Ultrafine particle emissions: Comparison of waste-to-energy with coal- and biomass-fired power plants

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EXECUTIVE SUMMARY

Combustion processes are the dominant anthropogenic sources of particulate matter emissions to the atmosphere. Recently, special attention has been directed to the potential health effects of ultrafine particles (UFP) of diameters less than 0.1μm. Numerous research studies show that inhalation exposure to UFP can lead to exacerbation of lung and cardiovascular diseases and that the effect is more severe than that of fine and coarse particles. It has also been shown that UFP can generate more reactive oxygen species (ROS) than larger particles and are able to cross epithelial cells and translocate to extrapulmonary organs.

In the combustion of coal, biomass, and municipal solid wastes, UFP mainly form by nucleation of metal vapor followed by growth through coagulation and condensation of other vaporized materials. Some of these particles are not captured in the air pollution control (APC) and are emitted to the atmosphere with the treated flue gas. UFP emissions from stationary combustion sources, in general, and from waste-to-energy (WTE) plants, in particular, are not well characterized as yet.

This study included a critical analysis of the literature with the objective to quantify the number concentration of UFP emitted from WTE facilities in comparison to coal and biomass power plants. Analysis of fly-ash samples from coal burned in laboratory combustors has shown that the ultrafine fraction contained higher concentrations of volatile metals, in some cases 50 times higher, than in fine and coarse particles. This supports the idea that volatile and semi-volatile metals in the fuel vaporize and nucleate to form UFP.

Research studies show that the number concentration of UFP in WTE stack gas is on the order of 10^5-10^6 particles/cm^3. However, this number varied significantly between different facilities and in some cases it was considerably lower. The number concentration of UFP in coal-fired and biomass-fired power plants stack gas was found to be on the order of 10^6-10^7 particles/cm^3. It is possible that the activated carbon particles used by the WTE industry to lower mercury and other heavy metals emissions, and also dioxins and furans, also result in reducing UFP emissions.

Further research is required in order to better characterize UFP emission from these combustion sources in terms of number concentration and also chemical composition. An experimental approach is discussed at the end of this paper.

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1. Introduction

Particulate matter (PM) refers to solid or liquid particles suspended in the air. The atmosphere contains considerable concentrations of PM, particles that range from a few nanometers up to 100 microns in diameter. The sources of PM are both anthropogenic and natural. PM can be emitted either directly from industrial activities, transportation, power plants and natural sources, or indirectly through conversion of gaseous precursors such as ammonia and sulfur oxides. PM is categorized according to its aerodynamic diameter: PM_{10}, also called "coarse PM", refers to articles with diameter of 10 μm or less, $PM_{2.5}$, also called "fine PM", refers to particles with diameter of 2.5 μm or less, and $PM_{0.1}$, also called "ultrafine particles" (UFP), have diameters of 0.1 μm or less.

The size of particles generated by combustion ranges from a few nanometers up to 1 μm, whereas the size of naturally occurring PM, such as pollens, plant fragments, and sea salt is generally larger than 1 μm. Particles generated by photochemical processes in the atmosphere are generally smaller than 1 μm (Seinfeld and Pandis). The number concentration of atmospheric PM varies spatially and temporally, with typical number concentrations being on the order of thousands of particles per cm^3 (Costabile et al, 2009).

In recent years, increasing attention has been given to ultrafine particles (UFP) due to the potential adverse health effects associated with them. UFP can stay suspended in air for several days and be transported over thousands of kilometers (Hinds, 1999). Comparing with larger particles, UFP have a much larger surface area to volume ratio and can carry large amounts of adsorbed pollutants (Oberdorster, 2001). UFP are ubiquitous in the atmosphere and dominate the total number of particles in urban PM (Lightly et al, 2000; Seinfeld and Pandis). However, their contribution to the total mass of PM in air is very small. For example, the mass of a 10 μm particle is equivalent to the mass of one billion 10-nm particles.

UFP are formed by combustion processes and also through gas-to-particle conversion. In the urban environment, a major fraction of UFP is related to road traffic and emissions from stationary combustion sources (Young and Keeler, 2004; Wehner et al, 2008). Heavy duty diesel vehicles are in fact one of the greatest contributors to urban UFP (EPA), emitting up to two orders of magnitude more UFP, as compared to gasoline trucks (Whener et al, 2009), with a typical UFP emission rate of 10^{15} particles per kilometer of travel (Morawska et al, 2008). Legislation regarding emissions and ambient concentrations of PM is based on particle mass concentration, to which UFP are a minor contribution.

Waste combustion in Waste-to-Energy facilities (WTE) is a method for reducing the waste volume by about 90% and at the same time recovering heat and using it to generate electricity. Concerns regarding emission of toxic combustion by-products such as dioxins and mercury, have been resolved within the past decade following successful measures, such as implementation of the Maximum Achievable Control Technology (MACT) of the U.S. EPA. However, as in all other combustion processes, waste incineration produces UFP. This study is a critical analysis of the literature on number concentration of UFP generated by WTE plants relative to coal and biomass power plants and, also, of the health effects associated with UFP exposure. Finally, we offer recommendations for decreasing UFP emissions to the atmosphere.

2. Health Effects of PM and UFP Emissions

Numerous epidemiological studies have found a positive correlation between the level of particulate air pollution and increased morbidity and mortality rates in both adults and children, especially for people with pre-existing pulmonary and cardiovascular diseases (de Hartog et al, 2003; Disney, 2004; Janssen et al, 2005; Pope et al, 2009; Halonen et al, 2009). The main exposure route to particulate matter is through the respiratory system. Inhalable coarse particles- those with diameter in the range of 2.5 to 10 μm- mainly deposit in the upper respiratory system and may be cleared via mucociliary actions (Cormier et al, 2006). $PM_{2.5}$ is able to penetrate the alveolar regions of the lungs, where the UFP fraction may cross the epithelium (Oberdorster, 2001). The toxicity and potential adverse health effects of airborne particulate matter are strongly affected by the size of the particle and its chemical composition.

Pulmonary effects

Clinical, epidemiological and experimental research studies indicate that airborne particles, particularly fine particles, may contribute to adverse respiratory effects (Ghio et al, 2000; Holgate et al, 2003). Several studies focused on the specific role of UFP in these effects. In a laboratory experiment, mice that were exposed to ultrafine (20 nm) TiO_2 particles expressed much greater pulmonary - inflammatory response than mice exposed to fine (250 nm) TiO_2 particles, presumably due to the high surface area to volume ratio of UFP (Oberdorster, 2001). Rats exposed to burning polytetrafluoroethylene (PTFE) fumes containing UFP exhibited severe pulmonary-inflammatory response. However, no toxic effects were observed after letting the fresh PTFE fumes stand for 3.5 minutes (Johnston et al, 2000). The authors attributed the diminished toxicity to the bigger particle size resulting by coagulation. Penttinen et al (2001) studied the relationship between PM exposure and exacerbation of symptoms in asthmatic adults. They found that elevated number concentrations of UFP had t stronger effects than fine and coarse particles. It has also been proposed that inhalation of carbon black UFP may induce allergic lung inflammation in susceptible individuals (de Haar et al, 2005; Allesandrini et al, 2009).

Cardiovascular effects

Inhaled particles may cause adverse health effects beyond the lungs. Epidemiological studies show associations between airborne PM and cardiovascular morbidity and mortality (Disney, 2004; Janssen et al, 2005; Timonen et al, 2006). Adverse cardiac effects, related specifically to UFP exposure, have been observed in adults with pre-existing cardiac conditions (Timonen et al, 2006). Exposure to concentrated ambient UFP was associated with increase in heart rate variability in healthy young adults (Samet et al, 2009). Experiments in rabbits and mice showed that PM exposure promotes atherosclerosis and that the effects are more severe for UFP (Araujo and Nel, 2009). The mechanisms by which inhaled UFP affect the cardiovascular system are not fully understood yet. However, according the most accepted model, their impact is either indirect through pulmonary inflammation, or direct by translocation to the bloodstream (Mills et al, 2006).

Translocation to extrapulmonary organs

Translocation of insoluble UFP into the bloodstream has been the subject of numerous research studies. In a laboratory experiment, gold UFP inhaled by mice exhibited translocation to extrapulmonary organs among which are the liver, kidney and heart (Furuyama et al, 2009). Experiments in rats showed translocation of UFP to the central nervous system along the olfactory nerve (Oberdorster et al, 2004; Elder et al, 2006). Geys et al (2005) observed translocation of UFP through human lung cells in vitro. Long-term exposure to UFP may result in accumulation of UFP in extrapulmonary organs, possibly leading to onset of diseases following decades of exposure (Peters et al, 2006).

Oxidative stress

Studies have demonstrated that UFP can generate more reactive oxygen species (ROS) than larger particles (Kang et al, 2008; Xia et al, 2006; Yang et al, 2008). ROS are reactive molecules that can react and interfere with the normal function of cellular macromolecules such as lipids, proteins, and DNA (Stone et al, 2007). The condition caused by elevated ROS levels is called "oxidative stress". Brauner et al (2007) observed a systemic oxidative stress leading to DNA damage in human subjects following exposure to UFP.

Health effects related to the chemical composition of combustion particles

The chemical properties of the particle's surface also play an important role in the biological effects it may cause. Combustion generated particles are of specific concern since they have been found to be associated with toxic compounds such as polycyclic aromatic hydrocarbons (PAH) and metals (Cromier et al, 2006). PAH promote oxidative stress and inflammation (Araujo and Nel, 2009), and many of the PAH compounds are also carcinogenic and mutagenic. PAH formed by combustion are mainly concentrated on the fine and ultrafine fraction of PM (Zielinska et al, 2004; Riddle et al, 2007; Lin et al, 2008; Wichmann et al, 2009). PM-associated metals, especially the water soluble ones, are stated to cause pulmonary and cardiovascular injuries (Wallenborne et al, 2008; Costa and Dreher, 1997). They have also been observed to cause DNA damage to human epithelial alveolar cells in vitro (Di Pietro et al, 2009). Transition metals are highly oxidative and reactive in biological systems and are able to generate more oxidative stress (Wilson et al, 2002; Balakrishna et al, 2009). Various metals such as Cu, Ni and Fe can become redox active and cause oxidation of biomolecules leading to severe oxidative stress within cells or tissues (Park et al, 2007). Studies have shown that transition metals can amplify the ability of ultrafine particles to induce lung inflammation (Wilson et al, 2007).

Long term exposure effects

Toxicology studies of chronic or subchronic exposure to combustion generated UFP are not common. However, Kodavanti et al (2003) reported that rats developed cardiac lesions following 16 consecutive weeks exposure to concentrations of combustion UFP, while rats exposed to the same particles for either four consecutive weeks or four consecutive days did not develop this condition. All three groups, however, developed pulmonary inflammation. In another laboratory experiment, exposure of rats to low concentrations of UFP rich in zinc for 16 consecutive weeks caused small changes in cardiac gene expression without causing substantial lung injury (Wallenborne et al, 2008).

Airborne UFP have tendency to form larger particles via coagulation or from condensation of vapors onto existing particles. This tendency, which may limit long term exposure, has also been observed for combustion generated UFP (Linak and Wendt, 1993; Chang et al, 2004). However, studies show that the compounds associated with combustion UFP may stimulate inflammatory responses, both in vivo and in vitro, even when bound to larger particles (Wilson et al, 2002). Thus, UFP emissions from combustion sources remain a matter of concern when physical processes, taking place post emission, are considered.

3. UFP in Coal, Biomass, and Waste Combustion Systems

In coal, biomass and waste combustion systems, ultrafine particles are assumed to form mainly by homogeneous nucleation of metal vapors (in either chloride or oxide form) followed by growth through coagulation and condensation of other vaporized materials (Chang et al, 2000; Carbone et al, 2008; Linak et al, 2007). Nucleation occurs in a supersaturated vapor phase. The saturation ratio of a solute A in air at a given temperature, is defined as:

$$S = \frac{P_A}{P_A^*}$$

where P_A is the partial pressure of A and P_A* the saturation vapor pressure. S= 1 for saturated vapor, S<1 for sub-saturated vapor, and S>1 for supersaturated vapor. At saturation, there is no nucleation of a stable phase of A from the gas phase; for nucleation to occur S has to be greater than 1. The aerodynamic radius of the smallest stable homogeneous nucleated particle is defined as the "critical size". Below the critical size particles will spontaneously decompose and return to the vapor phase. According to the classical nucleation theory (Seinfeld and Pandis), the free energy of formation of the critical size cluster is expressed by:

$$\Delta G^* = \frac{16\pi v_1^2\sigma^3}{3(kTlnS)^2}$$

where v_1 is the volume of the condensable molecule in the condensed phase, σ the surface tension, k the Boltzmann constant and T the temperature. The critical size, r*, is derived from Kelvin's equation:

$$r^* = \frac{2\sigma v_1}{kTlnS}$$

Increasing the saturation ratio, S, decreases both the free energy barrier and the critical radius. Since σ, the surface tension of the metal in the liquid state (Table 1) is very high for metals and considering the cubic term in the free energy equation, a very high saturation ratio is required in order to overcome the free energy barrier. For these extreme supersaturation ratios, r* has to be very small. Supersaturation conditions are reached when the vapor is cooled or/and when the partial pressure of the metal vapor increases beyond saturation. In general, homogeneous nucleation of metal vapor is enhanced by high cooling rates and low system pressure (Singh et al, 2002).

Table 1: Surface tension values of selected heavy metals in liquid state (Gale and Totemeier, 2004)

Metal	*Temperature (K)*	*Surface Tension (milliNewton/meter)*
Ag	1233.7	966
Al	933	914
Cd	594	570
Cr	2148	1700
Cu	1356	1303
Fe	1809	1872
Hg	234.13	498
Mn	1514	1090
Ni	1727	1778
Pb	600	458
Zn	692	782

The classical nucleation theory has been criticized as being inadequate to predict the critical particle size because it assigns surface tension bulk properties to clusters of particles (Lightly et al, 2000; Martens et al, 1987). However, more recent models and experimental work have shown that homogeneous nucleation of metal vapor does result in stable particles with diameter of less than 10 nm (Bahadur and McClurg, 2004; Lummen and Kraska, 2005; Carbone et al, 2008). Finney and Fink (2008) hypothesized that for transition metals, the critical size of the nucleus depends on the strength of the metal-metal bonds. Since the heat of vaporization of a metal, ΔH_{vap} (Table 2), can be used as a measure of the metal-metal bond strength and since the critical size strongly influences the final particle size, there should be a correlation between ΔH_{vap} of a metal and the size of the metal nanoparticle. More specifically, for a metal with high value of ΔH_{vap}, nucleation should be favored over growth, thus resulting in smaller particles. This hypothesis is consistent with literature data regarding the preparation of transition metals nanoparticles (Finney and Fink, 2008; Wegner et al, 2001; Li and Truhlar, 2008; Bondi et al, 2009; Schladt et al, 2009; Narayana and El-Sayed, 2005).

Table 2: ΔH_{vap} values of selected heavy metals at boiling point. (Gale and Totemeier, 2004)

Metal	ΔH_{vap} *(kJ/mol)*
Ag	257.8
Al	290.9
Cd	99.6
Cr	342.1
Cu	304.8
Fe	340.4
Hg	61.1
Mn	231.1
Ni	374.3
Pb	178.8
Zn	114.3

Following combustion, as the gaseous products travel through the flue gas system, the heavy metals are subject to homogeneous nucleation and condensation onto fly ash particles (Hester and Harrison, 2002); these reactions are followed by surface growth.

The final particle size depends on the residence time of the flue gas in the combustion system. Longer residence times provide more opportunity for particles to coagulate and grow (Liank and Wendt, 1993).

3.1 UFP emission from coal power plants

Combustion of coal in thermoelectric power plants is a major source for direct emission of UFP. In 2007, almost 50% of the approximately 4.16 billion MWh generated in the United States, corresponding to 1.047 billion tons of coal, were produced by coal-fired power plants (DOE).

Coal naturally contains metals in various concentrations. Table 3 shows the concentration ranges of some of the trace elements found in coal. Li et al (2009) investigated the particle size distribution produced by burning coal in a pilot scale pulverized coal combustor, where a single-wire tubular electrostatic precipitator (ESP) was used to collect fly ash particles. The particle size distribution at the ESP inlet showed a high number concentration of particles with the mode around 75nm and number concentration of about 10^8 particles/cm^3. At the ESP outlet, the number concentration of particles larger than 70 nm decreased about three orders of magnitude. However, the reduction in the number concentration of particles smaller than 70 nm decreased significantly at particle sizes below 70 nm. Wang et al (2008) observed a number concentration of $6x10^8$ particles/cm^3 for particles in the size range of 5.6-560nm in a coal-fired power plant in China.

Table 3: Concentrations of some trace elements in coal (Ruth, 1998)

Element	*Concentration range (ppm)*
Zn	10-400
Pb	5-55
Cu	8-30
Ni	3-30
Cd	<0.1-7
Hg	0.1-0.2

Analysis of fly-ash samples from coal burned in laboratory combustors showed that the chemical composition of the ultrafine fraction is significantly different than the composition of the fine and coarse fractions. It was observed that the ultrafine fraction contained higher concentrations of trace elements such as Cu, Pb, Cr, Mn, Zn, As, and Se. In some cases, the metal concentration in the UFP was 50 times higher than in the fine and coarse particles (Seames and Wendt, 2000; sui et al, 2007; Wang et al, 2008). The enrichment of metals in UFP has also been observed in stack gas samples taken from coal-fired power plants (Yoo et al, 2005). These observations are consistent with the hypothesis that metal vapors nucleate to form UFP. Table 4 summarizes findings from various research studies regarding metal enrichment in UFP from coal combustion.

Table 4: Enrichment of metals in UFP- summary of various studies

Study	*UFP Enriched*
Seames and Wendt, 2000	As, Cd, Se
Yoo et al, 2005	Cd, Cr, Mg, Mn, Ni, Pb,
Sui et al, 2007	Ca, Cr, Cu, Fe, Mg, Mn, Pb, Zn
Wang et al, 2008	As, Cd, S, Sb, Se, Pb, and to a lesser extent: Zn, Cr, Ni, Cu, V, Co, Cr, Ni, Mg, Mn

3.2 UFP emission from biomass-fired power plants

Biomass combustion is used extensively to generate heat and power from organic-matter derived fuels, such as agricultural and wood residues (Wiinikka et al, 2006). The energy from biomass combustion used for heat and electricity generation and for transportation (biofuels) accounts for approximately 1.4% of the energy use worldwide (Goldemberg and Johansson, 2004). Utilization of biomass within the European Union has increased significantly over the last decades and has almost tripled in comparison to its use in 1999 (Khan et al, 2008). In 2007, 1.3% of the electric power in the United States was generated by biomass combustion (DOE).

Biomass contains various trace elements such as Zn, Cd, Cu, Cr, Pb, and Hg, which vary considerably, depending on the biomass type and source (Demibras, 2005). Biomass combustion leads to relatively high PM emissions and the particle size distribution varies for different fuels and combustor systems (Nussbaumer, 2003). Combustion of wood in different laboratory scale combustion systems showed that the majority of the particles emitted were in the size range of 30 to 300 nm with a mode of 80 nm (Wieser and Gaegauf, 2000). The particle number concentration varied widely from10^7-10^{10} particles/cm^3. Chemical analysis of particles emitted from wood combustion showed that metals such as Zn and Mg were concentrated in the UFP (Johansson et al, 2003; Tissari et al, 2008). The number concentration of particles in the size range of 17 to 300 nm emitted from boilers operating on three different biofuels (sawdust, wood pallets, and forest residues) ranged from 6.3×10^7 to 7.7×10^7 particles/cm^3 with a particle size mode ranging between approximately 70-100 nm (Weirzbicka et al, 2005). The concentration of Zn, Cd, Pb and Cr in the particles depended on the type of fuel, but was generally significantly higher in the smaller particles.

3.3 UFP emission from WTE plants

Incineration of waste is used throughout the world for treatment and disposal of MSW. In the U.S. there are 87 WTE facilities that process a total of approximately 29 million U.S. tons of municipal solid waste (MSW) (Themelis, 2005). A typical calorific value for MSW is about 9000 kJ/kg (Hester and Harrison, 2002). In 2007, WTE accounted for 0.3% of the electric power produced in the United States (DOE).

MSW contains variable amounts of metals. Sources of heavy metals in MSW include batteries, electronic devices, light bulbs, house dust and paint chips, food containers, used motor oils, plastics, yard wastes and some papers (Hasselriis and Licata , 1995).

In a WTE facility in Germany, Ultrafine particles were detected in the flue gas at various gas temperatures. Samples were collected from the flue gas at 700°C (prior to boiler), 300°C (after the boiler), and from the stack at 80°C (Maguhn et al, 2003). At the 700°C sampling point, a bimodal size distribution was observed with the one maximum at about 80 nm and the other at 40 nm. At the 300°C sampling point, the observed size distribution showed a shift of the main maximum to a larger particle size of about 140 nm, probably due to coagulation of particles upon travel through the boiler section. The particle size distribution of the stack samples showed a significant reduction in the total number of particles in comparison with the 300°C, due to particulate control (ESP and fabric filters).The measured particle size distribution ranged from approximately 1 μm to 17 nm with a broad maximum at about 40-70 nm and a UFP number concentration of about 10^5 particles/cm^3.

In a very recent study, the particle size distribution observed in the stack flue gas of a WTE plant in Italy (Buonanno et al, 2009) was somewhat similar to that observed by Maguhn et al, with a peak at around 80nm. The number concentration varied among the different samples, from $2x10^5$ to $1x10^5$ particles/cm^3, and 65% of the total number of particles were in the ultrafine range. The flue gas cleaning methods used in this facility were selective non catalytic reduction of NOx (SNCR), ESP, spray absorber system and fabric filters.

UFP emissions from WTE are challenging to characterize due to the variability of MSW fed to the combustion unit. Cernuschi et al (2009) observed a significant variability in the mean number concentration of UFP in the stack gas of three Italian WTE plants ranging from $4x10^3$ to $7x10^4$ particles/cm^3.

Chemical composition of WTE UFP

In the above studies, information about the waste composition, particularly the metal content, was not provided and the UFP were not analyzed for their chemical composition. However, since UFP generated from both coal and biomass combustion were found to be enriched in metals, it is expected to find the same trend in UFP generated by MSW combustion. In a laboratory experiment, Tsukada et al (2008) showed that the particle size distribution in a modeled WTE exhaust gas, generated by sublimation of $PbCl_2$ and $CdCl_2$, ranged from 3 nm to 300 nm and most of the particles were of diameter less than 100 nm. This finding is in agreement with the concept of UFP formation from metal vapor (in this case in chloride form) generated by waste combustion.

3.4 Comparison of UFP number concentration in gas from combustion of different fuels

Table 5 provides a comparison of the number of ultrafine (or near ultrafine) particles emitted by combustion of several types of fuels as observed in different research studies. Of course, the number concentration of the particles measured in the stack is likely to be affected by the air pollution control methods used. It is therefore difficult to relate the observed number concentration solely to the fuel composition and combustion process. However, in a laboratory-scale test of fluidized bed combustion of different solid fuels, the number concentration of UFP generated by the combustion of refuse derived fuel (RDF; shredded and pre-sorted MSW) was about half than from biomass and about 14 times lower than from bituminous coal combustion (Urciuolo et al, 2008). This observation is interesting since refuse derived fuel should contain a smaller amount of metals than as-received MSW.

Table 5: UFP emission from combustion of different fuels*

Paper ***(System)***	***Fuel used***	***Air pollution control (APC) system***	***UFP number concentration in stack gas (particles/cm^3)***
Wang et al, 2008 (Industrial power plant)	Coal	ESP	$6\text{x}10^8$ ($5.6<d_p<560$ nm)
Wierzbicka et al, 2005 (district heating units)	Sawdust, Pellets, Forest residues	Multicyclones	Sawdust: $6.7\text{x}10^7$ Pellets: $6.3\text{x}10^7$ Forest residues: $7.7\text{x}10^7$ ($17<d_p<300$ nm)
Urciuolo et al, 2008 (Experimental fluid bed reactor)	Bituminous coal, Biomass (pine seed shells), Commercial pre-dried granulated sludge (GC), Refuse derived fuel (RDF)	No Air Pollution Control system	Coal: $1.1\text{x}10^7$ Biomass: $1.49\text{x}10^6$ GC: $1.06\text{x}10^7$ RDF: $7.52\text{x}10^5$
Buonanno et al, 2009 (Industrial WTE facility)	As-received MSW	ESP, spray absorber, fabric filter, selective non-catalytic reduction	~ $1\text{x}10^5$
Cernuschi et al, 2009 (3 industrial WTE facilities)	As-received MSW	**Plant 1**: ESP, dry absorption system, fabric filter, selective non-catalytic reduction. **Plant 2**: dry absorption system, fabric filter, selective non-catalytic reduction. **Plant 3**: dry absorption system, fabric filter, selective non-catalytic reduction, wet absorption system, quencher.	**Plant 1**: $3\text{x}10^3$-$1.7\text{x}10^4$ **Plant 2:** $4\text{x}10^3$-$7\text{x}10^3$ **Plant 3:** $2.45\text{x}10^4$-$7\text{x}10^4$

* Data are not normalized

3.5. Comparison of UFP emissions from coal-fired and WTE plants in the U.S.

As shown in Table 5, on the basis of studies carried out on different fuels the overall number concentration of UFP emitted from coal-fired power plants in the U.S. is at least one order of magnitude higher than that observed in WTE power plants. Based on the quantities of fuel combusted annually in the U.S. and the corresponding volume of stack gas generated and assuming that the average UFP number concentration in coal-fired power plants is 10^7 particles/cm^3 and in WTE facilities 10^6 particles/cm^3, it is possible to compare the respective contribution of these two types of power plants to UFP emission in the United States (Table 6). However, it should be noted that the number concentration of UFP emission from WTE plants may be considerably lower, as demonstrated by Cernuschi et al (2009).

Table 6: Comparison of total UFP emitted from coal-fired and WTE power plants in the United States assuming comparable number concentration of UFP in the stack gas. (Data for biomass-fired power plants were not available).

Fuel	*U.S. tons/year*	*Stack gas per fuel combusted (dscm/ton)*[1]	*Total stack gas (dscm/year)*	*Total UFP (particles/year)*
Coal	1.05×10^9	10000	1.05×10^{13}	1.05×10^{26}
MSW	2.90×10^7	5000	1.45×10^{11}	1.45×10^{23}

[1] Themelis, 2005

It can be seen that the volume of gas emitted from U.S. coal-fired power plants is so much greater than from WTEs that even if the same number concentration is assumed to exist in these two types of plants, the amount of UFP emissions from coal combustion is at least fifty times greater than that from WTE facilities.

4. Factors Affecting Metal Partitioning in WTE

Since ultrafine particles are associated with metals, it is first crucial to review the factors influencing the partitioning of metals between bottom ash, fly ash and flue gas. This section treats metal partitioning in WTE alone since unlike in coal and biomass fired power plants the fuel properties can be controlled to some degree. The factors that affect metal vaporization can be divided into three main categories: metal properties, waste properties, and combustion conditions.

4.1 Metal properties:

Heavy metal partitioning in waste combustion systems is related to the physiochemical properties of the metal. For example, volatile heavy metals such as Cd and Hg, are likely to be found in the flue gas while other metals with low vapor pressure and high boiling points such as Fe and Cu are usually trapped in the bottom ash (Hester and Harrison, 2002). Toledo et al (2005) found that among the tested heavy metals, Ni and Cd exhibited the most extreme behavior. Ni showed the lowest and Cd the greatest partitioning into the gas phase; this can be explained by the high and relatively lower boiling points of these two metals.

4.2 Waste properties

Metal partitioning in MSW is also affected by other factors, namely chlorine and alkaline content as well as moisture in the waste.

4.2.1 Cl content

Heavy metals are known to react with chlorine during the incineration process (Corella et al, 2000). The presence of chlorine compounds in the waste enhances formation of metal chlorides, which are more volatile than the elemental metals (Pavageau et al, 2004, Poole et al, 2007). Increased chlorine content in the waste is predicted to favor volatilization of Co, Cu, Fe, Mn, Ni, Pb, and Zn (Poole et al, 2008). Controlled lab experiments showed that addition of chlorine significantly increased the transfer of different heavy metals into the gas phase and reduced the temperature at which the transfer occurred (Belevi and Langmeier (2), 2000). For example, at 700°C 70% of Zn was transferred to the vapor phase. Addition of chlorine increased the transfer of Zn into the vapor phase to 94% The gaseous chlorides of Cd and Pb were found to be the dominant species for these metals in the temperature range of ~600°K to 1400°K. Above this temperature the dominant form is elemental gas for Cd and oxide for Pb.

The gaseous chloride of Hg was found to the major Hg species below 900°K, while above this temperature elemental gaseous Hg dominated (Abanades et al, 2002). This result is in agreement with other research studies that reported that at high temperatures increased chlorine content seems to have a negligible effect on Cd and Hg, both of which are observed to exist mostly in the elemental gaseous form (Durlak et al, 1997). The effect of chlorine is of course dependent on its availability in the waste. PVC and C_2Cl_2 compounds are effective sources of chlorine ions as they are readily decompose (Wang et al, 1998). The weight percent of Cl in MSW ranges between 0.2-0.6 while in coal it ranges between 0.005-0.6.

4.2.2 Moisture

The effect of waste moisture is interesting. On one hand, increased waste moisture results in increased HCl production, which in turn reduces the amount of available Cl that can react with metals (Durlak et al, 1997). On the other hand, waste moisture tends to reduce the flame temperature due to the latent heat of evaporation resulting in a less efficient process (Yang et al, 2004) and in conditions that are less favorable for HCl formation. It has been observed that when the temperature is kept constant, increasing waste moisture shifts lead from $PbCl_2(g)$ to $PbO(l)$ due to reduction in available chlorine. When temperature is allowed to change, increasing moisture content results in lower temperature, which in turn shifts the lead from the oxide form to the gaseous chloride form (Durlak et al, 1997). Similar results were observed in a different experiment by Wang et al (1998): They observed that heavy metals tend to form metal chlorides in the fly ash in the presence of chlorine; and to form metal oxides in the bottom ash in the absence of chlorine. The effect of moisture should be more pronounced in MSW combustion than in coal combustion since the weight percent of moisture in MSW is 25 comparing to 5 in coal (Ruth, 1998).

4.2.3 Alkaline content

The effect of alkaline metals in the waste (mainly Na and K) somewhat resembles the effect of moisture. In both cases, speciation is governed by the affinity of hydrogen, in the case of moisture, or alkaline metal to chlorine. Na and K have a stronger affinity to chlorine than heavy metals (Wang et al, 1998). This again results in less available chlorine and a consequent reduction in heavy metal volatilization. Durlak et al (1997) reported that increased Na content in the waste significantly reduced the percentage of lead in the fly ash and increased the percentage of lead in the bottom ash.

4.3 Operating conditions

Different operating conditions, such as retention time and mixing, influence the combustion kinetics and thus may affect metal partitioning (Falcoz et al, 2009). Brunner and Monch (1986) reported that when a waste of a similar composition was combusted under two different conditions but with the same gas cleaning equipment, different partitioning was observed for the metals Zn, Cd and Pb .

5. Recommendations for Reducing Metal Content of MSW and UFP Emissions in WTE Stack Gas

UFP emissions from stationary combustion sources may possibly be reduced further. This section discusses some options for decreasing such emissions. The first section below refers to WTE and the second part may be extended to coal and biomass combustion as well.

5.1 Reduction of metals in the MSW stream

For the reasons described above, reducing the metal content in the MSW feed to WTE facilities may result in a significant reduction of ultrafine particles emitted by WTE facilities. Table 7 shows that the metal content in the U.S. MSW has remained relatively constant in the period 2000-2007.

Aucott et al (2009) investigated trends in MSW metal content on the basis of approximately 12 years of data from monthly summary reports of two WTE facilities in New Jersey. An increasing trend in Cd in both facilities was observed and the authors attributed it to the increased use of Ni-Cd batteries in inexpensive electronic items. On the other hand, both facilities showed a decreasing trend in Hg, which is consistent with the decreased use of Hg in the United States. The Pb content seems to have remained constant throughout the investigated period.

Table 7: Metal content in U.S. MSW (as % of total MSW) in 2000-2007 (EPA)

Metals	***2000***	***2004***	***2005***	***2006***	***2007***
Ferrous	5.6	5.7	5.8	6.0	6.1
Aluminum	1.4	1.5	1.5	1.6	1.5
Other Nonferrous	0.3	0.3	0.3	0.3	0.3
Total	*7.3*	*7.5*	*7.7*	*7.8*	*8.0*

The U.S. RDF-type of WTE plants shred the MSW and separate some metal-containing objects from the waste delivered to the plant. However, it is not possible to completely eliminate metals in waste by using technology alone so extensive recycling practices by consumers must be reinforced. In particular, it is important for major urban centers like New York to devise ways of collecting used batteries. In Switzerland, for example, battery recycling rates increased from 61% in 2002 to 67% in 2004, following a massive battery recycling campaign that started in 2002; specially designed battery collection boxes were provided to all retail businesses selling batteries. In addition, advertising in all forms of major media took place in order to raise public awareness (Hansmann et al, 2009).

5.1.1 Reduction of metal disposal by consumers

The concept of metal elimination from MSW goes hand in hand with "reduce, reuse and recycle" philosophy of waste management. This approach requires that the consumer and the manufacturer buy recycled materials as well as recycle the product at its end use (Ludwig et al). Although "zero waste" is impossible if "recycle" does not include energy recovery by thermal treatment, wasting reduction in general and of metals in particular can be achieved by reinforcing recycling practices.

Reduction of metals in the waste stream seems to be correlated with fewer metal vapor emissions. For example, it was reported that following an aggressive recycling campaign in Warren County, New Jersey, the amount of mercury in the waste stream was reduced from 3 ppm to less than 1 ppm. The mercury emission from the WTE facility in Warren County was also the lowest compared to the other WTE facilities in the study (Themelis and Gregory, 2001).

In Germany, the law requires all manufacturers and distributors of packages to take back and recycle their used packaging. This includes food containers and plastic packaging that may contain metals. The Recycling and Waste Management Act assumes product responsibility on the producer. Among the products for which regulations have been specified are electronic scraps and batteries, both of which are significant sources of heavy metal in MSW (Ludwig et al). Switzerland employs the "polluter pays principle" (Sinha-Khetriwal et al, 2005), according to which households are taxed based on the volume of waste they generate. In order to encourage recycling, the government created financial incentives by not charging for disposing of recyclable material at designated recycling points (Burdick, 2003). Reinforcement of recycling policies requires the public's awareness and cooperation. In Sweden, for example, the law requires households to separate packaging material from the waste, clean them, use the collection systems provided by the producers, and sort different packaging material in designated recycling bins. Although these actions are obligatory, they are rarely controlled. However, according to official statistics, households in Sweden recycle vast amounts of waste regardless of the lack of control (Hage et al, 2009). Even though there are no federal regulations regarding recycling in the United States, raising public awareness in conjunction with increasing the availability of collection points may result in a significant increase in the amount of waste recycled.

Switzerland was the first country to create a formal system to manage electronic waste. Electronic waste includes smaller items such as computers and TVs as well as larger items such as washing machines and refrigerators. The Swiss system for collection and management of electronic waste is based on the "Extended Product Responsibility" (EPR) principle. According to the EPR principle both the physical and the financial responsibility for the safe disposal of electronic waste is placed on the manufacturers and importers of the products. There are hundreds of collection points as well as thousands of retail locations throughout Switzerland that are required by law to take back old electronic equipment free of charge. To help finance the system, the Swiss government introduced the "Advanced Recycling Fee" charged on all new appliances (Sinha-Khetriwal et al, 2005). In the United States, there are no federal regulations regarding recycling of electronic waste. Some states have issued mandatory electronics recovery programs (EPA). However, these are not as comprehensive as the Swiss program, for example.

An interesting observation is that those countries that have rigorous recycling programs are also the ones that combust most of their MSW and try to avoid land filling (figures 1 and 2). Switzerland, for example, combusts 100% of its unrecyclable MSW. Norway is currently on a trend of increasing waste recovery (including energy recovery) while reducing landfilling. As of 2008, the amount of waste combusted exceeds the amount of waste landfilled (SOE Norway).

Figure 1: MSW incinerated in Europe in 2007 (kg/person/year). (Eurostat)

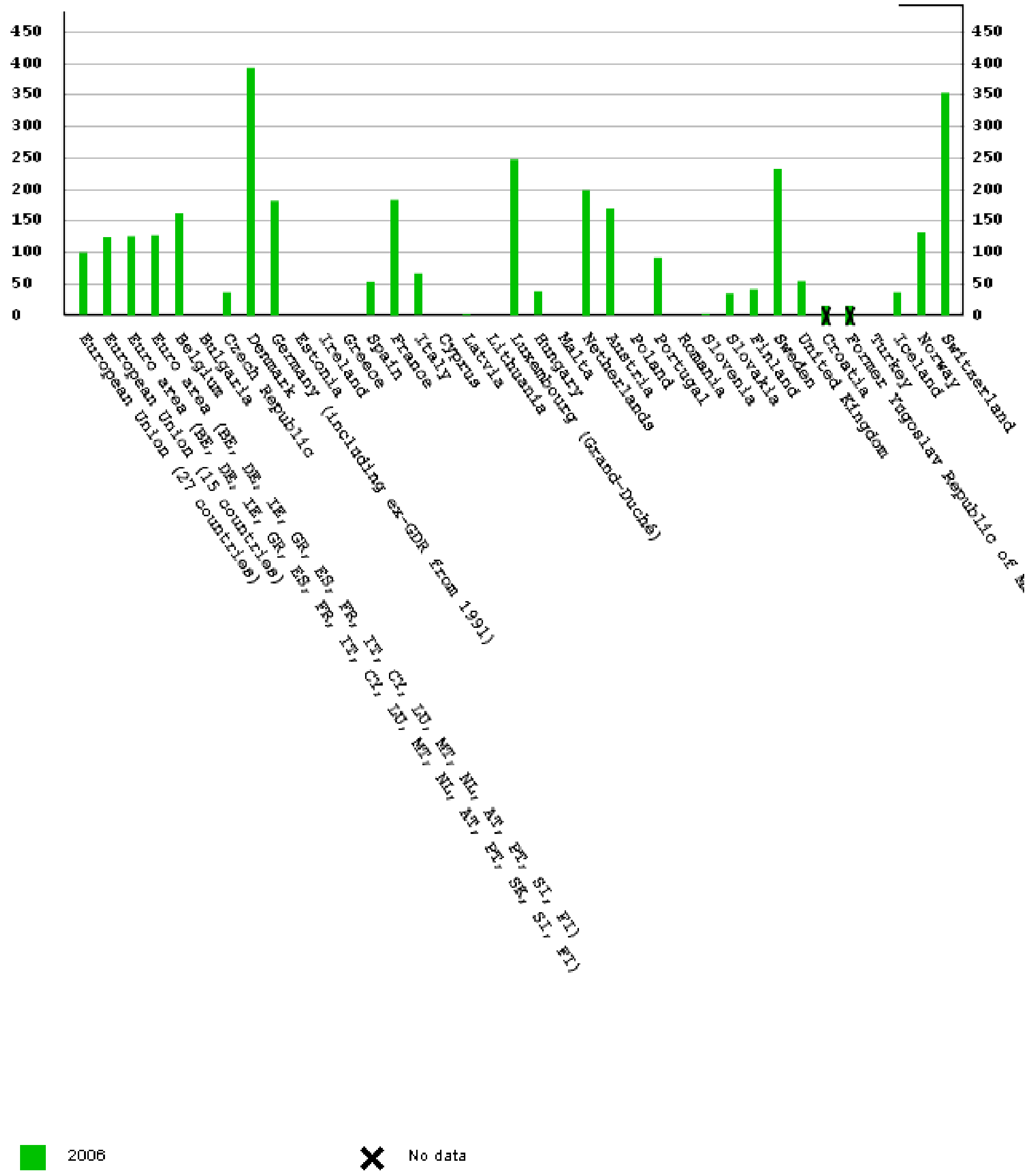
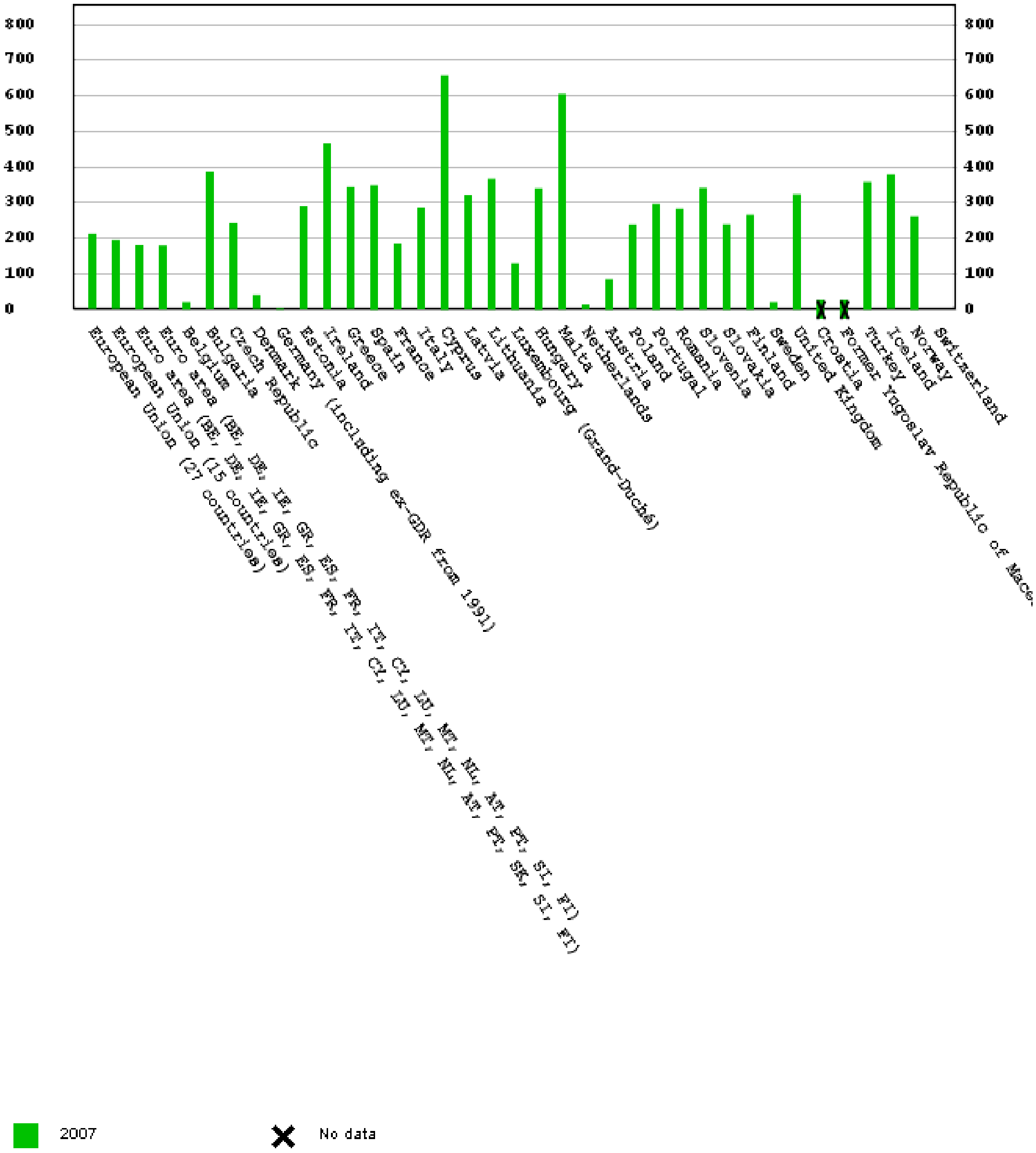


Figure 2: MSW landfilled in Europe in 2007 (kg/person/year). (Eurostat)



5.1.2 Metal separation from waste at the facilities

Sorting out the waste stream can significantly reduce metals in MSW before combustion. However, sources of metals in MSW also include coated plastics and papers which are not easy to separate. Table 8 shows the concentration of various metals in U.S. MSW and the amounts recovered in 2007. Figure 3 shows the trend in metal generation and recovery in the United States between the years 1960 to 2007. Although recovery of metals has increased over the years, more metals are being generated. Clearly, a significant improvement in metal recovery is still required. Different metal separation technologies exist:

Magnetic Separation

Magnetic separation is used to separate ferrous items such as tin cans, wires and household equipment from the waste. There is an additional value for magnetic separation if it is used before shredding the waste (in facilities that shred the waste pre combustion). In these cases, separation of ferrous items also helps reduce the wear and tear on the shredders.

Eddy-current Separation

Eddy current separation is used to separate nonferrous metals and alloys from the waste stream. It is also used to separate various nonferrous metals from each other. Eddy current separator is usually used after the ferrous metals have been sorted out using magnetic separation

Density Separation

Density separation is used to separate materials based on their density and aerodynamic properties. Usually, two products are obtained by density separation: the light fraction, which is mainly composed of organic materials, and the heavy fraction, which is mainly composed of inorganic materials such as metals, construction debris and heavy plastics.

Ballistic Separation

Other than the methods described above, there are additional separation techniques that are based on different combinations of density, resilience and surface friction (Wilson et al, 1994). These methods separate organic from inorganic materials as well as heavy from light materials. The separators based on these methods are usually low cost since the required equipment is relatively simple (Kutz, 2007).

Table 8: Distribution of metal products in MSW and the amount recovered (in millions of tons), USA 2008 (EPA)

Product Category	***Generation***	***Recovery***
Durable goods		
Steel	13.3	3.6
Aluminum	1.31	negligible
Other Nonferrous Metals*	1.76	1.21
Total Metals in Durable Goods	16.37	4.81
Containers and Packaging		
Steel	2.55	1.61
Aluminum	1.88	0.72
Total Metals in Containers and Packaging	4.43	2.33
Total Metals	***20.8***	***7.14***

* Includes lead from lead-acid batteries

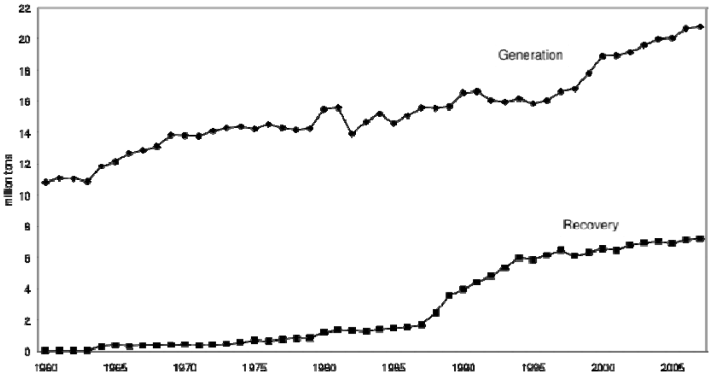


Figure 3. Metal waste generation and recovery in the US between 1960-2007. (EPA)

5.1.3 Reducing volatility of metals

Avoiding supersaturation conditions by reducing metal vapor is an additional option by which nucleation of UFP in WTE may be mitigated. Chlorine, which is contained in MSW substances such as PVC and NaCl, enhances the volatility of most heavy metals. Better waste separation and recycling measures would reduce chlorine contributed by PVC. However, it would certainly not eliminate chlorine completely. Durlak et al (1996) suggested that addition of sodium to the waste may inhibit metal chloride formation due to the greater affinity of chlorine to sodium than to heavy metals.

5.2 Improving air pollution control methods

Particulate control methods vary amongst different combustion processes. Coal power plants use mainly electrostatic precipitators (ESP) but some use fabric filters. Biomass power plants generally use either ESP or fabric filters, in some cases coupled with cyclones. Nearly all WTE plants in the U.S. use fabric filters (Ruth, 1998) but some WTE facilities in Europe, such as Technoborgo in Italy and TREA Breisgau in Germany, use ESP in addition to fabric filters.

Fabric filters are made from woven, felted, and knitted materials and are generally found as an array of long cylindrical bags called "baghouse" (Wark et al, 1998; Niessen, 2002). The collection efficiency of fabric filters is high due to the filters themselves but more importantly due to the contribution of the cake of collected dust. In general, the collection efficiency of fabric filters in terms of mass is 99.99% and over for loaded filters and below 99% for clean filters (Wark et al, 1998). The efficiency generally drops with decreasing particle size. However, the UFP collection efficiency of fabric filters as a function of size is not known (Biswas and Wu, 2005).

Electrostatic precipitation is based on the use of electrical fields as charged particles are attracted to electrodes of the opposite polarity. The collection efficiency of ESP ranges from 90% to 99.9%. However, the collection efficiency is significantly lower for UFP and goes down with decreasing particle size. For example, Li et al (2009) observed that the penetration of UFP in a coal power plant increased as the particle size decreased below 70 nm, where the most significant penetration was observed for particles smaller than 50 nm.

Cyclones operate on inertial collection principles. They are widely used as particulate control devices due to their simplicity and low cost (Biswas and Wu, 2005). However, their collection efficiency drops for particles below 100 nm (Wark et al).

Table 9: Comparison of the main particulate control methods used by power plants (after Hester and Harrison, 1994).

Method	***Description***	***Key points***
Fabric Filters	Permeable bags which allow the passage of gas but not particles. Build up of cake on the filter surface acts as a medium itself.	- High efficiency for removal of particles down to submicron range - UFP collection efficiency as a function of size is not known - Efficiency affected by pressure drop
ESP	Particles in gas stream are electrically charged and separated from the gas stream.	- High efficiency for removal of particles down to submicron range - Efficiency is significantly lower for UFP - Collection efficiency depends on gas velocity and residence time
Cyclones	Inertial separation	- Not efficient for UFP removal - Cannot handle wet flue gas - Fluctuations in flow volume and density affect efficiency

This section discusses a few options for improving the efficiency of current particulate control methods.

5.2.1 Condensation/adsorption of metal vapors onto injected particles

Homogeneous nucleation may be suppressed if metal vapor is condensed onto sorbent particles that are injected into the flue gas, thus avoiding supersaturation conditions. It is desirable to promote condensation of metal vapors on larger particles since they are more easily and effectively captured by air pollution control methods. This concept is used in combustion systems for removal of gases such as

HCl and SO_2 (dry scrubbing). Also in WTE facilities, activated carbon particles, added at concentration levels of 0.1-0.5 g/m^3 of flue gas, are used to efficiently remove Hg and Cd vapor (Hester and Harrison, 2002). After adsorption the activated carbon particles are captured by the fabric filters which further increases the efficiency of the filters. It might be possible to extend this method for reducing metal vapors before nucleation occurs if the injected particles are introduced early enough in the process. Linak et al (2003) investigated the possibility to reduce homogeneous nucleation of metal vapor from residual oil combustion. They found that injection of powdered kaolinite sorbent (d=1.4 μm) into the combustor post flame reduced the ultrafine particle volume by approximately 35% due to the interaction of Fe, Ni, V and Zn vapors with the sorbent.

5.2.2 Scavenging of nuclei

Once nucleation occurs, particles may grow by intra-coagulation or by inter-coagulation with the coarse particles. Intra-coagulation and inter-coagulation are competitive processes because the former increases the particle size, which makes it more difficult to be scavenged by larger particles. However, injection of coarse particles into the flue gas may increase inter-coagulation rate at the expense of intra-coagulation, resulting in a larger particle size distribution that may be more easily captured using current air pollution control technologies. Friedlander et al (1991) argued that if the inter-coagulation rate is small compared to the residence time, a considerable scavenging of ultrafine particles by coarse particles can be achieved.

Lee and Wu (2005) developed a model that predicts higher inter-coagulation rates with the following conditions: high number concentration of coarse mode, a large mean size difference between fine and coarse modes, and a narrow geometric standard deviation of the fine mode. They argue that for practical application of the model, monodisperse sorbent particles with mean diameter close to 1μm and a number concentration of more than $10^7/cm^3$, would be the most effective. It is very likely that the activated carbon particles injected in WTE plants to capture metal and organic molecules also reduce the number concentration of emitted UFP by scavenging some of the particles. However, more research is required in order to determine the effect of activated carbon particles in WTE on nuclei scavenging.

5.2.3 Enhancing charging process

The relatively low UFP collection efficiency of ESP is attributed to the low charging fraction of ultrafine particles (Biswas and Wu, 2005). Different charging techniques may increase the collection efficiency of ultrafine particles by ESP (Li and Christofides, 2006). In a laboratory experiment, a higher collection efficiency of ultrafine particles was achieved after the particles were charged by means of a dielectric barrier (Byeon et al, 2006). Another laboratory study showed that increasing the voltage applied on the particles increased the charged UFP fraction, thus resulting in a higher collection efficiency of the ESP (Suriyawong et al, 2008). However, further research is required to determine the feasibility of this approach.

6. Conclusions

Research studies have demonstrated that exposure to UFP can cause adverse health effects, some of which are severe. Power plants are a significant anthropogenic source of UFP and current particulate control methods are not very efficient in controlling these emissions. In the United States, the overall contribution of WTE and biomass to UFP emission is very small relative to coal power plants, commensurate with the lower scale of energy production from MSW and biomass. Assuming 10^6 and 10^7 particles/cm^3 of stack gas generated by WTE and coal-fired power plants respectively, it was estimated that the total number of UFP emitted annually by coal-fired power plants is nearly three orders of magnitude higher than the emissions of WTE facilities. Although UFP emissions from WTE plants are not well characterized as yet, the few research studies conducted show a significant variability ranging from a few thousands to about 10^6 particles/cm^3. Thus, the overall actual contribution of WTE to UFP emission might be even lower than calculated based on the above assumptions.

WTE has a greater potential of becoming even cleaner process since, unlike in coal and biomass, the fuel properties, namely metal content and to a lesser extent chlorine content, can be controlled by means of pre-sorting the MSW, as in the case of RDF WTE facilities. High moisture content in MSW may be responsible for inhibiting some UFP formation, while the activated carbon used in the WTE APC systems certainly helps to capture volatile metals and may also reduce UFP emission by means of adsorption. Moreover, the issue of UFP emission from WTE and the associated health impacts should be considered in the context of alternatives for waste treatment. For example, it has been determined that the health effects of landfills are greater than those of WTE. However, both were considered to present insignificant health risks ($1/10^6$ risk of cancer for landfills and $2/10^7$ for WTE) (Pearl Moy, MS thesis, 2005).

Waste combustion is an environmentally conscious process when it is done properly. Countries such as Switzerland, Norway and Sweden, that were ranked as the top three nations in the Environmental Performance Index (2008), combust almost all of their non-recyclable waste. These countries have rigorous recycling policies, which play an important role in the environmental impacts of waste combustion. The primary responsibility in separating metals from waste should be on the consumers. Thus, public awareness and legislation in the United States are necessary. Recovery of metals at the source, such as small batteries, fluorescent lights, etc. should become customary. Improvement in the particulate control methods, to address UFP emissions, is needed in all stationary combustion sources. A few possibilities exist but more research in that area is required.

Finally, further research is required in order to better characterize UFP emission from coal-fired, biomass-fired and WTE power plants in terms of chemical composition and number concentration.

7. Suggestions for future research

This study indicated that further research is needed in order to better characterize UFP emission from coal-fired, biomass-fired and WTE power plants, both in terms of chemical composition and number concentration. The initial plan in this thesis was to also include field tests on the analysis of samples of stack gas from a coal-fired and a WTE power plant for total number concentration and elemental composition. Although this part was not carried out eventually, we have designed an experimental setup that may be implemented in the future, as described below.

The collection and separation of particles into size bins is to be done using an impactor. The most widely used instruments for this purpose are: Microanalysis Particle Sampler (MPS), Micro-Orifice Uniform-Deposit Impactor for nanoparticles (nano-MOUDI), and Dekati Low Pressure Impactor (DLPI). Both MPS and nano-MOUDI separate particles into three size bins and gravimetric analysis of the samples would give a rough size distribution. DLPI has 13 stages and thus would be a better fit for isolating UFP. The particle size distribution can be done using Scanning Mobility Particle Sizer (SMPS). We have not decided whether or not to include sample dilution, which is used to simulate the evolution of the particle population after emission from the stack. The departmental laboratories do not have the necessary equipment for dilution. However, it can be purchased from the DLPI manufacturer or borrowed from our collaborators at Clarkson University.

Elemental composition analysis can be done using proton induced x-ray emission (PIXE) or using x-ray fluorescence (XRF). Both methods are sensitive even to very low concentrations so sampling time may be short. In addition, Samples are analyzed directly from the substrate (either Nylon or Teflon) so no preparation is required.

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