

The emission of nanoparticles from MSW combustion

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Summary

The small (nano-size) airborne particles released into the atmosphere grow during their lifetime by a dynamic process of accretion, mostly by inorganic salts such as nitrates. The origin of the original nuclei is overwhelmingly from combustion processes. Nanoparticles stay in suspension for long periods, and are transported over intercontinental distances. Organic compounds and metals are found to some extent in all samples of ambient air. The species which are responsible for health impacts are present in material from all sources. Motor vehicles produce most fine particles and dominate the generation of urban pollution. Uncontrolled emissions from bushfires, backyard burning and other internal combustion engines are also a significant contributor to poor air quality. Because the particulate emissions from a well-designed waste-to-energy (WtE) plant, before release into the atmosphere, are of the same order as in the ambient air above a modern city site, they will have a negligible addition to the overall sum of particulates in an urban environment.

Introduction

A nanometre, which is one billionth of a metre ($1 \text{ nm} = 10^{-9} \text{ m} = 0.001 \text{ micrometres } (\mu\text{m})$), is the dimension used to measure extremely fine particles. Recent developments in aerosol technology have indicated that particles smaller than $0.1 \mu\text{m}$ (100 nm) or “ultrafines” are responsible for the adverse effects on human health associated with particulate air pollution. Recent toxicological studies have shown that concentrated airborne particles ($\text{PM}_{2.5}$, i.e. Particulate Matter with a size less than or equal to $2.5 \mu\text{m}$) can induce pulmonary inflammation, chronic bronchitis, pulmonary hypertension and electrocardial changes (Sondreal et al 2000). It is hypothesised that PM pulmonary irritants trigger a nerve response that increases the heart rate and decreases heart rate variability. There is an inflammatory response to ultrafine particles (defined here as $< 20 \text{ nm}$) and the chemical effects of acids, peroxides, nitrates, sulphates, organic carbon and acid aldehydes must be considered.

The tiny mass involved in airborne solids and the small size of the particles requires that measurement is often reported as the number of particles per unit volume of gas. A typical sample of urban air contains $10 - 100 \times 10^3$ particles per cm^3 , which on a mass basis may be 20 to $100 \mu\text{g}\cdot\text{m}^{-3}$. The number concentration is dominated by fine particles, and the mass loading by large particles. Special equipment is required for sampling fine particles to avoid artefacts. The

number analysis of ultrafines is carried out by recently developed instruments, namely the scanning mobility particle sizer (SMPS) and the electronic low pressure impactor (ELPI), which also gives a size distribution.

The fine particles in the atmosphere are called aerosols because they form by condensation on a smaller nucleus while in suspension. In general, aerosols begin life in the nucleation mode (1 – 2 nm), formed predominantly in combustion systems. This process is depicted in Figure 1 for diesel particulates. Both gasoline and diesel motors are responsible for a large part of the nucleation mode particles in the urban environment. The particles then grow by coagulation when moisture, organic molecules, and ammonium, sulphate and nitrate radicals are adsorbed onto their surface (accumulation mode). The large sizes (“coarse fraction” > 1 μm) settle quickly and are not present in high concentrations.

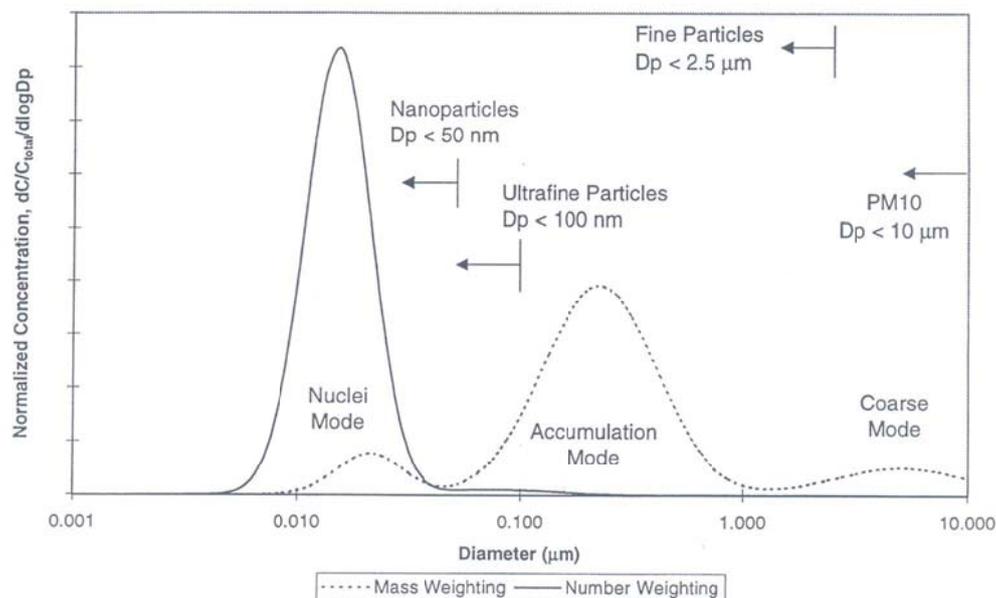


Figure 1 Diesel exhaust emissions showing the effect of accumulation

The adverse health effects noted can be related to the species listed above, and to polyaromatic hydrocarbons (PAH), which with their oxy and nitro derivatives, are present on the surface. To understand the likely effects of WtE operations on air quality, a comparison will be made between ambient city air and WtE and other emission sources.

Ambient air

Airborne primary particles are often carbonaceous and can be separated into two categories: 1) elemental carbon (EC) and 2) organic carbon (OC) of low volatility. The ratio OC/EC is normally much higher than unity in both urban and rural atmospheres. The material comprising the

remainder is mainly inorganic ions such as ammonium, nitrate and sulphate. The atmosphere contains numerous fine particles, of the order of 200 per cm^3 in "clean" air over the ocean, and more than 100,000 (10×10^4) per cm^3 at a polluted city site. The bulk of these are very fine particles smaller than 100 nm. The concentrations averaged over 24 hour periods in January and February 1966 at Pasadena California ranged from 7,100 to 14,000 cm^{-3} (Hughes et al 1998).

On a mass basis the values may lie between 2 and 500 $\mu\text{g.m}^{-3}$, and are dominated by micron size particles. The annual means for 1999 at 5 sites in the city of Lyon ranged from 23 to 44 $\mu\text{g.m}^{-3}$ (Coparly 2000). Pollution levels increase with population density; mean PM_{10} concentrations were 35 $\mu\text{g.m}^{-3}$ near Zurich, 80 $\mu\text{g.m}^{-3}$ near Paris and 110 $\mu\text{g.m}^{-3}$ near Tokyo (Zhiquiang et al 2000). In Mexico City which is badly polluted, the mean spatial averages across three sites in the period 2000-2002 were 35 $\mu\text{g.m}^{-3}$ for $\text{PM}_{2.5}$ and 76 $\mu\text{g.m}^{-3}$ for PM_{10} (Chow et al 2004). However, some 24 hr mean values of PM_{10} rose to as high as 184 and 267 $\mu\text{g.m}^{-3}$ in the winter of 1997.

The history of a packet of air passing over the Los Angeles basin was traced by Hughes et al (1999, 2000). The size distributions and compositions of particles sampled at two sites, Long Beach and Riverside, as measured by OPC are shown in Figures 2a and 2b. Very few particles with a diameter greater than the cut-off size of 2.6 μm appear to be present. The clean air at Long Beach had come off the ocean and after travelling inland across the city was sampled again at Riverside. The number count was converted to a mass basis using a mean particle density of 1300 kg.m^{-3} . The size distributions show an increase in the larger sizes at Riverside, and also a higher mass concentration. The concentrations of some species increase significantly, namely organic compounds, nitrate and ammonia, due mainly to the accumulation of additional material from industry and motor traffic. Although the mass of the sample had increased, the number of particles did not show a similar increase, as much of the additional mass was adsorbed onto existing particles.

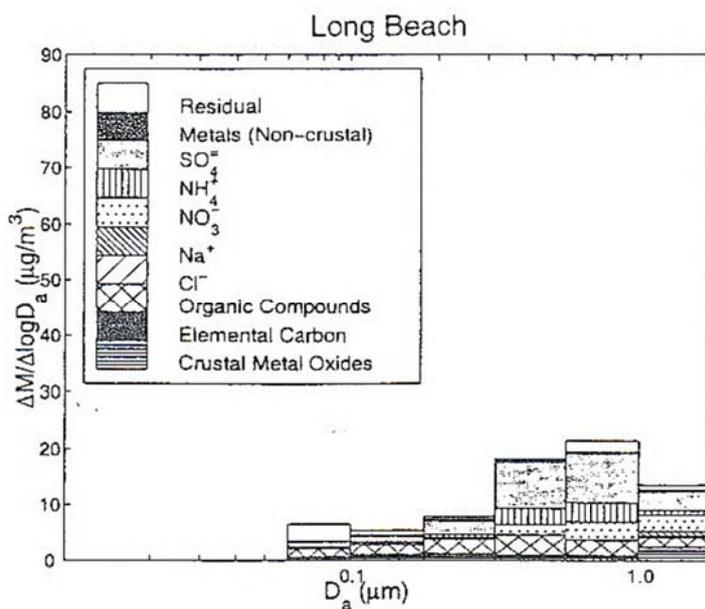


Figure 2a Particle size distribution and composition at Long Beach

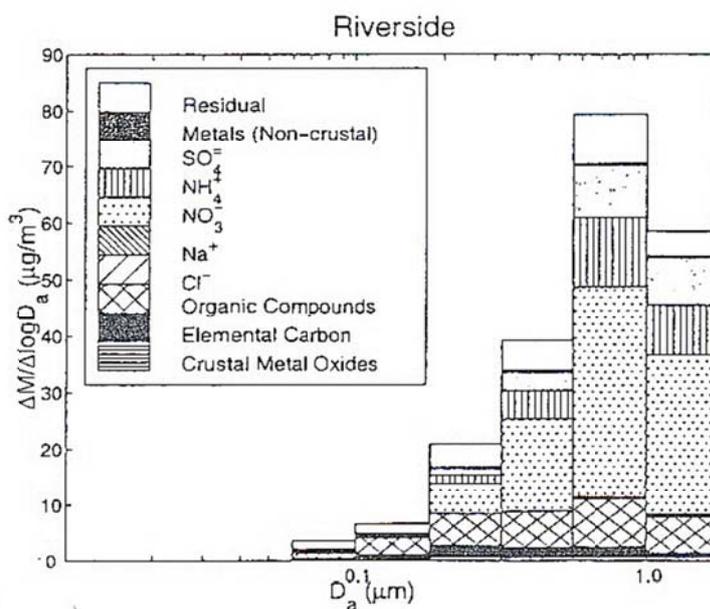


Figure 2b Particle size distribution and composition at Riverside

The metal content of these samples is divided into crustal elements (Si, Al, Fe) from dust, and non-crystal. In both cases the concentration is small. However metals are present in all samples, and have been identified in airborne particulates sampled in the arctic circle, well removed from human activity. A plot of the distribution of six common metals of environmental interest is shown as Figure 3 (Lüdke et al), which illustrates the extraordinary extent of dispersion through the atmosphere of these small particles.

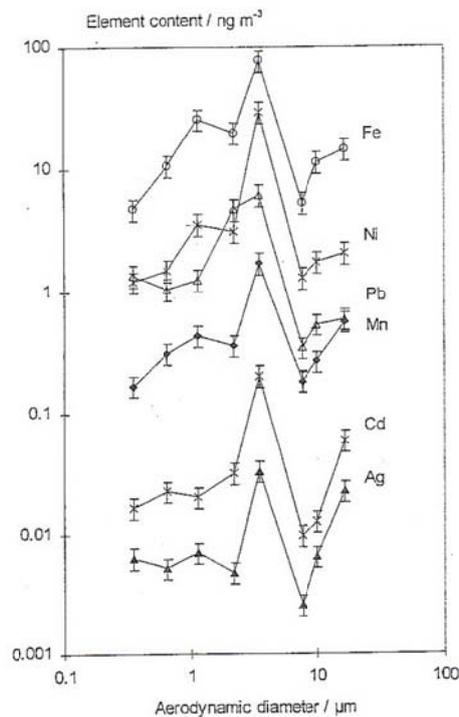


Figure 3 Concentration distribution of metals with particle size in arctic air aerosol (Lüdke et al 1999)

In the USA, the original standard for the concentration of particulate matter in ambient air was for Total Suspended Particulates (TSP). This was replaced in 1987 by a PM_{10} standard for particles with an aerodynamic diameter less than $10\ \mu\text{m}$. Later concerns focussed around the effects of even smaller particles, so that a $PM_{2.5}$ standard was added in 1997 (USEPA 1998). The current PM_{10} standard is $50\ \mu\text{g}\cdot\text{m}^{-3}$ for an annual arithmetic mean and $150\ \mu\text{g}\cdot\text{m}^{-3}$ for a 24 hour mean. The value for $PM_{2.5}$ is $15\ \mu\text{g}\cdot\text{m}^{-3}$ (as an arithmetic mean), with a 24 hour average of $65\ \mu\text{g}\cdot\text{m}^{-3}$ (Gertler 2005). There tends to be a good correlation between particle number and mass in the atmosphere, and $PM_{2.5}$ tends to be a relatively fixed proportion of PM_{10} mass (Harrison et al 2000b, 1999).

Sources

Analytical techniques have been developed to identify the original source of airborne particulates by sophisticated spectrometric techniques e.g. Cass et al 1998, Kleeman et al 1999.

Transport Vehicles

The origin of 'ultra-fine' i.e. $<100\ \text{nm}$ airborne particulates in the UK is depicted in Figure 4, where it can be seen that the majority are generated by road transport vehicles (Harrison et al 2000). Both petrol and diesel engines emit fine particles in high concentrations; untreated diesel exhaust from a modern engine contains $\sim 10^7\ \text{particles cm}^{-3}$ and a spark ignition engine $\sim 5 \times 10^5\ \text{particles cm}^{-3}$

(Kittelson). The emission rates for light duty i.e. petrol vehicles is in the range 5 to 10 mg of $PM_{2.5}$ per km, while the figure for heavy duty vehicles (trucks) is in the range of 70 to 135 $mg.km^{-1}$ (Gertler). In terms of particulate numbers in the range from 10 to 700 nm, a survey by Morawska et al (2005) shows good agreement between a number of researchers and techniques. Petrol-fuelled cars emit $1.5-2 \times 10^{13}$ particles per km and large diesel vehicles $2-4 \times 10^{14}$ per km. The percentage contribution of vehicular traffic to air quality therefore depends on the nature and use of the motor fleet. The distribution of particle sizes at the exhaust pipe of a diesel engine and later after experiencing accumulation is shown in Figure 1 (Kittelson).

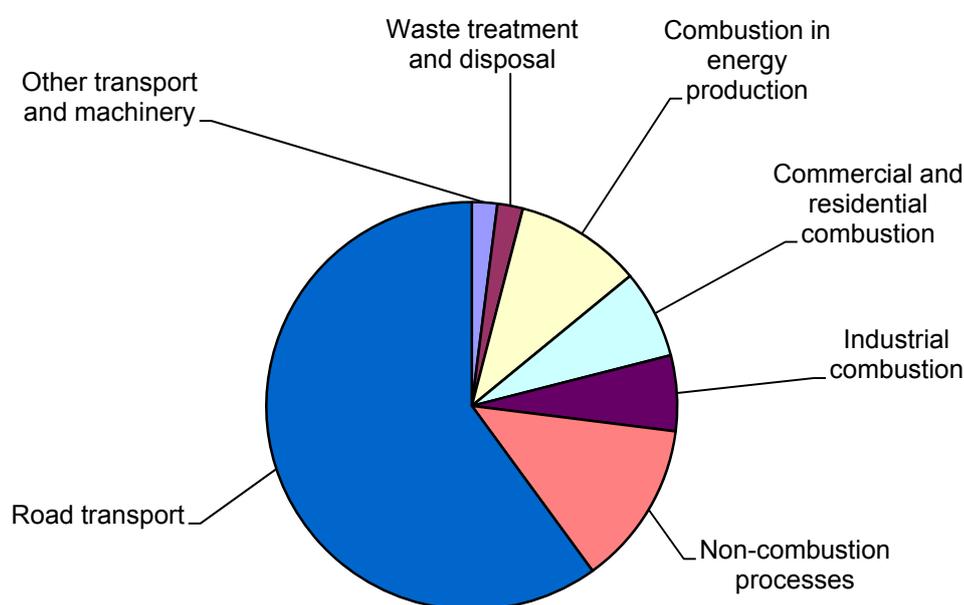


Figure 4 Ultrafine particle sources, UK 1996 (after Harrison et al 2000b)

Harrison et al (1999) and Shi et al (1999) report the size distributions of particulates sampled from a site near a busy road in Birmingham, UK. The average number concentration at the roadside over a four day period was between 1.6 and $1.9 \times 10^5 \text{ cm}^{-3}$. The background values were more consistent than the roadside ones, which could change very quickly. The background samples on a number basis showed an apparently log-normal distribution with a single mode around 30 nm. The roadside distribution also showed the 30 nm peak, together with a second one below 10 nm, the smallest size measured.

Diesel emissions contain a high fraction of elemental carbon (soot), which is a good adsorbent of organic compounds. These particles contain a range of toxic materials including metals and

organic compounds or SOF (soluble organic fraction). SOF consists of polyaromatic hydrocarbons (PAH) adsorbed onto the surface of the particulate, and comprises about 5% of the total mass at full engine load, but as much as 60% at idle. These compounds are known carcinogens, and some are present in the concentrations listed in Table 1 below. Their presence in both the gas phase and on the solid particulates is recorded. The smaller molecules tend to remain in the gas phase, while the larger ones, which are more carcinogenic, are preferentially adsorbed onto the particulates. It has been shown that there is more toxic organic free-radical activity in the smaller particles (Kittelson). The concentrations of PAH and NPAH compounds in ambient air range from 20 ng.m⁻³ in a residential area to about 100 ng.m⁻³ in the vicinity of heavy traffic. The high concentrations in the tunnel at Birmingham compared to the general urban level demonstrates the effects of local dispersion.

Table 1. Mean concentrations of PAH in urban air (ng.m⁻³)

Compound	Phase	Zurich			Birmingham		Damascus	Milan*	Rome*
		road	road	resd'l	tunnel	urban			
1-nitronaphthalene	V				1.59	0.09	0.21		
2-nitronaphthalene	V				1.25	0.07	0.16		
9-nitroanthracene	V				0.16	0.06	0.17		
	P				0.36	0.13	0.25		
1-nitropyrene	P				0.56	0.09	0.20		
benz(a)anthracene	P							2.5	0.35
benz(j)fluoranthene	P							5.5	2.0
indeno(1,2,3-cd)- pyrene	P							4.0	1.7
Total PAH	P	92	46	19				60	37

* Cecinato et al V = vapour, P = particulate; resd'l = residential

Calcium and zinc were the most common metals present in diesel particulates, at around 0.05 % concentration (Lowenthal et al).

Stationary Combustion Sources

Coal-fired power stations

Large coal-fired power stations utilise pulverised fuel firing, in which the feed is ground into a fine powder below 100 µm in size. As the carbonaceous matter burns away, the included mineral matter forms small, spherical particles generally below 10 µm in size. The conversion processes are depicted in a simplified fashion in Figure 5 (Haynes et al). In Australia the emission limits are

commonly set at around 20 - 50 mg.Nm⁻³ so that a significant amount of ash material is released into the atmosphere. However most of it is of a larger particle size which means that it readily settles to the ground.

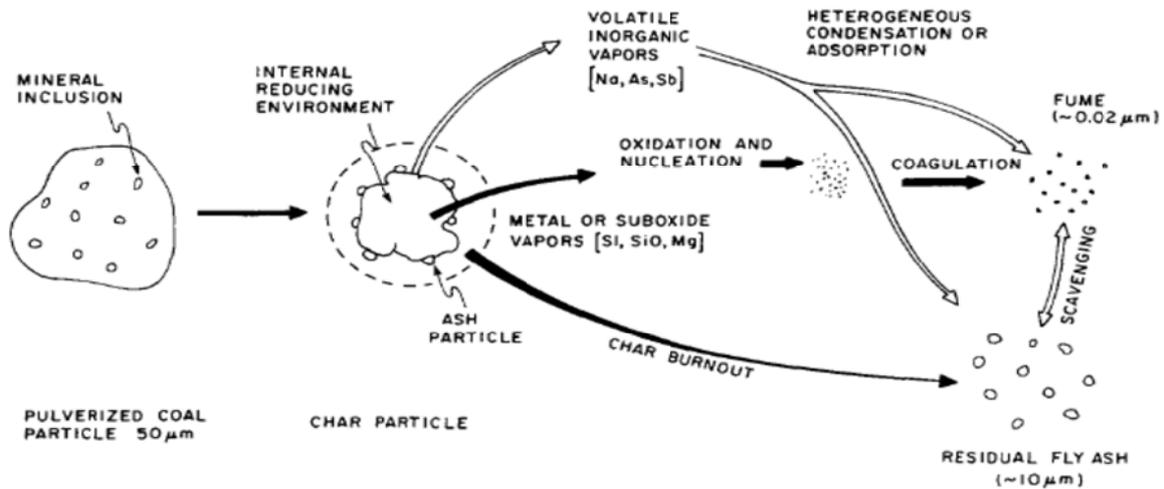


Figure 5 Ash release from a burning coal particle

Coal contains trace metals which during firing migrate into the flyash particles. Some more volatile metals are vapourised in the flame and condense as fume as the gases cool. The distributions of four metals, copper, zinc, cadmium and lead, across the size range of flyash particles from a full-scale coal-fired power station are depicted in Figure 6 (Senior et al).

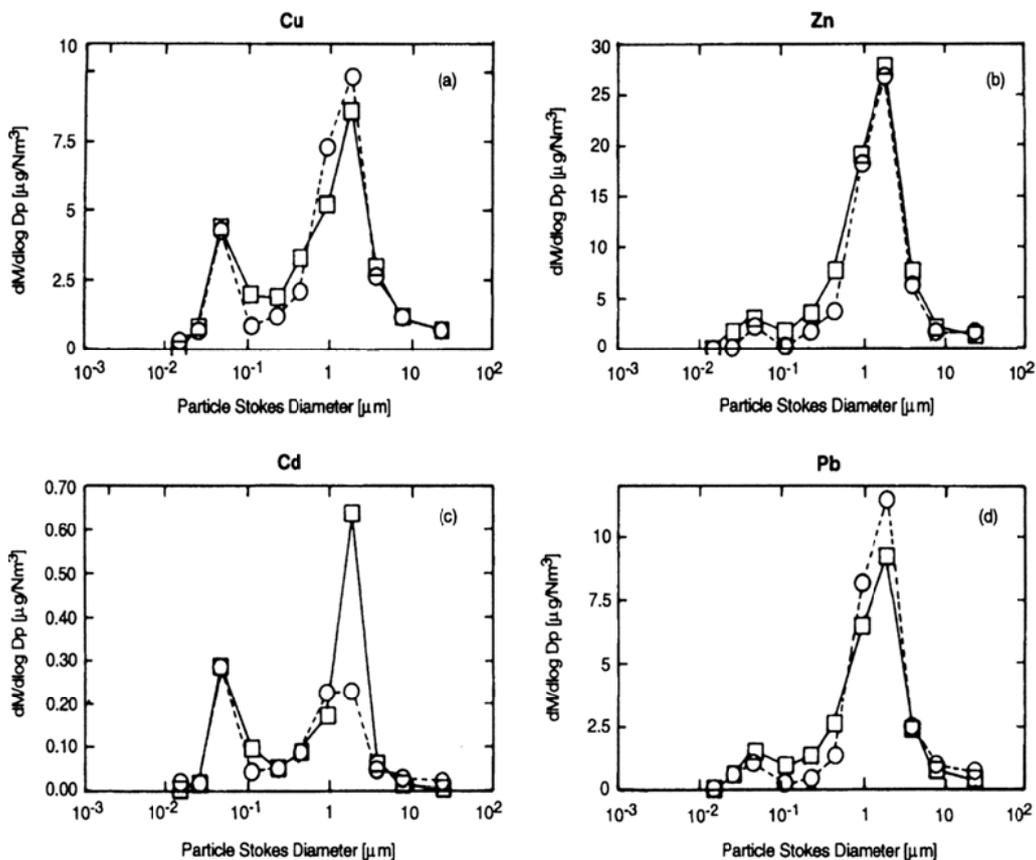


Figure 6 Metal distribution in the flyash generated by pulverised coal firing

It can be seen that there are peaks around 2 μm and 50 nm. The former would be removed almost completely in the gas cleaning system, while some of the latter would escape into the atmosphere and persist for some time. A summary by Pavagau et al of metal emissions in both the vapour/fume and solid phases from one coal-fired station is given in Table 2, with mean values reported here.

Table 2 Metal concentrations in the vapour and solid phases from coal firing ($\mu\text{g Nm}^{-3}$)

Metal	Cd	Hg	Tl	As	Se	Te	Sb	Cr	Co	Cu	Sn	Mn	Ni	Pb	V	Zn
Vapour	14	18	<1	3	7	21	62	265	10	58	37	20	37	340	4	1500
Solid	<1	<0.06	<1	62	<0.5	<1	5	27	10	44	3	120	29	45	96	530

Waste-to-Energy Plants

The amount of emissions depends on the fuel and the combustion aerodynamics, but primarily on the gas cleaning technology. Fabric filters capture 99.99% of particles released, but they are least efficient (~95%) in the 400 to 1000 nm range. Most fine particles consist of salts (Zeuthen et al). MSW stack emissions have been measured at $6.9 \times 10^4 \text{ cm}^{-3}$ (Zeuthen) and $10 - 20 \times 10^4 \text{ cm}^{-3}$ (Buananno et al). The mass loadings for tests on two MSW stack emissions were $100 \mu\text{g.m}^{-3}$ (Buananno) and $300 \mu\text{g.m}^{-3}$ (Lind et al). Thus the particulate concentrations emerging from these stacks would be indistinguishable from ambient air sampled at a central city site, and only 5 to 20 times higher than in the relatively clean air of a coastal suburb (see above). In a very short time after discharge they would have been diluted to ambient levels.

Jay and Stieglitz (1995) sampled the stack of a WtE plant in 1994 and found hundreds of organic compounds in very low concentrations, with sums of $189 \mu\text{g.Nm}^{-3}$ for aliphatics and $291 \mu\text{g.Nm}^{-3}$ for aromatics. The plant studied was of a previous design generation, and the emissions from a modern plant would be much lower than this. The concentration of heavy metals in MSW particulates has been extensively measured. The mean emissions from four Italian plants are given by Consonni et al as $\text{PM}_{10} = 10 \text{ g per tonne of MSW}$, and the metals cadmium 55 ng.t^{-1} and lead 520 ng.t^{-1} .

Buananno gives an interesting comparison between emissions from a modern WtE plant and a 3 kilometre stretch of highway. It requires only 20 vehicles (7% trucks) to traverse the section in order to produce the same particulate emissions as the plant operation for one hour.

Other Sources

In a study of aerosols in the USA, Sarofim (2001) totalled the relative emissions from a range of sources and concluded that “wood stoves therefore emit twice as much particulate matter as coal-fired power plants. There is a trend of the small, less regulated sources, becoming the dominant contributors to the particulate emissions. Wood stoves, leaf burning, off-road vehicles, snowmobiles, burning trash in barrels contribute to total particulate emissions a far greater fraction than is represented by the fraction of energy release because their emissions are uncontrolled”. In Australia the equivalent to snowmobiles would be outboard motors and jet skis. It is of interest that the Australian EPA attributes measured peaks in dioxin concentrations in city atmospheres during winter to uncontrolled burning i.e. domestic fires burning poor quality fuels (EPA 2004).

The fine particles ($PM_{2.5}$) emitted during the fireplace combustion of woods grown in the USA were studied by Fine et al (2001, 2002). The mass emissions averaged 4 g per kg of wood burned, and over 80% of this mass consisted of organic carbon. Particulate emissions at these levels are about 1000 times higher than from MSW combustion on an equivalent energy release basis. The particle size distributions showed little variation in timber from tree to tree, with the peak in number distribution occurring between 100 and 200 nm. Detailed analyses of the compounds present in the smoke from woods sampled from both the north-eastern and southern parts of the USA are presented.

Evaluation of the likely impact of a WtE plant on air quality

Two considerations are relevant:

1. Most of the mass of aerosols is not due to the primary source, but has accumulated during transport in the atmosphere. As a result their toxicity will primarily be the result of accreted material.
2. The emissions of particulate matter from a modern WtE plant are inherently low and are insignificant against the background of particulates in an urban airshed. Using the figures of Morawska et al, the average emissions from the motor vehicle fleet with 7% heavy duty units is 7.5×10^{13} particles per km. At an average yearly distance travelled of 15,000 km, this gives 1.1×10^{18} particles emitted per annum per vehicle. A 100,000 tpa WtE plant would emit about 7×10^{19} particles per year if the figures of Buananno et al are used. The plant would therefore contribute the same nanoparticle emissions as about 65 vehicles. If the figure of Zeuthen et al is used, the number falls to 30. In absolute terms, the Perth airshed is estimated to absorb about 1.3×10^{24} particles per annum from > 1,000,000 motor vehicles, so that a plant would contribute on a percentage basis either $7 \times 10^{19} \times 100 / 1.3 \times 10^{24}$, i.e. 0.0054%, or half that amount, if the figure of Zeuthen et al is used.

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