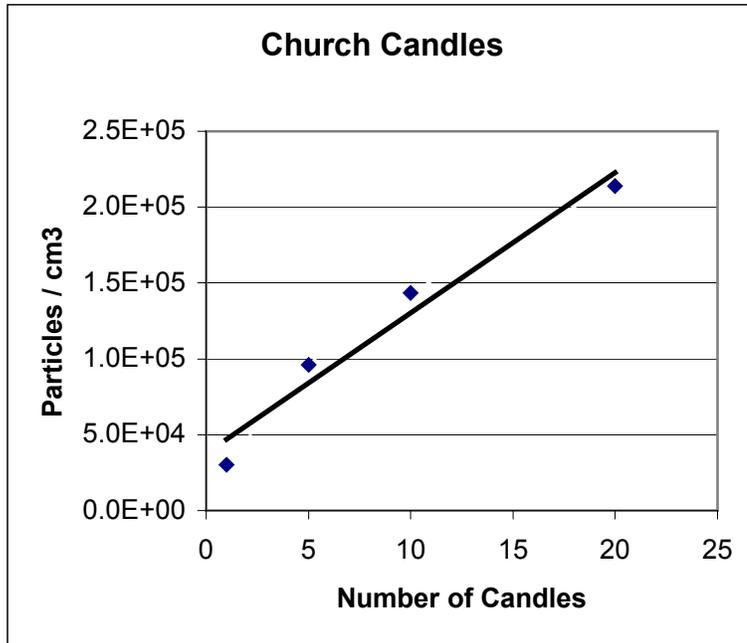
Figure 20. Regression analysis of church candles particle number concentration with candle number, unventilated test conditions. (R=0.98)

Ventilated Test Room Particle Measurements

Simulated room particle data in a ventilated room provided the most common workplace condition in workplaces. Environmental parameters for the test room were a constant 22-23°C and 45-50% relative humidity.

In ventilated environments, filtration and air exchanges condition and clean the air inside the room. To provide a typical ventilated environment, the door or window was not sealed, however, the door was kept closed. Graphical representation of particle counts in a ventilated test room illustrated the effect of an air handler and filter on peak concentrations, steady state conditions and coagulation (Figures 21, 22, 23, and 24). A cyclic relation of repeat particle buildup and then decay occurred over time in the ventilated test room. As with the sealed room, particle number concentration quickly rose to a peak and then decayed. When the air handler unit started, the particle decay rate transitioned to increased particle concentration generation. Larger particles were removed by filtration and allowed for the generation of more nucleation mode particles without the immediate effects of coagulation. There is more space between nucleation mode particles. The number of air changes causes steady state conditions. The peak particle number concentration is roughly half the concentration in the sealed room.

64
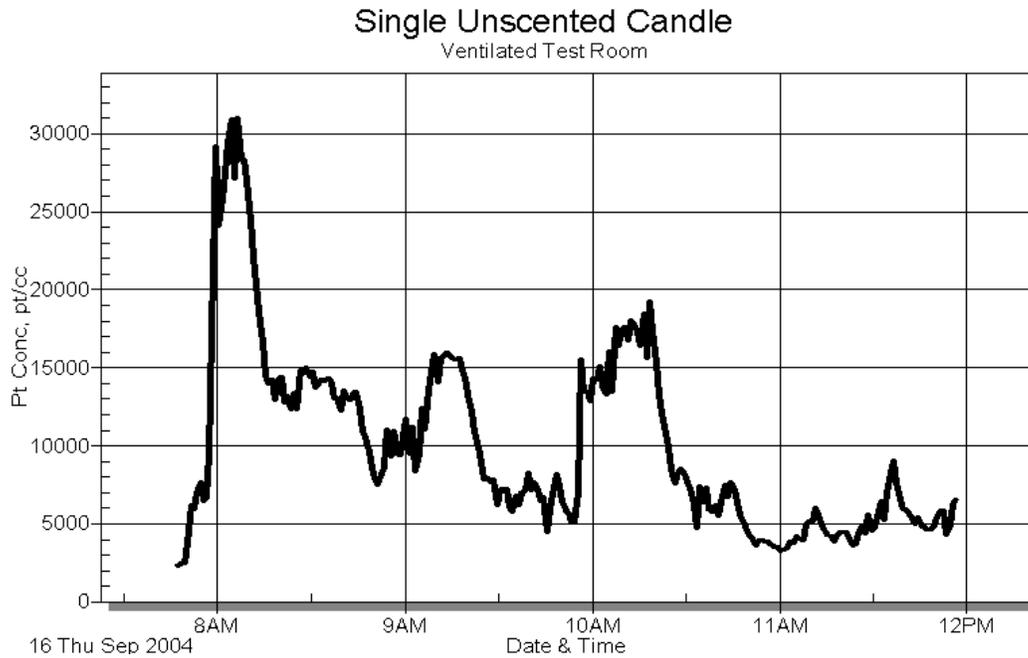
Figure 21. Single-candle burn, ventilated test conditions. Average = 10,102 particles/cm^3.
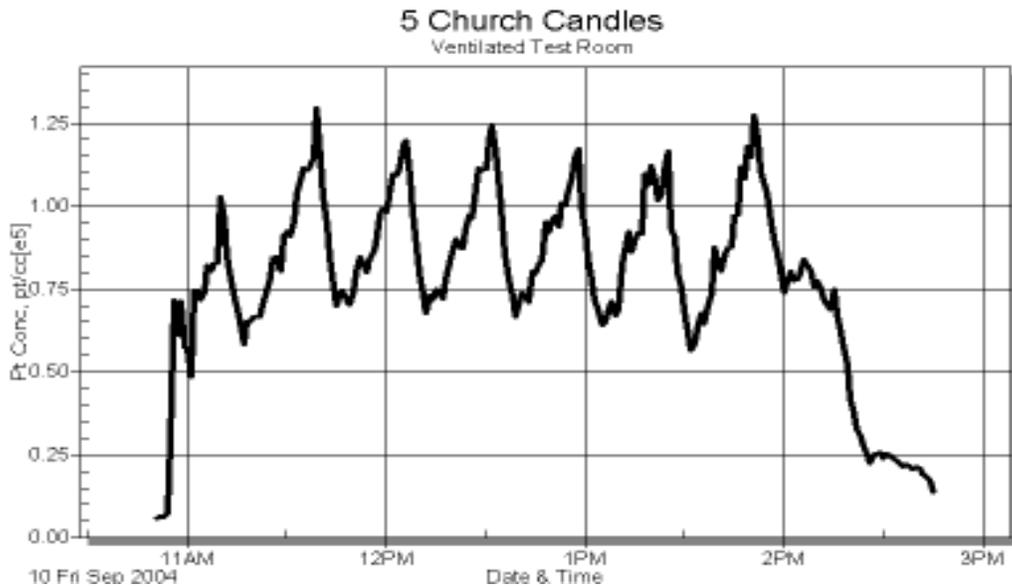
Figure 22. Five-candle burn, ventilated test conditions. Average = 77,851 particles/cm^3.

65
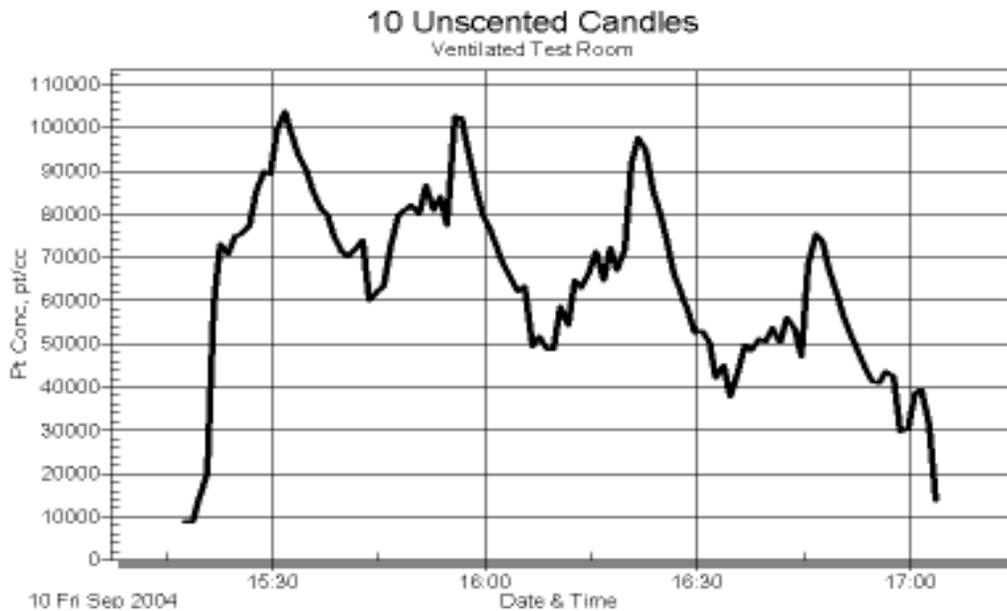
Figure 23. Ten-candle burn, ventilated test conditions.. Average = 64,219 particles/cm^3.
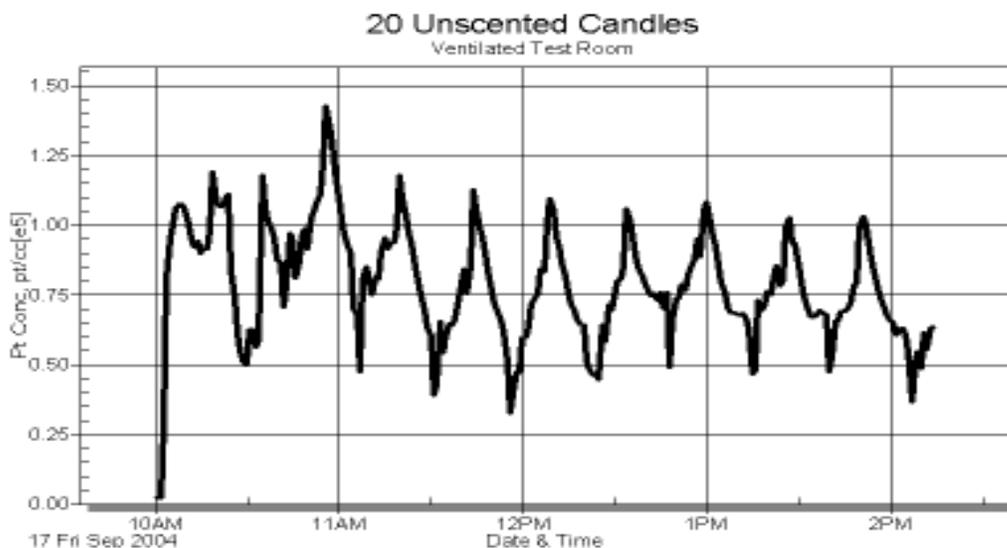
Figure 24. Twenty-candle burn, ventilated test conditions. Average = 79,044 particles/cm^3. The peaks and valleys along a horizontal line display a steady state concentration. Experimental results show the same peak concentrations with ventilated and non-ventilated rooms.

Candle Soot Generation Rate Differences among Unscented, Scented and Church Candles

Particle count data from same number sets of unscented, scented and church candles were tested for differences using 1-way analysis of variance. The F statistic was relatively small, indicating very little difference between

66

candle sets. One-way analysis of variance (ANOVA analysis) revealed no significant variation between types of candles and particle concentration (Table 13).

Table 13

ANOVA, differences among types of candles

Source of Variation	Sum of Squares	Degrees of freedom	Mean Squares	F
		Statistical Parameter		
	Five Ventilated – Unscented, Church, Scented			
Between	1.0105×10^8	2	5.0525×10^7	0.2413*
Error	4.3969×10^9	21	2.0938×10^8	
total	4.4979×10^9	23		
	Ten Ventilated Candles – Unscented, Church, Scented			
Between	3.0549×10^8	2	1.5274×10^8	0.8828**
Error	3.6334×10^9	21	1.7302×10^8	
total	3.9389×10^9	23		
	20 Ventilated Candles – Unscented, Church, Scented			
Between	1.9304×10^7	2	9.6521×10^6	0.0283***
Error	7.1654×10^9	21	3.4121×10^8	
total	7.1847×10^9	23		

*p=0.79, **p=0.43, ***p=0.97

Candle Soot Generation Rate Differences among Candle Set Numbers

Particle count data from same number sets of unscented, scented and church candles were tested for differences using 1-way analysis of variance. The F statistic was much greater than one, indicating significant difference between candle sets. One-way analysis of variance (ANOVA analysis) revealed significant variation between types of candles and particle concentration (Table 14).

Table 14

ANOVA, differences among 5, 10 and 20 candles

Source of Variation	Statistical Parameter			
	Sum of Squares	Degrees of freedom	Mean Squares	F
Unscented, 5, 10, 20 candles				
Between	6.9177×10^9	2	3.4589×10^9	12.21*
Error	5.9513×10^9	21	2.8339×10^8	
total	1.2869×10^{10}	23		
Church, 5, 10, 20 candles				
Between	6.2769×10^9	2	3.1385×10^9	15.91**
Error	4.1435×10^9	21	1.9731×10^8	
total	1.0420×10^{10}	23		
Scented, 5, 10, 20 candles				
Between	6.0854×10^9	2	3.0427×10^9	12.53***
Error	5.1010×10^9	21	2.4290×10^8	
total	1.1186×10^{10}	23		

Note. $*p=.0003$, $**p<.0001$, $***p=.0003$

Linearity of Candle Number with Particle Concentrations

Differences in steady state concentrations occurred with candle types, candle numbers, room volume, ventilation and sinks. Varying the ventilation rate and the number of candles had more of an effect on particle concentration than the type of candle. Average particle concentration data from four sets of numbered candle data were plotted against particle number concentration. The data is linear and useful for predicting the increase in steady state concentration with candle number (Figures 25-27).
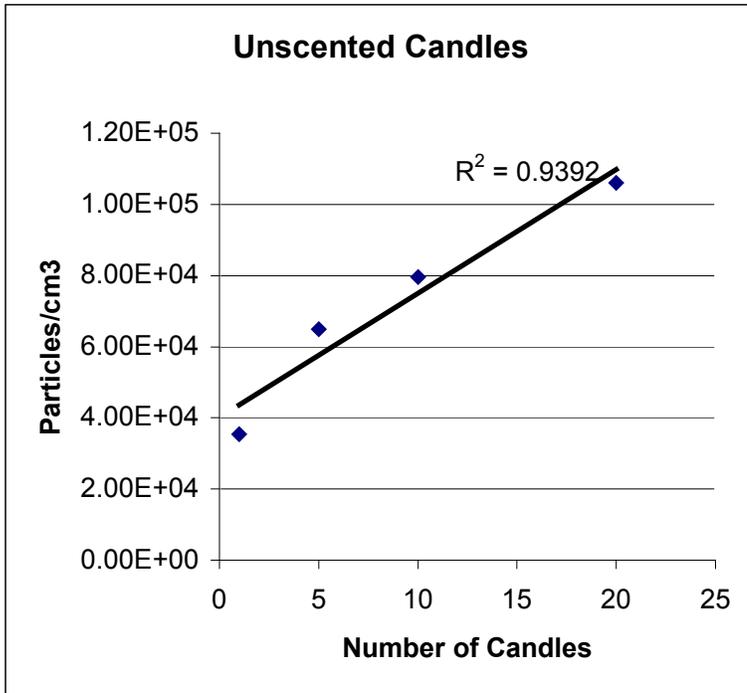
Figure 25. Regression analysis of unscented candles particle number concentration with candle number, ventilated test conditions. (R=0.97)
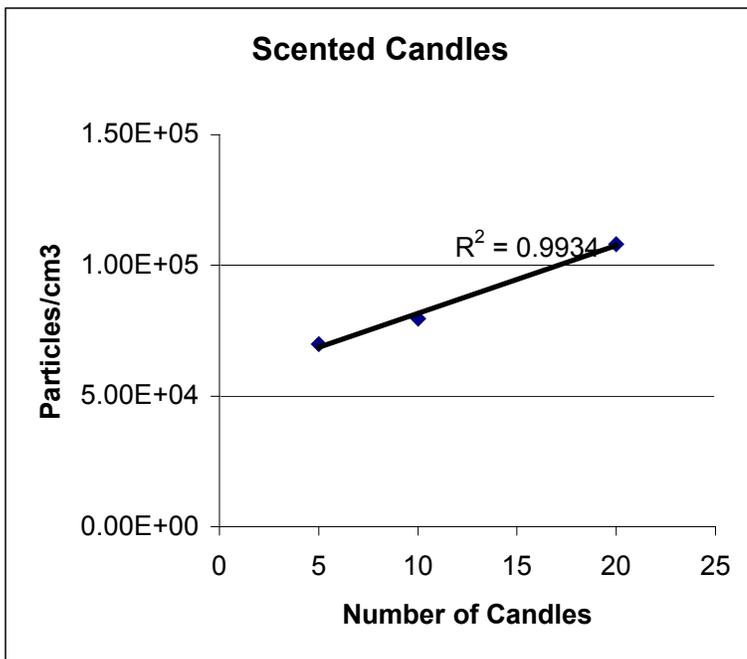
Figure 26. Regression analysis of scented candles particle number concentration with candle number, ventilated test conditions. (R=0.997)
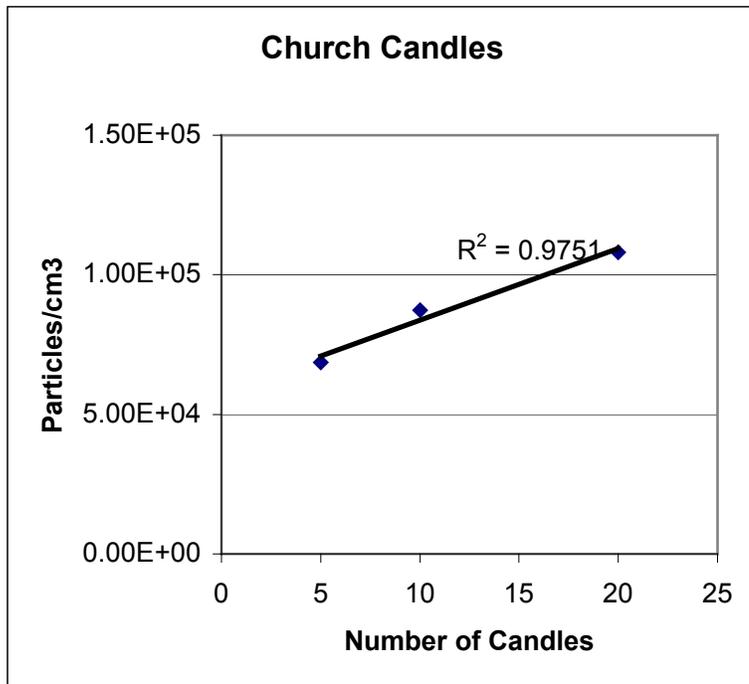
Figure 27. Regression analysis of church candles particle number concentration with candle number, ventilated test conditions.   (R=0.99)

Testing the Difference of Soot Generation between Scented and Unscented Candles in our Simulated Work Environment

Scented candles generated more soot than unscented candles when burned inside a chamber (Krause et al., 1999).   Candle generated soot was collected, analyzed and reported in terms of µg/min per wick, for both scented and unscented candles. Krause reported that the unscented candles mean rate was 83 µg/min-wick with range 20-175 µg/min-wick; Krause's scented candles mode was 180 µg/min-wick, and mean 165 µg/min-wick with range 20-3100 µg/min-wick.

Ten unscented paraffin candles and ten strongly scented paraffin candles were burned inside the test room to determine the validity of results from chamber studies. Mean differences (unscented; 1.183×10^5, scented 2.021×10^5 particles /cm^3) were tested with a two-tailed t-test, t=-8.25, 95% C.L. (Table 15). Test room particle concentration results provided evidence that the difference in means was statistically significant.   Figure 28 depicts the differences between the two types of candles.

Table 15

t-test results between test room data of scented and unscented candles

Parameter	Candle Types	
	Unscented N=10	Yankee Scented N=10
Mean	1.183×10^5	2.021×10^5
95% confidence interval for Mean	1.0321×10^5 thru 1.3337×10^5	1.8699×10^5 thru 2.1715×10^5
Standard Deviation	2.583×10^4	1.906×10^4
High/Low	$1.657 \times 10^5 / 7.672 \times 10^4$	$2.318 \times 10^5 / 1.750 \times 10^5$
Median	1.184×10^5	2.014×10^5
Average Absolute Deviation from Median	1.968×10^4	1.508×10^4
t*	-8.25	

Note. *$p<0.0001$
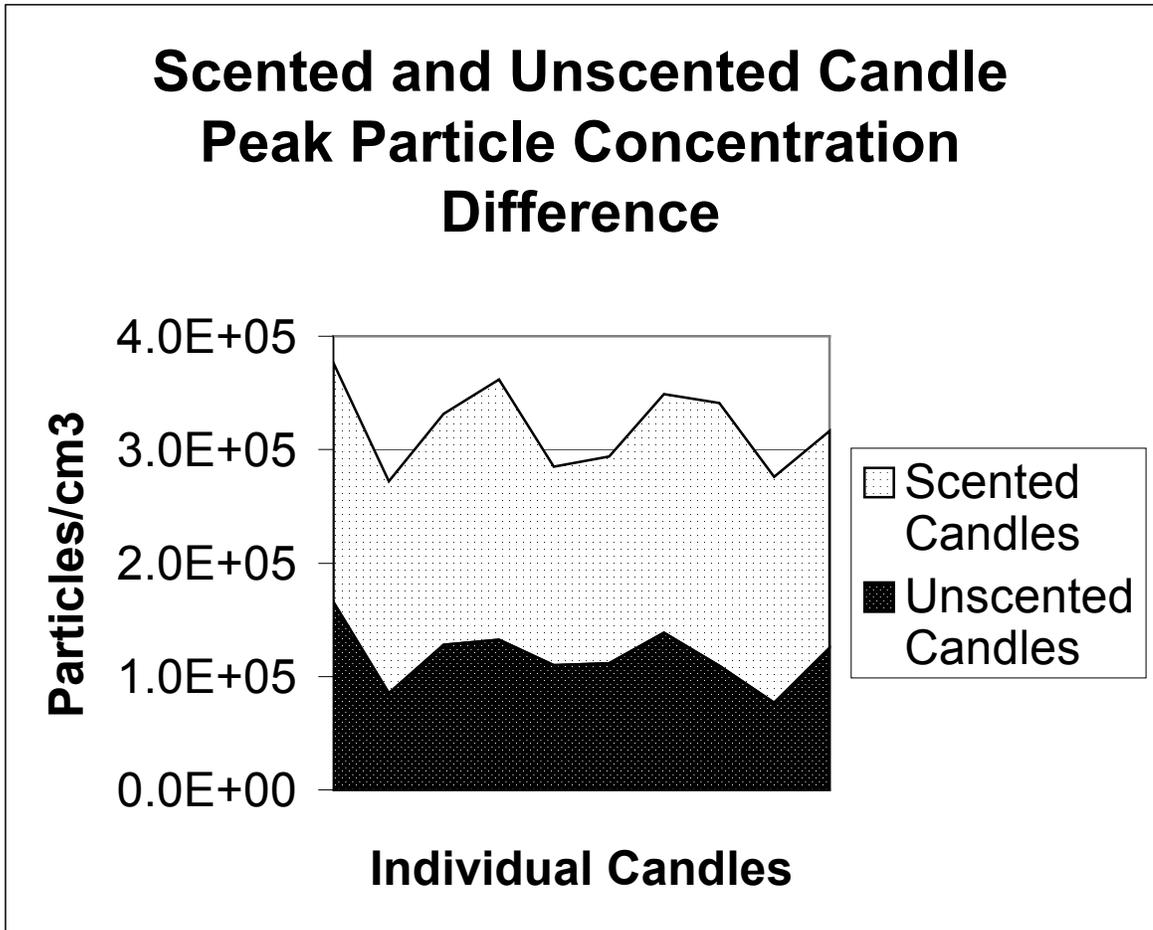
Figure 28. Scented and Unscented Candle Soot Peak Particle Concentration
Difference

Data from PAH Analysis

To determine the concentration of PAHs that are emitted in a test room, a
relatively large number of candles were burned in the sealed test room. Groups
of ten scented, unscented and church candles were selected for testing and
comparison of analytical results to OSHA Permissible Exposure Limits. OSHA
regulates and reports five carcinogenic PAHs as coal tar pitch volatiles (CTPV).

Sets of ten scented, unscented and church candles were burned inside a
40 m^3 non-ventilated, sealed room creating an atypical maximum exposure of an
occupational setting. Typically, most workplaces use fewer candles in a
ventilated volume of air. OSHA Method 58 for the sampling and analysis of
PAHs recommends 960 liters of air at 2 liters per minute. OSHA uses this
method to average an 8-hour period to determine the time weighted average
concentration for workers. In the study, modification of the sampling method took
advantage of the limited burn time and expected lower concentrations by

72

increasing the flow rate and sample volume. Schneider Laboratories of Richmond, Virginia, an AIHA accredited laboratory, analyzed the samples.

Soot was collected on glass fiber filters (GFF) by drawing a volume of air through pre-calibrated pumps. The laboratory's two-step analysis first measured the mass of the benzene-soluble fraction (BSF) by extracting with benzene.   If the BSF exceeds the appropriate PEL, specific PAHs are analyzed.   The lab analyzed the sample by high performance liquid chromatography (HPLC) with a fluorescence (μL) or ultraviolet (UV) detector to determine the presence of selected polynuclear aromatic hydrocarbons (PAHs). Each GFF was transferred to a separate scintillation vial after sampling and the vial sealed with a PTFE-lined cap. Samples were protected from direct sunlight. The method was validated to the established evaluation procedures of the Organic Methods Evaluation Branch of OSHA. Laboratory analytical data revealed that airborne concentrations of PAHs from candles burned in our test room are less than OSHA target concentrations (CTPV).

Laboratory results revealed levels below the limit of quantification.   Tables 16 and 17 list the target concentrations of the five PAH carcinogens. The time-weighted exposure for each candle resulted in levels less than the target concentrations.

Table 16

Actual exposure to coal tar pitch volatiles is less than 200 μg/m^3

Sample Type	Flow Rate (L/min)	Sample Volume Liters	Total Sample CTPV mg	8-hour TWA μg/m^3
Background-Inside Test Room	7.30	3416.4	<0.05	<14.2
Background – Outside	7.50	1837.5	<0.05	<14.9
10 Unscented Candles	7.91	1716.5	<0.05	<13.2
10 Scented Candles	7.87	1936.0	<0.05	<13.2
10 Church Candles	8.14	2271.1	<0.05	<12.8
Field Blank	N/A	N/A	<0.05	

Table 17

Target concentration of CTPV is less than 200 µg/m³

Analyte	Target Concentration
Coal Tar Pitch Volatiles (PEL)	0.20 mg/m³ (200 µg/m³)
Coke Oven Emissions (PEL)	0.15 mg/m³ (150 µg/m³)
Phenanthrene	8.88 µg/m³
Anthracene	0.79 µg/m³
Pyrene	9.00 µg/m³
Chrysene	3.27 µg/m³
Benzo(a)pyrene	2.49 µg/m³

Comparative Risk Analysis, Candle Soot Dose and Ambient Ultrafine Particle Dose

Using EPA human factor data, Lifetime Average Daily Dose (LADD) was calculated for candle use occupations versus ambient ultrafine particle concentrations. LADD refers to the amount of material absorbed by a person throughout a lifetime of exposure. LADDs are calculated from exposure duration, absorption rates, life expectancy and body weight.

The LADDs for candle soot are calculated with occupational exposure concentration and exposure duration, worklife expectancy, life expectancy and human factor data for inhalation rate based on physical activity and assumed adult age. Ambient soot LADD is calculated based on ambient ultrafine particle concentrations and average adult daily inhalation volume for a 24 hour day throughout a life expectancy of 70 years.

Candle measurement data is presented in Table 18 for both ventilated and unventilated test rooms.

Candle UFP Data

Table 18

Ultrafine particle number concentration for candle types, candle numbers and environmental conditions

Candle Type and Numbers (8 trials)	Test Room			
	Unventilated		Ventilated	
	Average Ultrafine Particle Number / cm^3	Standard Deviation	Average Ultrafine Particle Number / cm^3	Standard Deviation
1 Unscented	34512	12754		
5 Unscented	94919	27300	64937	16924
10 Unscented	142705	21729	79667	10167
20 Unscented			105982	21457
1 Church	30269	7579		
5 Church	95890	19441	68550	11058
10 Church	143513	39335	87257	16100
20 Church			108143	13569
1 Scented	36609	17627		
5 Scented	108605	21028	69769	13856
10 Scented	167568	48920	79710	11701
20 Scented			107403	17570

Ambient UFP Data

Ambient UFP exposure concentration was obtained from both literature values and measurement data taken in the Clearwater, Florida area.

Measurements of particle count data from various locations in Clearwater, Florida (Figure 29) averaged 20,281 particles/cm^3 with a range of 6017 to 116,500 particles/cm^3. Clearwater represents a busy urban area characterized by vehicle traffic, commercial and residential zones. The ambient particle data was the benchmark for comparison with candle soot particles.

Figure 29. Countryside Mall located in Florida U.S.A.

Measurements included a non-candle restaurant to determine the output from secondary sources such as an open grill restaurant using gas broilers and flat grills. Closer proximity inside the dining room to the kitchen resulted in higher readings. Among restaurants, a back kitchen produced much lower secondary sources than did an open kitchen.

Table 19

Average levels of particle number concentrations at various urban locations

Sample Type	Location	Particle Number / cm^3
Ambient	Lifestyle Parking lot	17000
Ambient	Ruby Tuesday/Mall parking lot	16300
Restaurant	Ruby Tuesday	6420
Interior	Inside Mall	5000
Interior	First floor of mall	6500
Ambient	Mall parking lot	17500
Ambient	Grill Smith parking lot	18000
Restaurant	Grill Smith, by open kitchen	111000
Restaurant	Grill Smith, dining room	70000
Ambient	Leather Express, parking lot	18800
Interior	Leather Express, interior	6760
Ambient	U.S. 19 and Countryside	17700
Ambient	Bally Timbers parking lot	13900
Restaurant	Bally Timbers	33400
Restaurant	Bally Timbers dining room	13900
Ambient	By U.S. 19	12700
Ambient	SR 580 by Belcher	11400
Ambient	Spoto's parking lot	9800
Restaurant	Spoto's dining room	13200
Ambient	SR 580 and Edgewater Drive	36,500-70000
Restaurant	Jollemon's Grill	153000
Average		28614

Note. Partial Data

Measurement of ultrafine particles in Clearwater agreed with airborne concentrations cited in morbidity and mortality studies in the U.S. and Europe (Table 20).

Table 20

Average particle number concentrations of ambient environments

Study of Urban Location Ultrafine Particle Concentration	Location	Particles / cm^3
Osunsanya (2001)	Aberdeen, UK	10241
von Klot (2002)	Erfurt, Germany	17300
Bloch (2002)	Santa Monica, CA	56104
Bloch (2002)	Santa Monica, CA	41276
Wichmann (2000)	Erfurt, Germany	25773
Pekkanen (2002)	Kuopio, Finland	44300
Penttinen (2001)		14500
Average from studies, Particles/cm^3		29927.71

Table 21

Maximum particle number concentrations of ambient environments

Study of Urban Location Ultrafine Particle Concentration	Location	Particles / cm^3
Osunsanya (2001)	Aberdeen, UK	60636
von Klot (2002)	Erfurt, Germany	46195
Bloch (2002)	Santa Monica, CA	300000
Bloch (2002)	Santa Monica, CA	428000
Wichmann (2000)		26094
Ayers (1998)	Six City Study	50000
Tiittanen (1999)	Kuopio, Finland	40200
Reponen (2003)	Cincinnati, OH	32000
Levy (2002)	Boston, MA	140000
Molnar (2002)	Gothenburg, Sweden	105114
Hussein (2004)		46500
Pekkanen (2002)	Helsinki, Finland	50310
Dennekamp (2001a)	Aberdeen, UK	100000
Average of Maximum from studies, Particles/cm^3		109619.2

Calculation of Occupational LADD of Candle Soot

Occupational human factors were inputted into the LADD model (Table 22). The worklife expectancy is 39.9 years for men and 30.1 years for women (Bureau of Labor Statistics, 1997 Environmental Scan). For moderate activity, inhalation rate and implied daily inhalation volume for men are 2.5 m^3/hour and 20 m^3, and for women are 1.6 m^3/hr and 12.8 m^3 (EPA Exposure Factors Handbook (1996), Table 5-18, adapted from U.S. EPA 1985). Women account for 70.7 percent of waiters, the majority of spa workers, and about half of, collectively, clergy, nuns and priests (Bureau of Labor Statistics – 20 leading occupations of employed women, 2002 annual average).

Table 22

Occupational Human Factors

Workplace	Dominant Gender	Activity	Hours worked in 24 hr cycle	Working days per year worked
Restaurant	Women	Moderate	6.1	250
Spa	Women	Moderate	1-8	250
Church	None	Moderate	8	50

Exposure Concentration

The number of candles normally used in restaurants, churches and spas was established from the survey results and influences concentration levels (Table 23). Maximum candle soot exposure assumptions provided a better risk comparison to ambient levels.

Table 23

Typical Number of Candles in an Occupation

Occupation	Expected number of candles per room area
Restaurant	Maximum of 1 per 20 sq ft
Spa	1-5 per 100 sq ft, unventilated; 5-10 per 100 sq ft, ventilated
Church	Maximum of 10-20 per 100 sq ft, unventilated

Exposure Duration

The time a worker is exposed to candle emissions in a restaurant, church or spa was established from the survey results, and influences total ultrafine particle concentration during a typical day (Table 24).

Table 24

Typical Exposure Duration in an Occupation

Occupation	Number of hours exposed in a day/week
Restaurant	6.1 hours per day, 30.5 hours per week
Spa	1-8 hours per day, 8-40 hours per week
Church	One 8-hour Sunday per week, 8 hours per week

Table 25

Candle Exposure Lifetime Average Daily Dose of Inhaled Particles

Number and types of candles	[a]LADD from peak concentrations, inhaled particles (unventilated)	[a]LADD from steady state concentrations, inhaled particles (ventilated)
5 candles – unscented – restaurant	9.07×10^{10} +/- 1.51	1.86×10^{11} +/- 0.282
1 candle –scented –spa – 1 hour/day	5.73×10^{09} +/- 1.60	
1 candle –scented –spa – 8 hour/day	4.59×10^{10} +/- 1.28	
5 candles –scented –spa – 1 hour/day	1.70×10^{10} +/- 0.191	1.09×10^{10} +/- 0.126
5 candles –scented –spa – 8 hour/day	1.36×10^{11} +/- 0.153	8.73×10^{10} +/- 1.01
10 candles –scented – spa – 1 hour/day		1.25×10^{10} +/- 0.107
10 candles –scented – spa – 8 hour/day		9.98×10^{10} +/- 0.852
10 candles – church – church	5.62×10^{10} +/- 0.895	3.42×10^{10} +/- 0.367
20 candles – church – church		4.23×10^{10} +/- 0.309

Note. [a]LADD is based on 30-year work exposure duration.

Calculation of Ambient LADD of Ultrafine Particles

Calculated LADDs presented in Table 26 factor adult average daily inhalation rates for both men and women is 16 m^3/day (EPA, 1985, Table 5-20). The exposure duration used in the calculation is 24 hours, 365 days per year for a 70-year life expectancy.

Table 26

LADD of Inhaled Particles from Ambient Particle Concentration Averages

Ambient Average	[a]LADD from peak concentrations, inhaled particles
Literature Averages	4.79×10^{11} +/- 1.62
Literature Maximums	1.75×10^{12} +/- 1.12
Clearwater	3.25×10^{11} +/- 1.59

Note. [a]LADD is based on 70-year life expectancy

Comparison of Occupational and Ambient LADDs

The ambient environment has 3 to 40 times greater ultrafine particle number concentration than the high end of normal occupational candle use. For the comparison, 5 unscented candles in unventilated test room were selected for restaurants; 5 scented candles in unventilated test room, 8 hour duration per day were selected as a "worst case scenario" for spas; and 10 church candles in unventilated test room were a "worst case scenario" for churches. (See Figure 30.)
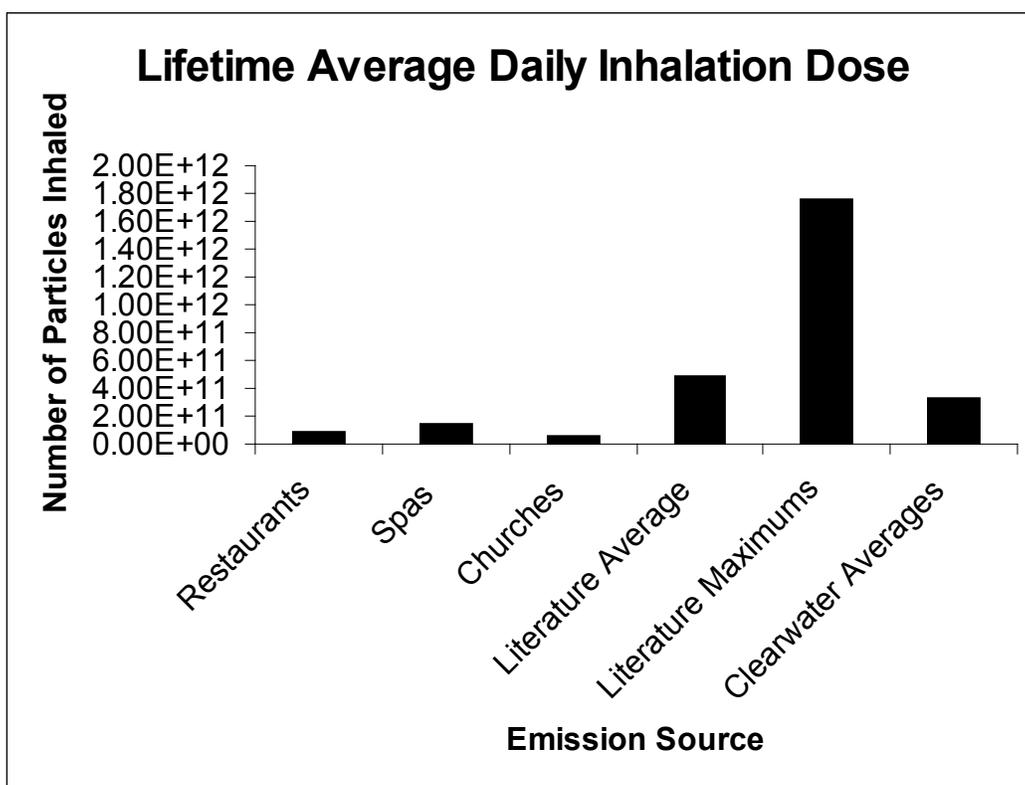

Figure 30. Lifetime Average Daily Dose of Inhaled Particles, Occupational Candle Soot and Ambient Ultrafine Particles

CHAPTER FOUR

DISCUSSION

The news media and the public seem to have long harbored misconceptions about the risk from exposure to combustion by-products. Often, an occupation is blamed for exposures that occur both in the workplace and in the ambient environment.

Apparatus Improvements

Candle chamber studies have found that ultrafine particles constitute the bulk of the candle emissions particle mass. However, these studies have used chambers that distort the particle size distribution relative to actual room conditions, and therefore have failed to capture particle dynamics such as particle agglomeration.

Our improved chamber simulated room conditions by lowering turbulence, equalizing pressure, and maintaining ambient room temperatures. We also generated and measured ultrafine particle concentrations from candle emissions in a real room with occupation-specific candles and numbers, in both ventilated and unventilated environments.

The present candle soot study employed state-of-the-art measurement technology and aerosol science, thus providing a more accurate perspective of risk than can be discerned from other studies. Multichannel optical particle counter measurement of the ambient urban environment demonstrated that the majority of particles are below 0.1 μm and 90% are in the 0.1-0.2 μm range. Particles sized 0.3 to 1 μm, contributed little to the distribution. OPC measurement of candle soot generation revealed that the most significant particle count increases were ultrafine particles below 0.1 μm. These results agree with previous candle soot particle size studies (Fine et al., 1999).

This work is reflective of current trends in occupational study that are focused on submicrometer particles. This "new science" is termed "nanotechnology" and has broad implications for technological developments in all aspects of industry and science. With this new trend, risk of cardiopulmonary disease from occupational exposures to "nano-sized" particles must be evaluated with animal and human studies.

Combustion Process

Real time analysis of candle soot UFP generation in a sealed, unventilated test room demonstrated that a peak concentration is reached soon after lighting, followed by a slower decline to slightly above background levels. These observations imply that coagulation effects take up nuclei particles rapidly. After the flame was extinguished, ultrafine particles increased in the room to a much greater concentration. These observations are consistent with other candle soot studies (Abt et al., 1999; Wallace, 2000). Exposure to ultrafine particles is brief during the short time the particle concentrations peak, approximately 30-60 minutes. Post peak concentrations diminish to near background concentrations within several hours. The exposure to soot particles and attached PAHs is minimized in an unventilated environment, therefore, the health risk is lower.

Ventilated test room data demonstrated a cyclic relation of repeat particle buildup, and then decay occurred over time. As with the sealed room, particle number concentration quickly rose to a peak and then decayed. When the air handler unit started, the decay transitioned to generation. Larger particles were removed by filtration and allowed for the generation of more nucleation mode particles without the immediate effects of coagulation. We found that there is more space between nucleation mode particles, the number of air changes causes steady state conditions, and the peak particle number concentration is roughly half the concentration in the sealed room. Steady state ultrafine particle concentration measured during a burn in a ventilated room is less than the peak concentration in an unventilated room. However, after the peak concentration in a ventilated room, the average ultrafine particle concentrations is less than the steady state average of a ventilated room. Hence, exposure is greater for workers in a ventilated environment compared to an unventilated environment. Therefore, candles burned in a ventilated environment pose more of a health risk than unventilated environments. However, compared to ambient ultrafine particle exposures, ventilated occupational environments pose less of an ultrafine particle health risk than the ambient environment.

Variables Affecting Emissions

Ventilated rooms maintain a higher concentration of ultrafine particles compared to sealed unventilated rooms. The higher ultrafine particle exposure implies increased health risk.

For both ventilated and unventilated test rooms, particle concentrations increased linearly with candle number. Scented candles generated more soot than unscented candles. Increased candle number and the use of scented candles increase the exposure and possible cardiopulmonary risks. Data collected from this study demonstrates that there are no significant differences in risk based on candle type. Based on survey data, candles reach a maximum

number in most occupations. Atypical numbers of candles may be found in some church areas, however, this is not common. The graphical findings imply that candle soot exposure can be predicted for an increasing number of candles and therefore the risk can be estimated.

Candle Emissions and Their Health Implications

Candle studies (Lau et al., 1997; Fine et al., 1999; Krause, 1999) have reported carcinogenic elements from their chemical analysis of candle emissions. Some of these studies plainly described the risks as minimal.   The studies cited just above were not able to demonstrate significant levels of PAHs. Malisch (1994) did not show that purple candles were a significant risk for dioxin exposures. Schwind et al. (1994) did not provide evidence that candles emit dangerous levels of organics or dioxins.

Candle chamber studies used modeling to extrapolate human doses from the measured chamber soot emissions. Their results were exaggerated because conditions inside the chambers did not simulate real room conditions. However, our improved chamber resulted in a lower generation rate, implying that PAHs, lead, and organics are lower and that the public and occupational health risks are lower.

Metals Emissions from Candles

The improved chamber detected no airborne lead. This was expected because candles with lead wicks were not burned inside the chamber. The chamber detected airborne zinc that translated to modeled concentrations that are below OSHA's permissible exposure level.

PAH Emissions from Candles

Krause's (1999) scented candles study assumes a similar toxicology for candle soot and diesel soot, and therefore comparable cancer risk. For example, he assumes that diesel soot's reference concentration of (RfC = 5 $\mu g/m^3$) applies to candle soot. Krause reported that candle emissions could cause significantly higher exposures to occupants (3-520 $\mu g/m^3$). The cancer risk for diesel exhaust was applied to exposures to candle soot, the estimated increased cancer risk for a lifetime exposure, would range from 9.7 x 10^{-5} to 3.0 x 10^{-4} for the lowest emitting candle to 1.5 x 10^{-2} to 4.7 x 10^{-2} for the highest emitting candle, using the range of unit cancer risk of 2.9 x 10^{-5} to 9.0 x 10^{-5} per $\mu g/m^3$.

However, the use of the diesel soot cancer slope factor as a surrogate for candle soot risk is invalidated by combustion studies demonstrating that soot is unique to the fuel source and to environmental conditions. These studies showed that carbonaceous soot from combustion has unique formation, PAHs, and generation rates that are dependent on temperature, fuel source and

environmental conditions. Moreover, PAH testing in our improved chamber and test room found the emission rate for PAHs to be < 1.14 ug/m^3 in the chamber and < 13 ug/m^3 in the test room. The literature review and measurements in the improved chamber and test room demonstrate that PAHs are not a risk factor for normal candle use. The undetected levels of PAH in the improved chamber and test room are consistent with other chamber candle emission studies, and provide strong evidence that PAHs emitted from candles are not a public or occupational health risk.

UFP Emissions from Candles

Few studies have targeted ultrafine particle counts rather than total particle mass, and have provided little evidence of associations. Oberdurster (1996), Wilson et al. (2002) and Donaldson et al. (2002) have provided strong evidence that inhalation of particles that are small and therefore have high surface area contribute to pulmonary and cardiovascular changes in test animals.

Mortality and morbidity studies have explored the association between ambient atmospheric ultrafine particle exposures and cardiopulmonary disease. Their findings are suspect, however, because of their having targeted asthmatics, misclassification due to central monitoring, and confounding due to extraneous pollutants and weak associations. Studies associating human exposure to ultrafine particles and health effects have been either weak or inconclusive. Peters et al. (1997) panel study of asthmatics and ultrafine particles provided evidence that pulmonary health effects from the 5 day mean UFP number were larger than the mass of fine particles.   Even though Wichmann et al. (2000) was able to show delayed association of UFP than of fine particles in a single day lag, no clear associations or patterns were observed for immediate or delayed effects. Von Klot et al. (2002), a panel study of 53 adult asthmatics with pulmonary symptoms, and mass concentration, ultrafine particles and ambient gases, did not produce solid evidence of a UFP association. Problems with recruitment, misclassification and results that did not correlate well, plagued this study.

Geographical locations cited in ultrafine particle morbidity studies have unique confounders and source apportionment. The uniqueness has lead to uncertainty in mirror studies because of the difference in pollution characteristics contributing to disease. Every city has a unique number and type of pollution sources. Morbidity and mortality rates can be quite different based on other factors contributing to the cardiopulmonary health effect under study. Urban environments are complex mixtures of atmospheric gases and particles, affecting persons with varied diets, smoking habits and cultural conditions. Study results are generally weak and provide inconclusive results. A specific referent ultrafine particle dose is not possible.

Occupational Lifetime Average Daily Dose from candle emission ultrafine particle concentrations based on the simulated occupational environment ranged from 3.91×10^{10} +/- 0.74 to 1.48×10^{10} +/- 0.20 inhaled particles, compared to the ambient environment concentrations of 1.18×10^{11} to 1.29×10^{12} inhaled particles.

Practical Implications of Findings

To provide a benchmark for the test room readings, measurements of ultrafine particle concentration were taken from representative urban locations in Clearwater, Florida. These averaged 20,281 particles/cm^3 and range from 6017 to 116,500 particles/cm^3, comparable to airborne concentrations cited in morbidity and mortality studies in the U.S. and Europe.

Ultrafine particle concentration data in the ventilated and unventilated test rooms provided accurate input for our dose calculation and risk model.

Candle soot dose was found to be one order of magnitude less than the ambient dose. Infrequent exposures from candle soot in comparison with constant inhalation of ambient ultrafine particles demonstrated that even with worst case soot volume densities, the risk is low.

Candle soot emissions do not appear to be an occupational hazard. Moderate use of candles seems to pose negligible risk in occupational and residential settings, because exposures to candle soot are too infrequent and transient to result in significant concentrations of hazardous chemical compounds.

Future Research Efforts

A more precise breakdown of ultrafine particle size would provide a better understanding of candle emissions, and could be accomplished using an electrostatic classifier. While beyond the budget for our study, this is suggested for future investigations.

This study is applicable to similar urban environments; however, other geographic locations have a wide variability of ultrafine particle concentrations and source apportionment. The ambient reference accuracy can be improved, and cofounders reduced, with more data collected from various locations throughout the United States.

With the invalid use of diesel soot as a surrogate for candle soot eliminated, the only thing left to focus on is the cardiopulmonary risks from the candle soot particle as another ultrafine particle.

Future research efforts should focus on improved soot characterization for PAH content and better-simulated environments. Monitoring workers for their specific exposure to ultrafine particles provides a more accurate quantification of dose. Improvements in predicting spatial distribution are helpful in understanding the nearness to the source and levels of exposure.

Occupation specific human factor data provide a better input for estimation of ultrafine particle dose. Restaurant workers tend to be younger, spa workers middle age and clergy in the upper age range. Using upper percentile respiration rates for restaurant workers because of their higher activity rate is recommended.

Occupation specific morbidity and mortality rates need to be developed for workers exposed to candles. An improvement over the simulated test environment is to conduct personal sampling on the candle exposed workers. Personal sampling for 24 hours would cover occupational and ambient exposure contributions. Controlling for tobacco, secondary sources, and the ambient atmosphere will reduce the potential for error.

REFERENCES

Abt, E., Suh, H. H., Allen G. A., & Koutrakis, P. (1999). Characterization of indoor particle sources: a study conducted in the metropolitan Boston area. *Environmental Health Perspectives, 108*, 35-44

Agency for Toxic Substances and Disease Registry (ATSDR) (1991). Toxicological Profile for Benzene (Draft). *Atlanta, GA: Agency for Toxic Substances and Disease Registry.*

Agency for Toxic Substances and Disease Registry (ATSDR) (1995). Public Health Statement for Polycyclic Aromatic Hydrocarbons (PAHs). *Government Printing Office.*

Agency for Toxic Substances and Disease Registry (ATSDR) (1998). *Toxicological Profile for Chlorinated Dibenzo-p-Dioxins*. Public Health Service, U.S. Department of Health and Human Services, Atlanta, GA.

Agency for Toxic Substances and Disease Registry (ATSDR) (1999). Tox Faqs for Chlorinated Dibenzo-*p*-dioxins(CDDs) . *Government Printing Office.*

Anderson, H. R., Limb, E. S., Bland, M. B., Ponce de Leon, A., Strachan, D. P., & Bower, J. S. (1995). Health effects of an air pollution episode in London. *Thorax, 50*, 1188-93.

Blake, C., & Silver, D. (2000). Candle emissions study using steel cylindrical chambers. *Unpublished data.* University of South Florida 2000

Bloch, J. Airport Vicinity Ultrafine Particle Measurements, Santa Monica-Malibu Unified School District, Abstract, April, 2002

Burton, R. M., Suh, H. H., & Koutrakis, P. (1996). Spatial variation in particulate concentrations within metropolitan Philadelphia. *Environmental Science & Technology, 30*, 400-407.

Business Wire. (1999). Candles Represent a $2 Billion Segment in the Giftware Market.

Chang, H. L., Kuo, M. L., & Lin, J. M. (1997). Mutagenic activity of incense smoke in comparison to formaldehyde and acetaldehyde in Salmonella typhimurium TA102. *Bulletin of Environmental Contamination and Toxicology, 58*, 394-401.

Crawford, M. (2003). Feasibility and Emissions of Compression Ignition Engines Fueled with Waste Vegetable Oil. *Thesis, University of South Florida*.

de Hartog, J. J., Hoek, G., Peters, A., Timonen, K. L., Ibald-Mulli, A., Brunekreef, B., et al. (2003). Effects of fine and ultrafine particles on cardio respiratory symptoms in elderly subjects with coronary heart disease: the ULTRA study. *American Journal of Epidemiology, 157*(7), 613-23.

Dennekamp, M., Howarth, S., Dick, C. A., Cherrie, J. W., Donaldson, K., & Seaton, A. (2001). Ultrafine particles and nitrogen oxides generated by gas and electric cooking. *Occupational & Environmental Medicine, 58*(8), 511-6.

Dennekamp, M., Mehenni, O. H., Cherrie, J., & Seaton, A. (2001a). Exposure to ultrafine particles and PM2.5 in different microenvironments. *University of Aberdeen, U.K. Inhaled Particles IX Conference.*

Dockery, D. W., Pope, C. A., III, Xu, X., Spengler, J. D., Ware, J. H., Fay, M. E., et al. (1993). An association between air pollution and mortality in six U.S. cities. *New England Journal of Medicine, 329*(24), 1753-1759.

Dockery, D. W., Schwartz, J., & Spengler, J. D. (1992). Air pollution and daily mortality: associations with particulates and acid aerosols. *Environmental Research, 59*, 362-373.

Donaldson, K., Brown, D., Clouter, A., Duffin, R., MacNee, W., Renwick, L., et al. (2002) The pulmonary toxicology of ultrafine particles. *Aerosol Medicine*. *15*(2), 213-20.

Donaldson, K., & Stone, V. (2003). Current hypotheses on the mechanisms of toxicity of ultrafine particles. *Ann 1st Super Sanikta, 39*(3), 405-410.

Donaldson, K., Stone, V., Gilmour, P. S., Brown, D. M., & MacNee, W. (2000). Ultrafine particles: mechanisms of lung injury. *Physical & Engineering Science, 358*, 2741-9.

Donaldson, K., Stone, V., Seaton, A., & MacNee, W. (2001). Ambient particle inhalation and the cardiovascular system: potential mechanisms. *Environmental Health Perspectives, 109*(Suppl. 4)*,*523-7.

Emission Inventory Improvement Program Particulate Emissions Committee (2001). Getting Started: Emissions Inventory Methods for PM-2.5.

European Candle Association (1997). Determining and Evaluating the Emissions of PCDD/PCDF, PAH and Short-Chain Aldehydes in Combustion Gases of Candles. *Oekometric GambH, Bayreuth, Germany*.

Ferin, J., Oberdorster, G., & Penney, D. P. (1992). Pulmonary retention of ultrafine and fine particles in rats. American Journal of Respiratory Cell Molecular Biology, *6*(5), 535-42.

Fine, P. M., Cass, G. R., & Simoneit, B. R. T. (1999). Characterization of fine particle emissions from burning church candles. *Environmental Science and Technology*, *33*, 2352-2362.

Fort Worth Star-Telegram. (1999). San Francisco Analyst Predicts Continued Growth for Candle Industry. *Knight-Ridder/Tribune Business News.*

Furtaw, E. J., Jr., Pandian, M. D., Nelson, D. R., & Behar, J. V. (1996). Modeling Indoor Air Concentrations Near Emission Sources in Imperfectly Mixed Rooms. Peer-reviewed paper in *Journal of. Air & Waste Management Association, 46*, 861-868.

Greene, J. F., Hays, S., & Paustenbach, D. (2003). Basis for a proposed reference dose (RfD) for dioxin of 1-10 pg/kg-day: a weight of evidence evaluation of the human and animal studies. *Journal of Toxicology & Environmental Health B Critical Review, 6*(2),115-59.

Guo, Z., & Miller, A. (1999). Characterization of Indoor Fine Particulate Matter. *EPA/NRMRL Grant.*

Guo, Z., Mosley, R., McBrian, J., & Fortmann, R. (2000). Fine Particulate Matter Emissions from Candles. *R Govt Reports Announcements & Index (GRA&I)*, Issue 24.

Hamins, Anthony (1993). Soot, Environmental Implications of Combustion Processes, *CRC Press, Florida*, pp. 71–95.

Hebgen, P., Goel, A., Rainey, L. C., Vander Sande, J. B. & Howard, J. B. (2001). Combustion Synthesis of Fullerenes and Fullerenic Nanostructures. *Department of Chemical Engineering Department of Materials Science and Engineering, Massachusetts Institute of Technology.*

Hinds, W. (1999). Aerosol Technology, Properties, Behavior, and Measurement of Airborne Particles. *Second Edition, John Wiley & Sons, Inc.*

Hussein, T., Hameri, K., Aalto, P., Asmi, A., Kakko, L., & Kulmala, M. (2004). Particle size characterization and the indoor-to-outdoor relationship of atmospheric aerosols in Helsinki. *Scandinavian Journal of Work, Environment & Health*, *30*(Suppl. 2), 54-62.

Iman, R. L., Helton, J. C., & Campbell, J. E. (1981) An Approach to Sensitivity Analysis of Computer Models: Part I --- Introduction, Input Variable Selection and Preliminary Assessment. *Journal of Quality Technology*, *13*(3), 174-183.

International Commission on Radiological Protection (1994) Human Respiratory Tract Model for Radiological Protection. Annals of the ICRP, Publication 66, Elsevier Science, Inc.

Jeong, C. H., Hopke, P. K., Chalupa, D., & Utell, M. (2004). Characteristics of nucleation and growth events of ultrafine particles measured in Rochester, NY. *Environmental Science & Technology*, *38*(7), 1933-40.

Johnston, C. J., Finkelstein, J. N., Mercer, P., Corson, N., Gelein, R., & Oberdorster, G. (2000). Pulmonary effects induced by ultrafine PTFE particles. *Toxicology & Applied Pharmacology*, *168*(3), 208-15.

Junker, M. H., Koller, T., & Monn, C. (2000). An assessment of indoor air contaminants in buildings with recreational activity. The Science of the Total Environment, *246*, 139-152.

Kidd, P. M. (1997). Glutathione: systemic protectant against oxidative and free radical damage. Altern Med Rev, *1*, 155-176.

Kittelson, D. B., Watts, W. F., Johnson, J. P., Remerowki, M. L., Ische, E. E., Oberdorster, G., et al. (2004). On-road exposure to highway aerosols. Aerosol and gas measurements. *Inhalation Toxicology*, *16*(Suppl. 1), 31-9.

Krause, D. (1999). Black soot and candles: new research and case studies. Indoor Environment '99 Proceedings. pp 157-164. *IAQ Publications, Inc.,* Bethesda, MD.

Lau, C., Fiedler, H., Hutzinger, O., Schwind, K. H., & Hosseinpour, J. (1997). Levels of selected organic compounds in materials for candle production and human exposure to candle emissions. *Chemosphere*, *34*(5-7),1623-1630.

Levy, J. I., Dumyahn, T., & Spengler, J. D. (2002). Particulate matter and polycyclic aromatic hydrocarbon concentrations in indoor and outdoor microenvironments in Boston, Massachusetts. *Journal of Exposure Analysis and Environmental Epidemiology, 12*(2), 104-14.

Levy, J. I., Houseman, E. A., Ryan, L., Richardson, D., & Spengler, J. D. (2000) Particle concentrations in urban microenvironments. *Environmental Health Perspective, 108*(11), 1051-7.

Li, X. Y., Gilmour, P. S., Donaldson, K., & MacNee, W. (1996). Free radical activity and pro-inflammatory effects of particulate air pollution (PM10) in vivo and in vitro. *Thorax, 51*, 1216-22.

Li, W., & Hopke, P. K. (1993). Initial size distributions and hygroscopicity of indoor combustion aerosol particles. *Aerosol Science and Technology*, *19*(3), 305-316.

Lippmann, M., Xiong, J. Q., & Li, W. (2000). Development of a continuous monitoring system for PM10 and components of PM2.5. *Applied Occupational & Environmental Hygiene, 15*(1), 57-67.

Löfroth, G., Stensman, C., & Brandhorst-Satzkorn, M. (1991). Indoor sources of mutagenic aerosol particulate matter: smoking, cooking, and incense burning. *Mutation Research 261*, 21-28.

MacNee, W., & Donaldson, K. (2003). Mechanism of lung injury caused by PM10 and ultrafine particles with special reference to COPD. *European Respiratory Journal Supplement, 40*, 47s-51s.
Malisch, R. (1994). Determination of dioxins and furans in colored candle wax. *Chemosphere*, *29*(9-11), 1957-1963.

Marvin, C. H., Leppard, G. G., West, M. M., Stern, G. A., Boden, A. R., McCarry, B. E. (2004). Refined Tunable Methodology for Characterization of Contaminant–Particle Relationships. *Journal of Environmental Quality, 33*, 2132-2140.

McBrian, J., Fortmann, R., Guo, Z., & Mosley, R. B. (2000). Test Methods to Characterize Particulate Matter Emissions and Deposition Rates in a Research House. *Govt Reports Announcements & Index (GRA&I),* Issue 24.

Molnar, A., Meszaros, E., Hansson, H. C., Karlsson, H., Gelencser, A., Kiss, G. Y., et al. (1999). The importance of organic and elemental carbon in the fine atmospheric aerosol particles, Atmospheric. Environment, *33*, 2745–2750.

Molnar, P., Janhall, S., & Hallquist, M. (2002). "Roadside measurements of fine and ultrafine particles at a major road north of Gothenburg." *Atmospheric Environment, 36*(25), 4115-4123.

Morawska, L., Bofinger, N., Kosic, L., & Nwankowala, A. (1998). "Submicrometer and supermicrometer particles from diesel vehicle emissions." *Environmental Science & Technology, 32*(14), 2033-2042

Morawska, L., Moore, M. R. & Ristovski, Z. D. (2003). "Desktop literature review and analysis of health impacts of ultrafine particles", *Report to Environment Australia*

Nazaroff, W. W. (2004) Indoor particle dynamics. *Indoor Air*, 14(Suppl. 7), 175-83. Review.

Nicas, M. (2000). Markov modeling of contaminant concentrations in indoor air. *American Industrial Hygiene Journal, 61*, 484-91.

Nriagu, J. O., & Kim, M. J. (2000). Emissions of lead and zinc from candles with metal-core wicks. *Science of the Total Environment, 250*, 37-41.

Oberdorster, G. (1996). Significance of particle parameters in the evaluation of exposure-dose-response relationships of inhaled particles. *Inhalation Toxicology*. *8*(Suppl.), 73-90.

Oberdorster, G., Finkelstein, J. N., Johnston, C., Gelein, R., Cox, C., Baggs, R., et al. (2000). Acute pulmonary effects of ultrafine particles in rats and mice. *Research Report/ Health Effects Institute, 96*, 5-74; disc. 75-86.

Oberdorster, G., & Utell, M.J., (2002). Ultrafine Particles in the Urban Air: To the respiratory Tract - And Beyond? *Environmental Health Perspectives, 110*(8), A440-441.

Osunsanya, T., Prescott, G., & Seaton, A. (2001). Acute respiratory effects of particles: mass or number? *Occupational & Environmental Medicine*, *58*(3), 154-9.

Özkaynak, H., Xue, J., Weker, R. A., Butler, D. A., Koutrakis, P., & Spengler, J. D. (1996). The Particle TEAM (PTEAM) Study: Analysis of the Data; Volume III: Final Report (EPA/600/R)

Pekkanen, J., Peters, A., Hoek, G., Tiittanen, P., Brunekreef, B., de Hartog, J., et al. (2002). Particulate air pollution and risk of ST-segment depression during repeated submaximal exercise tests among subjects with coronary heart disease. The exposure and risk assessment for fine and ultrafine particles in ambient air *(ULTRA) study. Circulation, 20*, 933-8.

Penttinen, P., Timonen, K. L., Tiittanen, P., Mirme, A., Ruuskanen, J., & Pekkanen, J. (2001). Number concentration and size of particles in urban air: effects on spirometric lung function in adult asthmatic subjects. *Environmental Health Perspective, 109*(4), 319-23.

Penttinen, P., Timonen, K. L., Tiittanen, P., Mirme, A., Ruuskanen, J., & Pekkanen, J. (2001a). Ultrafine particles in urban air and respiratory health among adult asthmatics. *European Respiratory Journal, 17*(3), 428-35.

Peters, A., Liu, E., Verrier, R. L., Schwartz, J., Gold, D. R., Mittleman, M., et al. (2000). Air pollution and incidence of cardiac arrhythmia. *Epidemiology, 11*, 11-17.

Peters, A., Wichmann, H. E., Tuch, T., Heinrich, J., & Heyder, J. (1997). Respiratory effects are associated with the number of ultrafine particles. *American Journal of Respiratory Critical Care Medicine, 155*(4), 1376-83.

Pope, C. A., III (2000) Epidemiology of fine particulate air pollution and human health: biologic mechanisms and who's at risk? *Environmental Health Perspective, 108*(Suppl. 4), 713-723.

Pope, C. A., III, Dockery, D. W., Spengler, J. D., & Raizenne, M. E. (1991). Respiratory health and PM_{10} pollution: a daily time series analysis. *American Review of Respiratory Disease, 144*, 668-674.

Pope, C. A., III, Verrier, R. L., Lovett, E. G., Larson, A. C., Raizenne, M. E., Kanner, R. E., et al. (2000). Heart rate variability associated with particulate air pollution. *American Heart Journal, 138*, 890-899.

Public Citizen's Health Research Group, Washington, DC. (2000). Commission to Immediately Ban and Recall All Candles With Lead-Containing Wicks, Candles in Metal Containers That Contain Lead, and Wicks Sold For Candle-Making That Contain Lead.

Reponen, T., Grinshpun, S. A., Trakumas, S., Martuzevicius, D., Wang, Z. M., LeMasters, G., et al. (2003). Concentration gradient patterns of aerosol particles near interstate highways in the Greater Cincinnati airshed. *Journal of Environmental Monitoring, 5*(4), 557-62.

Riley, W. J., McKone, T. E., Lai, A. C., & Nazaroff, W. W. (2002). Indoor particulate matter of outdoor origin: importance of size-dependent removal mechanisms. Environmental Science & Technology. 36(2):200-7. *Erratum in: Environmental Science & Technology, 36*(8), 1868.

Ristovski, Z., Morawska, L., Bofinger, N., & Hitchins, J. (1998). "Submicrometre and Supermicrometre Particulate Emission from Spark Ignition Vehicles." *Environmental Science & Technology, 32*, 3845-3852.

Ristovski, Z., Morawska, L., Thomas, S., Hitchins, J., Greenaway, C., & Gilbert, D. (2000). "Particle emissions from natural gas engines." *Journal of Aerosol Science, 31*, 403-413

Roveri, P., Andrisano, V., Di Pietra, A. M., & Cavrini, V. (1998). GC-MS analysis of incenses for possible presence of allergenic nitromusks. *Journal of Pharmaceutical and Biomedical Analysis*, *17*, 393-398.

Schwind, K. H., & Hosseinpour, J. (1994). Analysis of Raw Materials and Candle Combustion Products. *Oekometric GambH, Bayreuth, Germany, for the Association of German Candle Manufacturers, Inc.,* Frankfurt, Germany.

Seaton, A., MacNee, W., Donaldson, K., & Godden, D. (1995). Particulate air pollution and acute health effects. *Lancet,* 345, 176-8.

Sexton, K., & Ryan, P. B. (1988). Assessment of Human Exposure to Air Pollution: Methods, Measurements and Models. In A. Y. Watson, R. R. Bates, & D. Kennedy (Eds.), *Air Pollution, the Automobile and Public Health* (pp. 207-238). Washington, DC: National Academy Press.

Shaddix, C. R., & Smyth, K. C. (1994). *Eastern States Meeting of the Combustion Institute, Clearwater, FL*, p. 329.

Siegmann, H. C. (1990) Combustion Aerosols. *VIERTELJAHRSSCHR NATURFORSCH GES ZUER. 135*(4), 197-219

Sobel, H. L., Lurie, P., & Wolfe, S. M. (2000). Letter: lead exposure from candles. *Journal of the American Medical Association, 284*(2), 180.

Spengler, J. D. (2003). Estimation of primary and secondary particulate matter intake fractions for power plants in Georgia. Department of Environmental Health, Harvard School of Public Health, *Environmental Science & Technology, 37*(24), 5528-36.

Spengler, J. D., & Thurston, G. D. (1983). Mass and elemental composition of fine and coarse particles in six U.S. cities. *Journal Air Pollution Control, 33*(12), 1162-1171.

Spengler, J. D., Treitman, R. D., Tosteson, T. D., Mage, D. T., & Soczek, M. L. (1985). Personal exposures to Respirable particulates and implications for air pollution Epidemiology. *Environmental Science Technology, 19*, 700-707.

Stone, V., Shaw, J., Brown, D. M., MacNee, W., Faux, S. P., & Donaldson, K. (1998). The role of oxidative stress in the prolonged inhibitory effect of ultrafine carbon black on epithelial cell function. *Toxicology In Vitro, 12*, 649-59

Symposium on Particulate Matter (2003). *MIT Laboratory for Energy and Management Endicott House, Bedham, Mass*

Timonen, K. L., Hoek, G., Heinrich, J., Bernard, A., Brunekreef, B., de Hartog, J., et al. (2004). Daily variation in fine and ultrafine particulate air pollution and urinary concentrations of lung Clara cell protein CC16. *Occupational and Environmental Medicine, 61*(11), 908-14.

Tiittanen, P., Timonen, K. L., Ruuskanen, J., Mirme, A., & Pekkanen, J. (1999). Fine particulate air pollution, re-suspended road dust and respiratory health among symptomatic children. *European Respiratory Journal, 13*(2), 266-73.

Tung, T. C. W., Chao, C. Y. H., Burnett, J., Pang, S. W., & Lee, R. Y. M. (1999). A territory wide survey on indoor particulate level in Hong Kong. *Building and Environment, 34*(2), 213-220.

Unity Marketing. (2000). Candle Report: The Market, The Industry, The Trends. *Unity Marketing, Stevens, PA.*

U.S. Environmental Protection Agency (1985). *Health Assessment Document for Polychlorinated Dibenzo-p-Dioxin.* Environmental Criteria and Assessment Office, Office of Health and Environmental Assessment, Office of Research and Development, Cincinnati, OH. EPA 600/8-84-014F.

U.S. Environmental Protection Agency (1997). 40 CFR, Part 50: National Ambient Air Quality Standards for Particulate Matter, *Final Rule (Regulation). Federal Register* 62:38651-38701.

U.S. Environmental Protection Agency (1997a). *Health Effects Assessment Summary Tables. FY 1997 Update.* Environmental Criteria and Assessment Office, Office of Health and Environmental Assessment, Office of Research and Development, Cincinnati, OH.

U.S. Environmental Protection Agency (1998, 1999, and 2001). The National Research Council's three reports (Research Priorities for Airborne Particulate Matter; 1998, 1999, and 2001)

U.S. Environmental Protection Agency (2000). Integrated Risk Information System (IRIS). Substance file- benzene. Washington, D.C.: National Center for Environmental Assessment.

U.S. Environmental Protection Agency (2003). Dioxin Reassessment, NAS Review Draft, Dioxin Information Sheet 1 Dioxin: Summary of the Dioxin Reassessment Science

U.S. Environmental Protection Agency (2004) Dioxin Reassessment, NAS Review Draft, Dioxin Information Sheet 4 Dioxin: Continuing EPA Efforts to Reduce the Public's Exposure to Dioxin Risks

Utell, M. J., & Frampton, M. W. (2000). Acute health effects of ambient air pollution: the ultrafine particle hypothesis. *Aerosol Medicine, 13*(4), 355-59.

van Alphen, M. (1999). Emission testing and inhalation exposure-based risk assessment for candles having Pb metal wick cores. *The Science of the Total Environment, 243/244*, 53-55.

Vigil, F. (1998). Black stains in houses: soot, dust, or ghosts? *Home Energy*, *15*(1), 15.

von Klot, S., Wolke, G., Tuch, T., Heinrich, J., Dockery, D. W., Schwartz, J., et al. (2002). Increased asthma medication use in association with ambient fine and ultrafine particles. *European Respiratory Journal, 20*(3), 691-702.

Wallace, L. A. (1987). The TEAM Study: Summary and Analysis. Vol I. U.S. EPA, EPA 600/6-87/002a, NTIS PB 88-100060. U.S. Washington, DC: Environmental Protection Agency.

Wallace, L. (2000). Real-Time Monitoring of Particles, PAH, and CO in an Occupied Townhouse. *Applied Occupational and Environmental Hygiene, 15*(1), 39-47.

Wasson, S. J., Guo, Z., McBrian, J. A., & Beach, L. O. (2002). Lead in candle emissions. *Science & Total Environment, 296*(1-3), 159-74.

Watson, A. Y., & Valberg, P. A. (2001). Carbon black and soot: two different substances. *American Industrial Hygiene Journal, 62*(2), 218-28.

World Health Organization (1989). Environmental Health Criteria for Formaldehyde. Vol 89. *Geneva: World Health Organization.*

World Health Organization (1999) World Health Organization Dioxin and their Effects on Human Health. *Fact Sheet 225.*

World Health Organization (1999a). World Health Organization Health Guidelines for Vegetation Fire Events. *World Health Organization, Geneva, Switzerland.*

Wichmann, H. E., Spix, C., Tuch, T., Woelke, G., Peters, A., Heinrich, J., et al. (2000). Daily mortality and fine and ultrafine particles in Erfurt, Germany. Part I: role of particle number and particle mass. Res Rep Health Eff Inst., (98), 5-86; discussion 87-94

Wilson, M., Lightbody, J. H., Donaldson, K., Sales, J. H., & Stone, V. (2002). Interactions between ultrafine particles and transition metals in vivo and in vitro. *Toxicology and Applied Pharmacology, 184*(3), 172-179.

ABOUT THE AUTHOR

David Jonathan Silver holds a Bachelor of Arts in Chemistry and Master of Science Degree in industrial hygiene from Wayne State University in Detroit, Michigan. In 1987, he founded Silver Star Environmental Consultants, an industrial hygiene consulting firm and in 1993, CIH Equipment Company, an environmental test equipment rental, and repair and calibration company. Experience as an analytical chemist, corporate industrial hygienist and consultant have prepared him well to take on consulting assignments with local businesses and government facilities throughout the Southeast United States, Southeast Asia the Caribbean and Europe. He is active with the Florida Industrial Hygiene Association and holds the office of Communications Director.