

Riverside Energy Park

Post Hearing Note on Public Health and Evidence

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1 Introduction

- 1.1.1 The application for the Riverside Energy Park (REP) Development Consent Order (DCO) was submitted to the Secretary of State on 16 November 2018. An application for an Environmental Permit (EP) to operate REP was submitted to the Environment Agency (EA) in December 2018.
- 1.1.2 Concerns have been raised about the impacts of REP on human health in a number of Relevant Representations and Written Representations submitted to the REP Examination, and were also raised or re-iterated orally at the Open Floor Hearing on 4 June 2019, and the Issue Specific Hearing (ISH) on Environmental Matters on 5 June 2019.
- 1.1.3 The Applicant responded to matters regarding human health raised in Relevant Representations in '**The Applicant responses to Relevant Representations**' (8.02.03; REP2-054) submitted at Deadline 2.
- 1.1.4 A written summary from the ISH on Environmental Matters held on 5 June 2019 has been submitted by the Applicant at Deadline 3 (8.02.19). The Applicant has also responded to Written Representations covering health in its responses to Written Representations submitted at Deadline 3 (8.02.14). This further information note has been provided in support of the Applicant's oral responses made during the hearings and its responses to both the Relevant Representations and Written Representations.
- 1.1.5 Specific environmental assessments have been carried out and were included in the DCO Application which are reported in the following documents:
- **Chapter 7 Air Quality** of the **Environmental Statement (ES)** (6.1; REP2-019);
 - **Appendix K.1 Health Impact Assessment (HIA)** of the **Environmental Statement (ES)** (6.3; APP-094); and
 - **Appendix C.3 Human Health Risk Assessment (HHRA)** of the **Environmental Statement (ES)** (6.3; REP2-040).
- 1.1.6 No significant effects were identified, but the detail relating to the conclusions of these assessments is not repeated in this note.
- 1.1.7 General concerns regarding the association of energy recovery facilities (ERFs) with health outcomes and the emission of ultrafine particles have been raised through the written and oral submissions. This note therefore seeks to respond to those general concerns and assertions, and respond referring to independent evidence.

1.2 Purpose of this Report

1.2.1 The purpose of this note is:

- to draw attention to recent research commissioned by Public Health England (PHE) on health impacts associated with ERFs; and
- to provide reassurance and further evidence regarding the emission of ultrafine particles.

1.2.2 This note demonstrates that:

- PHE considers that *“While it is not possible to rule out adverse health effects from modern, well regulated municipal waste incinerators with complete certainty, any potential damage to the health of those living close-by is likely to be very small, if detectable”*;
- research commissioned by PHE and published in 2018 and 2019 shows that there is **no evidence** that living close to an ERF is associated with increased infant mortality or other infant health risks; and
- abatement systems in place for particulate matter in ERFs are very effective at avoiding emissions of ultrafine particles.

2 Energy Recovery Facilities and Health

2.1 Public Health England Statement

- 2.1.1 The Health Protection Agency (HPA), whose role has now been taken over by PHE, published a note RCE-13 “The Impact on Health of Emissions to Air from Municipal Waste Incinerators” in 2009¹. The summary of this note is as follows:

“The Health Protection Agency has reviewed research undertaken to examine the suggested links between emissions from municipal waste incinerators and effects on health. While it is not possible to rule out adverse health effects from modern, well regulated municipal waste incinerators with complete certainty, any potential damage to the health of those living close-by is likely to be very small, if detectable. This view is based on detailed assessments of the effects of air pollutants on health and on the fact that modern and well managed municipal waste incinerators make only a very small contribution to local concentrations of air pollutants. The Committee on Carcinogenicity of Chemicals in Food, Consumer Products and the Environment has reviewed recent data and has concluded that there is no need to change its previous advice, namely that any potential risk of cancer due to residency near to municipal waste incinerators is exceedingly low and probably not measurable by the most modern techniques. Since any possible health effects are likely to be very small, if detectable, studies of public health around modern, well managed municipal waste incinerators are not recommended.” (Summary, page 1)

2.2 Public Health England (PHE) Research

- 2.2.1 While this statement is considered by the Applicant to be clear, and has been referred to in many planning decisions, PHE commissioned further research in 2012, while continuing to state that the conclusions of RCE-13 remain applicable. These studies were commissioned from the Small Area Health Statistics Unit (SAHSU), which is based at Imperial College London and Kings College London. Details of the study can be found at <https://www.sahsu.org/content/incinerators-study>. The following statement describes the aims of the study:

“The study has been commissioned to extend the evidence base and to provide further information to the public about any potential reproductive and infant health risks from MWIs [municipal waste incinerators]. The study proposes to investigate the following questions:

Are the emissions from incinerators required to operate under the standards set by the EU Waste Incineration Directive (WID)

¹ <https://www.gov.uk/government/publications/municipal-waste-incinerators-emissions-impact-on-health>

(2000/76/EC) linked with adverse reproductive and infant health outcomes?

Is living near a municipal waste incinerator linked with adverse reproductive and infant health outcomes?"

2.2.2 The methodology and results of the studies have been published in a series of papers in scientific journals. The two most recent papers are the most relevant.

2.2.3 The primary paper which provides the results of the main study is known as Ghosh et al (2018)² (provided in **Appendix A**). It considered a range of health outcomes for babies and infants - term birth weight, small for gestational age (SGA) at term, stillbirth, neonatal, post-neonatal and infant mortality, multiple births, sex ratio and preterm delivery – covering 1,025,064 births and 18,694 infant deaths. The study considered outcomes close to all 22 UK municipal ERF plants which operated at some point between 2003 and 2010. This includes the Edmonton, South East London Combined Heat and Power (SELCHP) and Lakeside facilities in and around London.

2.2.4 An earlier part of the study had involved carrying out dispersion modelling of emissions from these ERFs over the period 2003-2010³. The modelling used actual operating data from all plants to calculate the average ground level concentration of particulates down to postcode level (i.e. an area of about 12 households). As explained in Ghosh et al (2018), particulates were used as a proxy for all emissions, as all emissions to atmosphere will disperse in a similar manner. The authors of Ghosh et al (2018) then used this data and the health data to look for associations between predicted particulate concentrations and health outcomes. No associations were found.

2.2.5 The authors also looked for associations between the listed health outcomes and proximity to an ERF. This was done by identifying whether the mother's usual residence at date of birth registrations was within 10 km of an operational ERF at the date of birth. This is a less sophisticated method of analysis, as it does not take account of operational data or atmospheric dispersion patterns, but it is more similar to previous studies carried out by others. Again, no associations were found.

² Ghosh RE, Freni Sterrantino A, Douglas P, Parkes B, Fecht D, de Hoogh K, Fuller G, Gulliver J, Font A, Smith RB, Blangiardo M, Elliott P, Toledano MB, Hansell AL. Fetal growth, stillbirth, infant mortality and other birth outcomes near UK municipal waste incinerators; retrospective population based cohort and case-control study. Environment International. 2018.

³ Reported in Douglas, P., Freni-Sterrantino, A., Leal Sanchez, M., Ashworth, D.C., Ghosh, R.E., Fecht, D., Font, A., Blangiardo, M., Gulliver, J., Toledano, M.B., Elliott, P., De Hoogh, K., Fuller, G.W., Hansell, A.L. Estimating Particulate Exposure from Modern Municipal Waste Incinerators in Great Britain, Environ. Sci. Technol.201751137511-7519, 2017

2.2.6 The conclusion of the study (on page 157) is clear:

“This large national study found no evidence for increased risk of a range of birth outcomes, including birth weight, preterm delivery and infant mortality, in relation to either MWI emissions or living near an MWI operating to the current EU waste incinerator regulations in Great Britain. The study should be generalisable to other MWIs operating to similar regulations and with similar waste streams.”

2.2.7 The most recent paper was published in April 2019 and is known as Freni-Sterrantino et al (2019)⁴ (provided in **Appendix B**). The objective of this paper was to determine whether it was possible to observe a change in infant mortality rates before and after an ERF is opened. The authors considered eight ERFs which opened over the time period considered (1996-2012), which included Lakeside and Allington close to London. For each facility, the authors identified all neighbourhoods within 10 km, working at the level of Middle Layer Super Output Area (MSOA), which is an area with a population of about 7,500 people. This gave a base area. The authors then identified comparator MSOAs within the same region which had similar characteristics to those within the 10 km radius in terms of deprivation, ethnicity, population density and nitrogen dioxide emissions, in order to give a comparator area.

2.2.8 The authors then compared infant mortality rates in the ERF area and the comparator area for five years before the opening of the ERF and five years afterwards. The data showed that infant mortality rates after the ERFs opened were lower than before the plants opened, both in the ERF areas and the comparator areas. The difference was actually slightly greater in the ERF areas (i.e. infant mortality improved faster in the areas within 10 km of ERFs), but the difference was not statistically significant. The authors considered a smaller buffer distance of 4 km and found the same result.

2.2.9 The authors carried out a similar study for the sex-ratio of births (i.e. the number of boys compared to the number of girls) and found no change between the periods before and after the opening of an ERF.

2.2.10 Hence, the authors concluded (on page 114) *“we did not find an association between the opening of a new MWI and changes in infant mortality trends or sex ratio at birth for 10 and 4 km buffers, using distance as proxy of exposure, after taking into account temporal trends in comparator areas and potential confounding factors.”*

2.2.11 The papers consider UK ERFs, operating under the same regulatory regime which would apply to REP and operating to current standards, Accordingly, this independent research and evidence is the most recent, most

⁴ Freni-Sterrantino, A; Ghosh, RE; Fecht, D; Toledano, MB; Elliott, P; Hansell, AL; Blangiardo, M. Bayesian spatial modelling for quasi-experimental designs: An interrupted time series study of the opening of Municipal Waste Incinerators in relation to infant mortality and sex ratio. *Environment International*. 128 (2019) 106-115 (Freni-Sterrantino et al, 2019)

comprehensive and most relevant research available. Given that neither paper found any evidence of an association of ERFs with the health outcomes considered, and that REP would actually operate to tighter standards, as it will use the reduced emissions limits from the Waste Incineration BREF (Best Available Techniques (BAT) reference documents), the Applicant is confident that the conclusions are directly relevant and support PHE's position statement.

3 Ultrafine Particulates

3.1 Monitoring of Particulates

- 3.1.1 It was suggested in the ISH on Environmental Matters on 5 June 2019 that ERFs do not monitor ultrafine particulates. This assertion is not correct.
- 3.1.2 The Operator will be required to monitor particulate emissions using the method set out in EN 13284-1. This method involves the extraction of flue gases through a filter to measure the dust concentration. The method requires that the filter efficiency must be at least 99.5 % on a test aerosol with a mean particle diameter of 0.3 μm , at the maximum flow rate anticipated. The filter efficiency for larger particles will be at least as high as this. This means that particulate monitoring data effectively captures everything above 0.3 μm and much of what is smaller. It is not expected that particles smaller than 0.3 μm will contribute significantly to the mass release rate / concentration of particulates because of their very small mass, even if present. This means that emissions monitoring data can be relied upon to measure the true mass emission rate of particulates.

3.2 Emissions of Ultrafine Particulates from Energy Recovery Facilities

- 3.2.1 Concerns have been raised by various parties about the releases of ultrafine particles. The impression is given that the abatement systems in place for particulate matter are ineffective for smaller particles (i.e. those smaller than 0.1 μm). This impression is incorrect.
- 3.2.2 Two separation principles apply within the bag filters.
- Absolute filtration – particles larger than the holes in the filter obviously cannot pass.
 - Adsorption – a layer of particles called “filter cake” builds up on the surface of the filter material which consists of reagents (lime and activated carbon) and reaction products. This layer is essential to the proper functioning of the flue gas treatment system. Within this layer, the final acid gas neutralisation and the absorption of heavy metals and complex organic compounds takes place.
- 3.2.3 It is the second reaction which accounts for the capture of the smaller particles which are adsorbed onto the surface of the particles in the filter cake. The smaller the particle, the greater the probability that it will be adsorbed onto another particle.
- 3.2.4 This mechanism can be demonstrated with reference to two published documents.

3.2.5 The International Energy Agency commissioned a report on fine particle emissions known as Wilen et al (provided in **Appendix C**)⁵. This report covered fine particle emissions from five combustion plants, being: (the first two are in Sweden and the remainder in Finland)

- Norrköping, a circulating fluidised bed (CFB) plant with a bag filter for particulate abatement, burning waste.
- Hässelholm, a grate-based plant with a bag filter, burning waste.
- Kautta, a CFB plant with an electrostatic precipitator (ESP) and a bag filter, burning waste, biomass and coke.
- Siilinjärvi, a grate based plant with an (ESP), burning biomass.
- Karhula, a CFB pilot plant with a bag filter, burning various fuels as a test facility.

3.2.6 It can be seen that the two ERFs (Norrköping and Hässelholm) are both equipped with a bag filter, which is the same abatement technology proposed for REP. The biomass combustion and co-incineration plants have ESPs and, in one case, a bag filter.

3.2.7 Section 6.3 of the paper presents the particulate emissions from the first three plants above and a number of pilot plant runs. Table 6 shows that particulate levels before the abatement system and Table 7 shows the levels after the abatement system. Both tables show concentrations of all particulates, PM2.5s and PM1s.

3.2.8 Table 7 in the paper shows that bag filter systems can achieve low concentrations of particulates. The total concentrations at operational plants are between 0.2 and 0.7 mg/m³, which is well below the emission limit in the Waste Incineration Directive of 10 mg/m³. Concentrations at the pilot plant were even lower. This data should provide clear reassurance that the emission levels assumed in the Applicant's air quality assessment of 5 mg/m³ for both PM10s and PM2.5s are conservative.

3.2.9 The data in Tables 6 and 7 in the paper can be combined to assess the effectiveness of bag filters. We have divided the top of the range of emitted particulate concentrations (from Table 7) by the bottom of the range of unabated particulate concentrations (from Table 6) for each of the plants. This showed that:

- the abatement of total particulates was between 99.984% and 99.997%;
- the abatement of PM2.5 was between 99.971% and 99.990%; and

⁵ Wilen, C., Moilenan, A., Hokkinen, J., Jokiniemi, J. "Fine Particle Emissions of Waste Incineration", March 2007

- the abatement of PM1 was between 99.864% and 99.991%.

3.2.10 This shows that bag filters are very effective at removing particulate matter, including PM2.5s and PM1s.

3.2.11 Secondly, a paper by Buonanno et al (2012)⁶ (provided in **Appendix D**) has considered the emissions of ultrafine particles from ERFs. The authors measured the ultrafine particle (UFPs) number distributions and total concentrations before and after the fabric filters at two ERFs. The conclusions of the paper (on page 110) are as follows:

“The core finding of the work is the modest amount of UFPs emitted by the analyzed incinerators, since particle number concentrations at the stack are always lower than 1.0×10^4 particles cm^{-3} , in particular, average particle number concentrations at the stack range from 0.4 to 6.0×10^3 particles cm^{-3} .

On the contrary, average particle number concentrations before the fabric filter were measured to be equal to 1.4 and 2.4×10^7 particles cm^{-3} at plants 1 and 2 respectively, leading to average removal efficiency of the fabric filters higher than 99.99%. In addition, measurements of the particle number distributions before and after the fabric filters show that the removal efficiency is quite constant all over the measurement range.”

3.2.12 Expanding on the final sentence, the abatement efficiency for particles between 5 nm and 40 nm was estimated to be 99.88%.

3.2.13 These papers demonstrate that bag filters are very effective at abating fine particulates, contrary to the concerns raised.

3.3 Particulate Emissions overall

3.3.1 Some of the written representations have referred to a report produced by UKWIN entitled “Waste Incineration and Particulate Pollution.” The Applicant notes that the Environment Agency has produced a response to this report, attached as **Appendix E**. This report notes that “*emissions from EfW plants make up just 0.03% / 0.05% of total UK PM10/ PM2.5 emissions. This is compared to 5.35% / 4.96% from traffic and 22.4% / 34.3% from domestic wood burning.*”

3.3.2 The report also confirms that EfW plants monitor total particulate matter and that this “*includes particles of all sizes including PM10, PM2.5, PM1 and ultrafine particles.*”

⁶ Buonanno, G, Scungio, M., Stabile, L, Tirlor, W. Ultrafine particle emission from incinerators: The role of the fabric filter, Journal of the Air and Water Management Association 62(1): 103:111, 2012

3.3.3 The Environment Agency also reiterates the PHE statement referred to in **Section 2** of this note.

4 Conclusions

- 4.1.1 PHE considers that “*while it is not possible to rule out adverse health effects from modern, well regulated municipal waste incinerators with complete certainty, any potential damage to the health of those living close-by is likely to be very small, if detectable*”.
- 4.1.2 Research commissioned by PHE and published in 2018 and 2019 shows that there is **no evidence** that living close to an ERF is associated with increased infant mortality or other infant health risks.
- 4.1.3 In addition, research has shown that abatement systems that are in place for particulate matter in ERFs are very effective at avoiding emissions of ultrafine particles.

Appendix A Ghosh et al (2018) Fetal growth, stillbirth, infant mortality and other birth outcomes near UK municipal waste incinerators; retrospective population based cohort and case-control study



Fetal growth, stillbirth, infant mortality and other birth outcomes near UK municipal waste incinerators; retrospective population based cohort and case-control study



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ABSTRACT

Background: Some studies have reported associations between municipal waste incinerator (MWI) exposures and adverse birth outcomes but there are few studies of modern MWIs operating to current European Union (EU) Industrial Emissions Directive standards.

Methods: Associations between modelled ground-level particulate matter $\leq 10 \mu\text{m}$ in diameter (PM_{10}) from MWI emissions (as a proxy for MWI emissions) within 10 km of each MWI, and selected birth and infant mortality outcomes were examined for all 22 MWIs operating in Great Britain 2003–10. We also investigated associations with proximity of residence to a MWI. Outcomes used were term birth weight, small for gestational age (SGA) at term, stillbirth, neonatal, post-neonatal and infant mortality, multiple births, sex ratio and preterm delivery sourced from national registration data from the Office for National Statistics. Analyses were adjusted for relevant confounders including year of birth, sex, season of birth, maternal age, deprivation, ethnicity and area characteristics and random effect terms were included in the models to allow for differences in baseline rates between areas and in incinerator feedstock.

Results: Analyses included 1,025,064 births and 18,694 infant deaths. There was no excess risk in relation to any of the outcomes investigated during pregnancy or early life of either mean modelled MWI PM_{10} or proximity to an MWI.

Conclusions: We found no evidence that exposure to PM_{10} from, or living near to, an MWI operating to current

Abbreviations: MWI, municipal waste incinerator; EU, European Union; PM_{10} , particulate matter $\leq 10 \mu\text{m}$ in diameter; SGA, small for gestational age; SO_2 , sulphur dioxide; NO_x , nitrogen oxides; HCl, hydrogen chloride; CO, carbon monoxide; VOC, volatile organic compound; POPs, persistent organic pollutants; PCDD/Fs, polychlorinated dibenzo-*p*-dioxins/furans; PCBs, polychlorinated biphenyls; PAHs, polycyclic aromatic hydrocarbons; EU-WID, European Union Waste Incineration Directive; COA, Census Output Area; MSA, Middle Layer Super Output Area; GAM, generalised additive model; OR, odds ratio; ONS, Office for National Statistics; EA, Environment Agency; SEPA, Scottish Environment Protection Agency; NRW, Natural Resources Wales; NHS, National Health Service; NHS NN4B, Numbers for Babies; NWS, NHS Wales' Informatics Service; HSW, Health Solutions Wales; NCCHD, National Child Community Health Dataset; ISD, Information Services Division Scotland

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EU standards was associated with harm for any of the outcomes investigated. Results should be generalisable to other MWIs operating to similar standards.

1. Introduction

Incineration of waste by Municipal Waste Incinerators (MWIs) has been increasing in the UK; since 2000 the tonnage of waste incinerated has more than tripled with approximately 35% of all local authority waste in England now being incinerated (Department for Environment, 2016). MWIs burn waste collected by local authorities that is not classified as hazardous or toxic and is generated mainly by households and commercial establishments (Committee on Health Effects of Waste Incineration, 2000). Air-borne emissions from MWIs depend on the composition of the feedstock incinerated but potentially include particulate matter, sulphur dioxide (SO₂), nitrogen oxides (NO_x), hydrogen

chloride (HCl), carbon monoxide (CO), volatile organic compounds (VOCs), persistent organic pollutants (POPs) such as polychlorinated dibenzo-*p*-dioxins/furans (PCDD/Fs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and heavy metals (Douglas et al., 2017).

MWI emission limit values are legislated by the European Union Waste Incineration Directive (EU-WID) (2000/76/EC) (Council E, 2000), which came into operation for new and existing MWIs on 28 December 2002 and 2005, respectively, and was subsequently incorporated within the current Industrial Emissions Directive (IED) (2010/75/EU) (European Union, 2010). Hazardous and medical wastes are handled by other types of incinerators and are not included in this

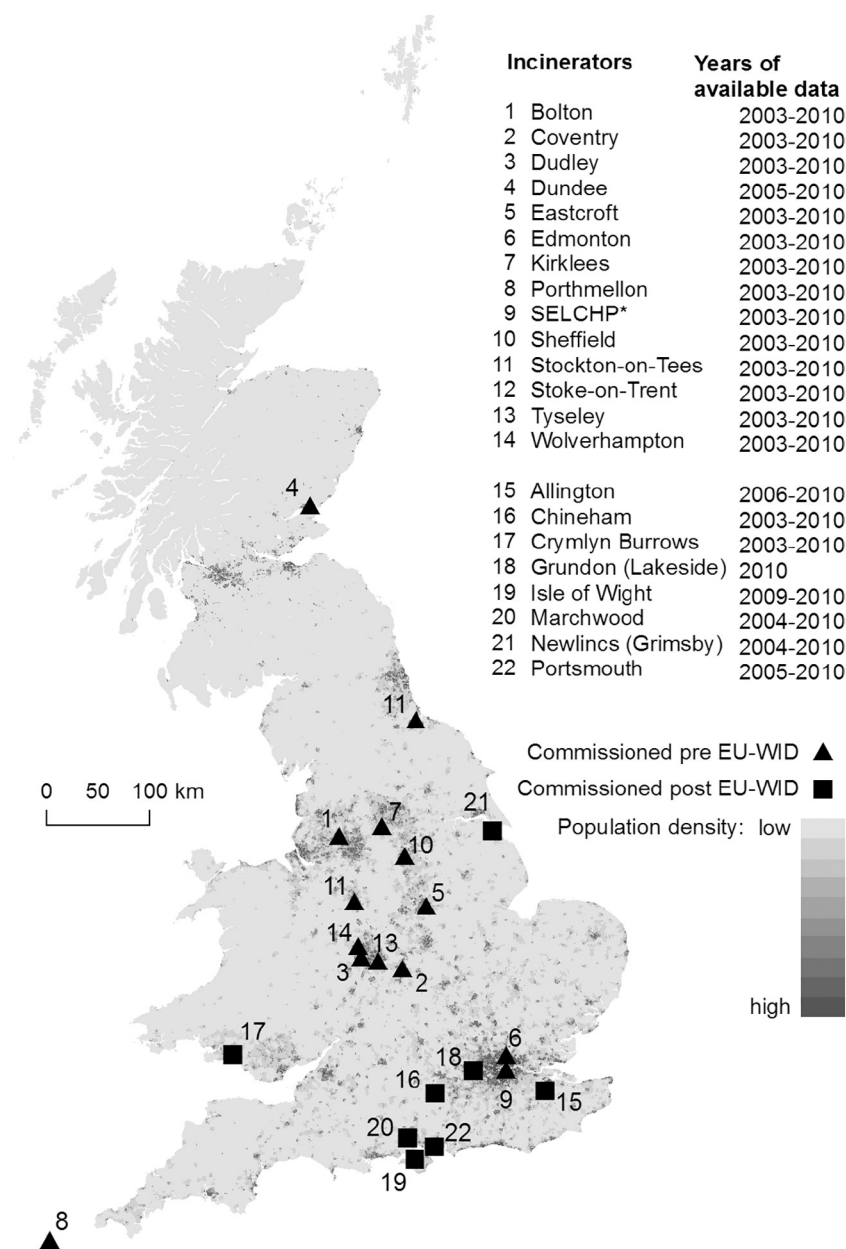


Fig. 1. Location of all municipal waste incinerators (MWIs) operating to the European Union Waste Incineration Directive (EU-WID) in Great Britain 2003–2010. The table denotes the years of data available.

*SELCHP is abbreviated for South East London Combined Heat and Power

study.

Although some studies have reported associations between MWI exposures and increased risks of e.g. preterm delivery (Candela et al., 2013), miscarriages (Candela et al., 2015) and congenital anomalies (Dummer et al., 2003; Cordier et al., 2010), reviews on health effects from MWIs (Health Protection Agency, 2009; Crowley et al., 2003; Maynard et al., 2010; Ashworth et al., 2014) have been inconclusive, and cite the need for improved studies with better exposure assessment. More generally, maternal exposure to ambient PM₁₀, mostly from road traffic sources, during pregnancy has been linked to fetal growth restriction (Dadvand et al., 2013; Pearce et al., 2012). The aim of this study was to investigate at the national scale possible health effects associated with (i) MWI emissions of particulate matter ≤ 10 μm in diameter (PM₁₀) as a proxy for MWI emissions more generally, and (ii) living near a MWI, in relation to fetal growth, stillbirth, infant mortality and other birth outcomes.

2. Methods

2.1. Municipal waste incinerators (MWIs) included

All MWIs in operation between 2003 and 2010 across Great Britain were eligible for inclusion in the study. One MWI in the Isle of Man (Richmond Hill) was excluded due to a lack of health and emissions data, while three other incinerators were excluded as they were not solely MWIs (Fawley, Hampshire; Ellesmere Port, Cheshire; Peak Load Boiler, Shetland) (Douglas et al., 2017). This left 22 MWI for inclusion in the study with the study area defined as a 10 km radius around each MWI. We modelled concentrations of PM₁₀ arising from MWI emissions (Douglas et al., 2017; Ashworth et al., 2013) as a proxy for MWI

emissions more generally (Douglas et al., 2017). Fig. 1 shows the location, name, and years of data availability for each MWI in the study.

2.2. Outcomes

Birth and mortality outcomes in the study area were obtained from routine administrative databases, 2003–10. Outcomes for investigation were selected by consideration of the evidence base and routine data availability e.g. there is no national data collection for spontaneous abortion so we are unable to study this. Congenital anomalies are considered elsewhere. Gestational age information, required for the fetal growth, and preterm birth outcomes, was only available from 2006 as was individual level ethnicity information.

2.2.1. Birth outcomes (2003–2010)

- Multiple births: ≥ 2 births from a single pregnancy.
- Sex ratio (all births): ratio of female births to male births.

2.2.2. Fetal growth and preterm birth (2006–2010)

- Preterm delivery (< 37 weeks gestation) among live singleton births.
- Birth weight among live, singleton term births ≥ 37 weeks to ≤ 44 weeks gestation.
- Small for gestational age (SGA) among live, singleton births, defined as birth weight below the sex and ethnicity specific 10th centile for gestational age for births ≥ 37 weeks gestation. Smoothed birth weight for gestational week centile curves were calculated by sex and ethnicity (Asian, Black, White, Other), using a method for

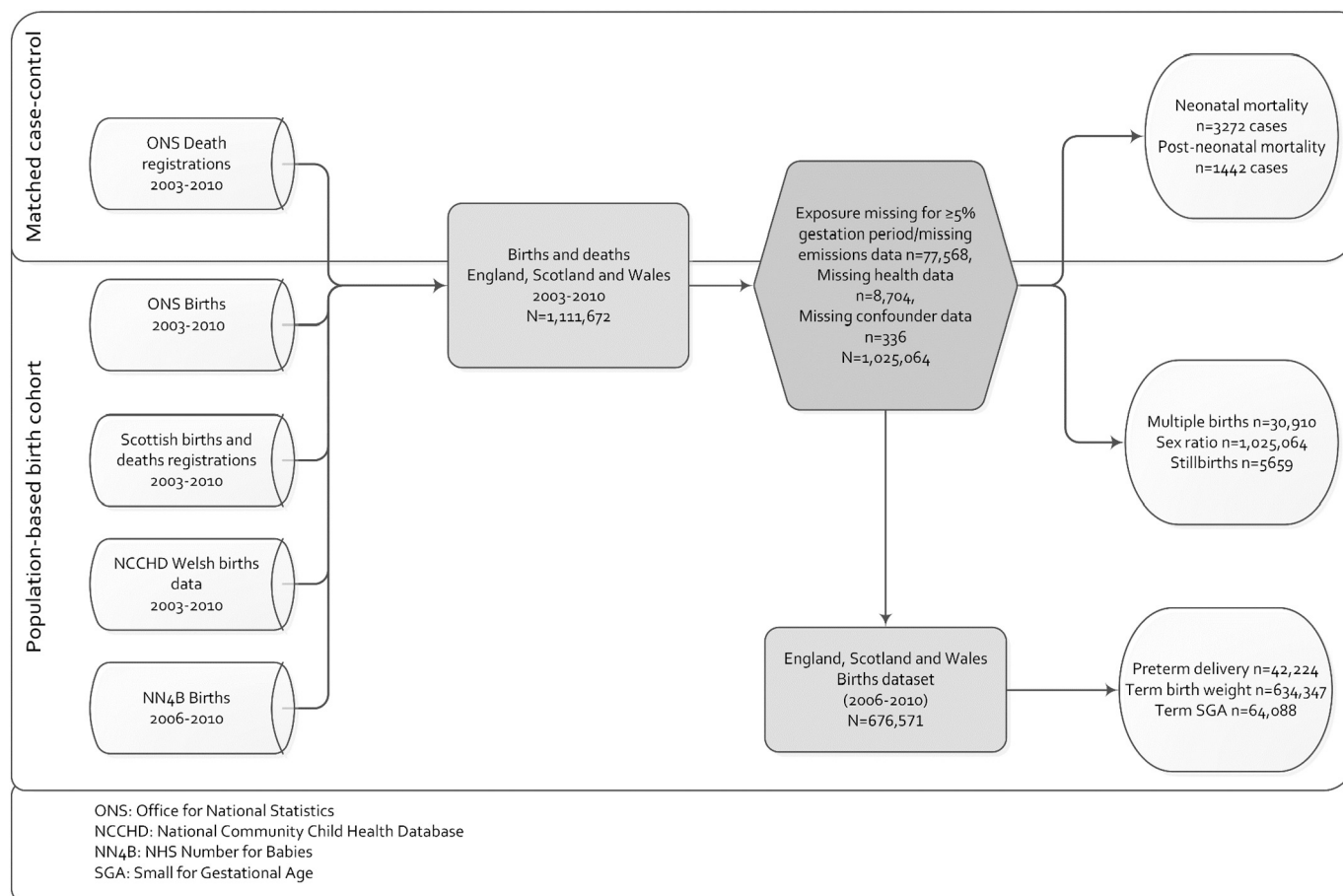


Fig. 2. Datasets and exclusions for each analysis and outcome in the study.

drawing centile curves, consisting of a generalised additive model for location, scale and shape as described by Cole and others (Rigby and Stasinopoulos, 2005; Cole and Green, 1992).

2.2.3. Mortality outcomes (2003–2010)

- Stillbirths: Births at ≥ 24 completed weeks gestation showing no signs of life
- Neonatal mortality: deaths among singleton births occurring between birth and ≤ 28 days after birth.
- Post-neonatal mortality: deaths among singleton births occurring between 29 and ≤ 365 days after birth.

2.3. Data

Outcome data were obtained from the Office for National Statistics (ONS) including the National Mortality register for England and Wales, the Births and Stillbirths register for England and Wales and the NHS Number for Babies dataset (NN4B); the National Child Community Health Dataset (NCCHD) from the NHS Wales Informatics Service (NWIS)/Health Solutions Wales (HSW); and from the Information Services Division (ISD) in Scotland. How these datasets were combined for each outcome and analysis is shown in Fig. 2.

Cases were taken from the combined outcome data on all births or deaths in Great Britain between 2003 and 2010 and were included if the postcode centroid (representing on average 12 households) of the mother's usual residence, provided at the time of birth registration, was within 10 km of an open MWI and the estimated date of conception (based on gestation weeks) and outcome were also within the study period (2003–2010).

2.4. MWI PM_{10} exposure assessment

ADMS-Urban (CERC, 2017), extensively validated Gaussian-based atmospheric dispersion model software, was used to estimate daily concentrations of ground-level PM_{10} resulting from MWI emissions as a proxy for MWI emissions more generally (Candela et al., 2013; Cordioli et al., 2013). Methods are as previously described (Ashworth et al., 2013). Input data were obtained from the relevant regulatory agencies in England (Environment Agency; EA), Wales (Natural Resources Wales, NRW) and Scotland (Scottish Environment Protection Agency, SEPA). They comprised within-flue particulate matter/total dust emissions monitored continuously as part of the EU-WID regulations, reported as daily mean concentrations ($\mu\text{g}/\text{m}^3$), and MWI operating characteristics. PM_{10} was estimated rather than total suspended particles as size fraction studies have found that particulate emissions from incinerators have diameters $< 10 \mu\text{m}$ (Buonanno et al., 2009). Results of the modelling for the 22 MWIs are published (Douglas et al., 2017), although since then new flue characteristics data have become available for Sheffield MWI, and thus the Sheffield MWI data have been remodeled as described in Supplement A. A daily estimate of PM_{10} was obtained for each postcode centroid within the study area.

For the birth outcomes and stillbirths the mean concentrations of PM_{10} during pregnancy were estimated (see Supplement B for further details). For neonatal and post-neonatal mortality outcomes, post-birth exposure to MWI emissions was also estimated. For the post-birth mean PM_{10} exposure to be similar for both cases and controls, we used a matched case-control design for these analyses, with controls (four per case) frequency matched by birth date, sex and the postcode of the mother's residence being within 10 km of the same MWI as the cases. Exposure was calculated from the date of birth (considered an exposed day) to date of death of the case.

2.5. Confounders

Potential confounders were selected a priori for each analysis – see

Supplement B - Supplemental Tables B.1–B.3 for details. Individual-level confounders included were sex, maternal age (continuous), season of birth (winter as reference), and, from 2006, gestational age (both linear and quadratic terms) and ethnicity of the baby as reported by the mother (White, Asian, Black or Other). Area-level confounders, derived from the 2011 UK Census, were Carstairs index categorised into fifths (Carstairs and Morris, 1990) as a measure of socio-economic status, calculated for census output areas (COAs); population density (population per square km for each COA); and ethnicity defined as the percentage non-white ethnicity among the female population of reproductive age (15–49 years) in each Middle Layer Super Output Area (MSOA, comprising contiguous COAs with an average population size of 7500). To adjust for other potential PM_{10} sources, models included local road density data from 2014, defined as the continuous total road length within 250 m of each residential postcode; road length within 10 km of each MWI; and industrial emission sources defined as the number of industries in 2003–10 within 10 km of each MWI, based on data included in the Environmental Permitting Regulations – Industrial sites 2010 (Supplemental Table B.3).

Sensitivity analyses additionally adjusted for (i) COA-level tobacco expenditure as a smoking proxy (England and Wales only because of data availability); and (ii) birth registration type (within marriage (reference category), joint-same address, joint-different address, sole registered) as a proxy for individual level socio-economic status (England only) (Graham et al., 2007).

2.6. Data and statistical analysis

There were a total of 1,111,672 births from 2003 to 2010; 77,568 (7%) were excluded due to missing emissions data or where $\geq 5\%$ of the exposure period had invalid values or the dispersion model was unable to estimate a concentration (Douglas et al., 2017; Ashworth et al., 2013). A further 8704 (0.78%) records were excluded due to missing health data (e.g. birth weight, gestational age, ethnicity) and 336 (0.03%) due to missing area-level confounder data (Fig. 2). This left a total of 1,025,064 births available for analysis from 2003 to 2010, including 30,910 (3.02%) multiple births, 5659 (0.55%) stillbirths, 3260 neonatal and 1442 post-neonatal deaths. For the period 2006 to 2010, there were 676,571 live singleton births with gestational age information. For 2006 to 2010 there were 634,347 (93.76%) term births and 42,224 (6.24%) pre-term births, of the term births 64,088 (10.01%) were SGA (Supplemental Tables B.4 and B.5). For the population-based cohort analysis (2003–2010), overall 51% of births were males, 25% of the areas had over 50% non-white ethnicity and 50% of all births were in the most deprived areas by Carstairs quintile. For the matched case-control analysis a slightly higher proportion were male (53%), 28% of the areas had over 50% non-white ethnicity and 54% of all births were in the most deprived Carstairs quintile.

As the mean PM_{10} exposures distributions were highly skewed, they were log transformed for analysis. Proximity to the nearest MWI was calculated as a continuous measure of linear distance (km) based on postcode centroid of maternal residence at birth. For the population based cohort analyses we also conducted sensitivity analyses with exposures analysed as quintiles.

Associations between mean ground-level PM_{10} concentrations and each outcome, adjusted for relevant potential confounders, were examined using multiple logistic regression (multiple births, sex ratio, preterm deliveries, term SGA and stillbirths) and multiple linear regression (birth weight). These regression models included a random intercept for each MWI to allow for differences in baseline rates between areas, and a random slope to account for unobserved heterogeneity. Conditional logistic regression was used for neonatal and post-neonatal mortality analyses. Since PM concentrations may vary seasonally (due to seasonal patterns in weather and wind direction), we checked for seasonal patterns using a generalised additive model (GAM), which did not show any evidence of seasonal effects (not

shown). All analyses were conducted in Stata version 13.

3. Results

3.1. Birth and fetal growth outcomes

The mean modelled PM₁₀ concentrations during pregnancy for all live births were 2.51 × 10⁻³ µg/m [IQR 0.47–2.86] (Supplemental Table B.6). For the birth, fetal growth and preterm birth outcomes we found no associations in the adjusted models between term birth weight (co-efficient 0.12 g [95%CI - 1.51, 1.75]), term SGA (Odds Ratio 0.99 [0.98, 1.00]), sex ratio (OR 1.00 [1.00–1.00]), multiple births (OR 0.99 [0.99, 1.00]) or preterm delivery (OR 0.99 [0.97, 1.01]) per doubling of PM₁₀ from MWIs during pregnancy (Table 1).

There were no associations between the same outcomes and proximity to an incinerator, in adjusted models; the coefficient for term birth weight was -0.56 g/km [95% CI -1.80, 0.68].

3.2. Mortality outcomes

For the mortality outcomes we found no associations in the adjusted models, for stillbirths (OR 0.98 [0.97, 1.00]), neonatal (OR 1.00 [0.96, 1.02]) and post-neonatal mortality (OR 1.02 [0.96, 1.07]) per doubling of PM₁₀ from MWIs (Table 2). Findings were similar when analyses were restricted to 2006–10, years when gestational age information was available (Supplement Table B.11). There was also no association between post-neonatal mortality and exposure after birth, in the adjusted models. Proximity to nearest MWI was not associated with mortality.

Sensitivity analyses by quintiles of PM₁₀ concentrations (Supplemental Tables B.12 and B.13) or with additional adjustment for tobacco sales and birth registration (Supplement Tables B.14 and B.15) did not alter the findings.

4. Discussion

This is, to our knowledge, the largest study to date to examine potential impacts of modern MWIs operating to current EU regulations on a range of fetal outcomes, preterm delivery and infant mortality.

Table 1

Risk of mortality outcomes associated with a doubling in ground-level PM₁₀ concentrations and proximity from residential address to an incinerator^a.

	n cases/N total	Risk per doubling of PM ₁₀ ^b		Proximity to MWI (continuous) per km	
		OR	95%CI	OR	95%CI
Multiple births					
Unadjusted	30,910/1,025,064	1.00	1.00, 1.00	0.98	0.97, 0.98
Adjusted		0.99	0.99, 1.00	1.00	0.99, 1.00
Sex ratio					
Unadjusted	525,272(males)/1,025,064	1.00	1.00, 1.00	1.00	1.00, 1.00
Adjusted		1.00	1.00, 1.00	1.00	1.00, 1.00
Preterm delivery					
Unadjusted	42,224/676,571	1.00	0.99, 1.00	1.01	1.00, 1.01
Adjusted		0.99	0.97, 1.01	1.00	0.99, 1.00
Term SGA					
Unadjusted	64,088/634,347	1.00	0.99, 1.00	1.01	1.01, 1.02
Adjusted		0.99	0.98, 1.00	1.00	0.99, 1.01
Term birth weight					
Mean difference in term birth weight (g)		Coefficient	95%CI	Coefficient	95%CI
Unadjusted	634,347	-5.43	-5.83, -5.04	-5.52	-6.01, -5.04
Adjusted		0.12	-1.51, 1.75	-0.56	-1.80, 0.68

^a All analyses adjusted for year of birth, maternal age, area-level deprivation, population density, road density, incinerator road density, other potential sources of emissions, and included a random effect for incinerator area and random slope for the exposure. Analyses additionally adjusted for one or more of sex, season of birth, ethnicity (area or individual level), gestational age depending on outcome as detailed in Supplement Table B.1.

^b The odds ratios from a log transformed exposure do not represent a risk per increase in PM₁₀ unit (µg/m³) on a linear scale so to aid interpretation we present the risk associated with a doubling of the ground-level PM₁₀ concentrations, for example an odds ratio of 1.05 would indicate a 5% higher risk of an outcome each time the PM₁₀ is doubled.

Table 2

Risk of mortality outcomes associated with a doubling in ground-level PM₁₀ concentrations and proximity from residential address to an incinerator^a.

	n cases/N total	Risk per doubling of PM ₁₀ ^b		Proximity to MWI (continuous) per km	
		OR	95%CI	OR	95%CI
Stillbirths					
2003–2010					
Unadjusted	5659/	1.01	1.00, 1.02	1.02	1.01, 1.03
Adjusted	1,025,064	0.99	0.97, 1.00	1.00	0.99, 1.02
Neonatal mortality (pregnancy exposure)^c					
2003–2010					
Unadjusted	3260/12,944	1.03	1.00, 1.06	1.03	1.02, 1.05
Adjusted		0.99	0.96, 1.02	1.01	1.00, 1.03
Post-neonatal mortality (pregnancy exposure)					
2003–2010					
Unadjusted	1442/5750	1.10	1.04, 1.16	1.03	1.01–1.05
Adjusted		1.02	0.96, 1.07	0.99	0.97, 1.02
Post-neonatal mortality (birth to death of case exposure)					
2003–2010					
Unadjusted	1442/5750	1.04	1.01, 1.08	-	-
Adjusted		1.01	0.98, 1.04	-	-

^a Stillbirth analyses are adjusted for year of birth, sex, season of birth, maternal age, area-level deprivation, area-level ethnicity, population density, road density, incinerator road density, other potential sources of emissions, random effect for incinerator area and random slope for the exposure. Infant mortality outcome analyses are case-control analyses adjusted for sex, maternal age, area-level deprivation, area-level ethnicity, population density, road density.

^b The odds ratios from a log transformed exposure do not represent a risk per increase in PM₁₀ unit (µg/m³) on a linear scale so to aid interpretation we present the risk associated with a doubling of the ground-level PM₁₀ concentrations, for example an odds ratio of 1.05 would indicate a 5% higher risk of an outcome each time the PM₁₀ is doubled.

^c Pregnancy exposure only is presented as babies are likely very sick and considered unlikely to leave hospital before death, therefore only the pregnancy window is relevant as an exposure.

Estimated ground-level PM₁₀ concentrations from MWI emissions, as a proxy for MWI emissions more generally (Douglas et al., 2017), were not associated with increased risk of adverse birth, fetal growth or mortality outcomes. Living near an incinerator was also not associated with increased risks to infant health.

We assumed that the predominant pathway of exposure to incinerator emissions would be through air. We used estimated mean ground-level concentrations of PM₁₀ as our main exposure measure. Ambient PM₁₀ concentrations have previously been reported to be associated with adverse birth outcomes (Dadvand et al., 2013; Lamichhane et al., 2015), but ambient concentrations of PM₁₀ are approximately 3–5 orders of magnitude greater than our estimates for the contribution from MWIs (Douglas et al., 2017), so the results are not directly comparable. We also examined distance from incinerator as has been done in several other studies (Obi-Osius et al., 2004; Tango et al., 2004; Lloyd et al., 1979; Williams et al., 1992). Although distance is a relatively crude (but readily obtained) measure, it may capture exposures not reflected in the emissions modelling (e.g. transport of waste to the MWI) as well as non-airborne exposure pathways.

Some studies have, like ours, used modelled PM₁₀ (Candela et al., 2013; Candela et al., 2015; Santoro et al., 2016) or modelled dioxin concentrations (Vinceti et al., 2008; Lin et al., 2006). The most comparable recent studies to ours were a multi-site (Candela et al., 2013; Candela et al., 2015) and a single site MWI study (Santoro et al., 2016), both from Italy, which used modelled PM₁₀ in relation to MWIs operating to the EU-WID. The Candela et al. (2013, 2015) study, where the outcomes chosen and the exposure modelling methods (ADMS-Urban) were comparable to this study, was generally consistent with our findings of a lack of any associations with sex ratio, multiple births, and SGA. Where this study was not consistent with ours was for preterm births – Candela et al. (2013), covering eight Italian incinerators, found an increased odds ratio of 1.30 (95% confidence interval [1.08–1.57]) comparing highest vs. lowest fifth of PM₁₀ concentrations. They also found an increased risk of spontaneous abortions (Candela et al., 2015) but that measure of fetal mortality was not directly comparable with our mortality outcomes. Candela et al. used a smaller buffer around each incinerator than our study, 4 km vs 10 km, which may have led to fewer outcomes with a lower estimated exposure being included in their study as compared to ours. However, overall they estimated a mean per-person exposure level of 0.57 ng/m³ which was lower but still of the same order of magnitude as our mean finding per pregnancy for the birth outcomes of 2.51 ng/m³. The Santoro et al. (2016) study also found an increased risk of preterm births however this was a study of only one incinerator in a highly industrialized area and included data from the incinerator from before the introduction of the EU-WID regulations. The lack of an association in our study of incinerator emissions or proximity with sex ratio (Tango et al., 2004; Lloyd et al., 1979; Lin et al., 2006; Rydhstroem, 1998), birth weight (Candela et al., 2013; Tango et al., 2004; Santoro et al., 2016; Lin et al., 2006), SGA (Candela et al., 2013; Santoro et al., 2016), stillbirths, neonatal and post-neonatal mortality (Dummer et al., 2003; Tango et al., 2004; Vinceti et al., 2008) is consistent with other studies.

Evidence on multiple births has been inconsistent: while two studies, Lloyd et al. (1979) in Scotland and Obi-Osius et al. (2004) in Germany, found increased risks of twinning, the Rydhstroem (1998) study in Sweden found decreased risks of multiple births. However these studies were of pre-EU-WID incinerators and used either distance from the incinerator as a proxy for exposure (Obi-Osius et al., 2004; Lloyd et al., 1979) or compared twinning rates for the periods before/after an incinerator opening (Rydhstroem, 1998), which may have been confounded by temporal trends in twinning rates. The more recent Candela et al. (2013) with comparable exposure modelling to ours also found no association between MWI emissions and multiple births.

4.1. Strengths and limitations

This is a large study, using comprehensive national birth and death registration data and included all MWIs nationally, avoiding selection bias. Over one million births were included in the analyses providing sufficient statistical power to be able to detect small associations. We used two methods to assess potential exposure to incinerator emissions: modelled PM₁₀ concentrations and distance from incinerator. We made the assumption that modelled PM₁₀ dispersion is a reasonable proxy of spatial exposure patterns to other components of MWI emissions (Douglas et al., 2017). This assumption was investigated in the following ways.

- For NO₂ and SO₂: We conducted dispersion modelling for NO₂ and SO₂ using in-stack measurements for a representative incinerator (details in Supplement C). Modelled NO₂ and SO₂ correlated well with modelled concentrations of PM₁₀ with Spearman's correlation values of $r = 0.88$ – 0.99 (Supplemental Table C.3).
- For heavy metals: We previously examined ratios in air pollutant metal concentrations measured at ambient monitoring stations near six incinerators (Font et al., 2015). We found no evidence that MWI emissions contributed to ambient air pollution metal concentrations near four MWIs and minimal contributions from the remaining two. (Dispersion modelling could not be used for heavy metals - or for PCDD/Fs, PAHs or PCBs emissions - as these are monitored for regulatory purposes using intermittent spot measurements, unlike particulates for which continuous measurements are taken.)

We only had information on residential address at time of birth. We did not have information on change in residence during the pregnancy, estimated at around 16% in the UK (Tunstall et al., 2010). Maternal mobility is more likely in younger mothers living in more deprived areas (Hodgson et al., 2009) which may have introduced some differential exposure misclassification. Gestational age at birth, a major risk factor in mortality outcomes, was not adjusted for in the mortality analysis as gestational age information was only available for 5 of the 8 years of the analysis. The assumption that all births prior to 2006, with no gestational age information, were term births is a potential source of exposure misclassification, particularly for outcomes likely not to go to full term e.g. stillbirths. However, sensitivity analyses for stillbirths and infant mortality restricted to 2006–10, the years when gestational age was available, were consistent with those for the whole study period. There was no evidence that emissions levels changed systematically between the earlier study years and following the implementation of the EU-WID at the end of 2005 (Douglas et al., 2017), but we did not have information on changes in feedstock for each MWI over that time period.

We controlled for major confounders including age, ethnicity and deprivation. We did not have individual-level information on maternal smoking and deprivation, but adjustments at the small-area level for tobacco sales and at individual-level for birth registration type (a proxy for socio-economic status as it is related to qualifications and housing tenure (Graham et al., 2007)) did not materially change the results.

The majority of exclusions were due to gaps in the emissions data. This did result in a study population that included more areas with higher percentage of ethnic minorities and that were more deprived (Supplement Table B.18). To explore the impact of this, we relaxed the criteria for missing emissions data from 5% to 15%, and reran the analyses (Supplement Tables B.16 and B.17) but results were similar to those reported here.

MWIs are not randomly located across the country and are often built in heavily industrialized areas with other sources of pollution (Font et al., 2015). The model adjusted for a count of relevant industries and for road density, which has been shown to perform well as a representation of spatial variation in road traffic air pollution (Rose et al., 2009). To account for remaining spatial variability not explained by the

covariates, we included a random effect for incinerator MWI area in the statistical models. We also included a random slope term in the statistical models to allow for heterogeneity in the exposure-response relationship, which should allow for differences in composition of waste incinerated at each MWI for which information is not available. Despite these efforts, we cannot exclude the possibility of residual confounding that may have affected our estimates.

5. Conclusions

This large national study found no evidence for increased risk of a range of birth outcomes, including birth weight, preterm delivery and infant mortality, in relation to either MWI emissions or living near an MWI operating to the current EU waste incinerator regulations in Great Britain. The study should be generalisable to other MWIs operating to similar regulations and with similar waste streams.

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Data

Births and deaths data were from the Office for National Statistics (ONS) National Mortality, Births and Stillbirth registers for England and Wales and the National Health Service (NHS) Numbers for Babies (NN4B). Welsh births data were from the National Child Community Health Dataset (NCCHD) from the NHS Wales' Informatics Service (NWIS)/ Health Solutions Wales (HSW). Scottish births and deaths were from the Information Services Division (ISD) Scotland.

Incinerator emissions data came from the Environment Agency (EA), Scottish Environment Protection Agency (SEPA), and Natural Resources Wales (NRW).

Data on industrial sites came from the Environment Agency Environmental Permitting Regulations – Industrial sites (England), Natural Resources Wales - Environmental Permitting Regulations – Industrial sites and the Scottish Pollutant Release Inventory.

2011 Census aggregate data came from the Office for National Statistics (for England and Wales) and the National Records of Scotland (2016): UK Data Service (Edition: June 2016).

Road length data came from Meridian 2014 road lengths. Ordnance Survey data © Crown copyright and database right 2014.

CACI tobacco expenditure data is © Copyright 1996-2014 CACI Limited. We attest that we have obtained appropriate permissions and paid any required fees for use of copyright protected materials.

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Declaration of interests

None. Anna Hansell declares a Greenpeace membership but has not received any money from the organisation nor been involved in campaigns; nor other relationships or activities that could appear to have influenced the submitted work. Brandon Parkes declares a Friends of the Earth membership but has not been involved in campaigns; nor other relationships or activities that could appear to have influenced the submitted work. The authors have no other relationships or activities that could appear to have influenced the submitted work.

The views expressed are those of the author(s) and not necessarily those of the NIHR, the Department of Health or Public Health England.

Ethical approval

The study uses SAHSU data, supplied from the Office for National Statistics; data use and link between UK National Births and Stillbirth register data and NHS Numbers for Babies (NN4B) was covered by approval from the National Research Ethics Service 17/L0/0846 and by the Health Research Authority Confidentiality Advisory Group (HRA-CAG) for Section 251 support (HRA – 14/CAG/1039). Approval for Scottish data was covered by SAHSU existing ethics and from the NHS National Services Scotland Privacy Advisory Committee (PAC) reference - PAC 17/14.

Data sharing

Health data are available from the data providers on application with appropriate ethics and governance permissions, but we do not hold data provider, ethics, or governance permissions to share these datasets with third parties.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2018.10.060>.

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Appendix B Freni-Sterrantino et al (2019) spatial modelling for quasi-experimental designs: An interrupted time series study of the opening of Municipal Waste Incinerators in relation to infant mortality and sex ratio



Bayesian spatial modelling for quasi-experimental designs: An interrupted time series study of the opening of Municipal Waste Incinerators in relation to infant mortality and sex ratio

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ABSTRACT

Background: There is limited evidence on potential health risks from Municipal Waste Incinerators (MWIs), and previous studies on birth outcomes show inconsistent results. Here, we evaluate whether the opening of MWIs is associated with infant mortality and sex ratio in the surrounding areas, extending the Interrupted Time Series (ITS) methodological approach to account for spatial dependencies at the small area level.

Methods: We specified a Bayesian hierarchical model to investigate the annual risks of infant mortality and sex-ratio (female relative to male) within 10 km of eight MWIs in England and Wales, during the period 1996–2012. We included comparative areas matched one-to-one of similar size and area characteristics.

Results: During the study period, infant mortality rates decreased overall by 2.5% per year in England. The opening of an incinerator in the MWI area was associated with –8 deaths per 100,000 infants (95% CI –62, 40) and with a difference in sex ratio of –0.004 (95% CI –0.02, 0.01), comparing the period after opening with that before, corrected for before-after trends in the comparator areas.

Conclusion: Our method is suitable for the analysis of quasi-experimental time series studies in the presence of spatial structure and when there are global time trends in the outcome variable. Based on our approach, we do not find evidence of an association of MWI opening with changes in risks of infant mortality or sex ratio in comparison with control areas.

1. Introduction

In recent years, incineration of municipal waste in Europe has increased in response to legislation to divert waste from landfills, leading to the construction of new municipal waste incinerator (MWI) plants. MWI emissions are regulated by the European Union Waste Incineration Directive (EU-WID) (2000/76/EC), later incorporated within the current Industrial Emissions Directive (IED) (2010/75/EU). The EU-WID was implemented in Great Britain for new MWIs on December 28th, 2002.

Emissions from MWIs include particulates, SO₂, nitrogen oxides

(NO_x), heavy metals, polycyclic aromatic hydrocarbon (PAH) and polychlorinated biphenyls (PCBs), as well as polychlorinated dibenzo-p-dioxins and furans (PCDD/Fs) which are reported to have a range of biological effects including enzyme induction, immunotoxicity, developmental toxicity and with reproductive and endocrine effects (Birnbaum, 1994). Exposures to particulates as a proxy for MWI emissions are very low from modern well-regulated incinerators (Douglas et al., 2017).

Despite high public concern about potential effects of waste management processes on birth and infant mortality outcomes, there are relatively few epidemiological studies available, particularly of modern

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incinerators. Some previous studies have found associations with infant mortality (Tango et al., 2004), miscarriages (Candela et al., 2015), sex ratio (Williams et al., 1992), preterm delivery (Candela et al., 2013), and congenital anomalies (Dummer et al., 2003; Cordier et al., 2010), with congenital anomalies and pre-term birth being the leading causes of infant mortality. The established reproductive toxicity of dioxins and heavy metals, present in emissions from incinerators, provides plausible biological mechanisms for these observed associations, although doses received by individuals at ground level are likely to be very low (Ashworth et al., 2014). In addition, sex ratio may behave as a sentinel for the endocrine disruption due to chemicals in MWI emissions including dioxins and furans (Candela et al., 2013; Van Larebeke et al., 2008).

A systematic review (Ashworth et al., 2014) found that the evidence for adverse birth and neonatal outcomes in relation to waste incineration was limited and inconclusive. Problems identified were the small numbers of available studies and low statistical power in each study, potential exposure misclassification, and residual confounding. Additionally, the current evidence-base was found to be difficult to synthesise due to the heterogeneity between studies in their design, location, time period, number of incinerators and the birth outcomes explored. Thus, the authors recommended further epidemiological investigation, particularly multi-site studies to increase statistical power, with better control for confounding, more accurate exposure assessment and robust methodology (Ashworth et al., 2014).

To help address these recommendations, we consider the opening of multiple new MWIs across England as a natural experiment to study annual trends in birth outcomes, with a focus on infant mortality and sex ratio, using an interrupted time series (ITS) approach (Zhang et al., 2014).

In an ITS study, a time series is used to establish an underlying trend, which is 'interrupted' by an intervention at a known point in time. Such methods have been applied in: epidemiology (Gasparrini et al., 2009; James Lopez, 2017), econometrics (Shadish et al., 2002), drug use research (Jandoc et al., 2015), evaluation of health care quality (Penfold and Zhang, 2013) and to assess impact of changes in public health policies (Kontopantelis et al., 2015) e.g. the effect of a smoking ban on birth outcomes (Bakolis et al., 2016). The ITS approach has also become popular in the road safety literature, where authors have focused on deriving indices to describe the effect of safety interventions both in an empirical Bayesian and fully Bayesian framework (Park et al., 2010; El-Basyouny and Sayed, 2011).

In its original formulation, ITS evaluates only the changes in the time series of the outcome of interest before and after the intervention, failing to account for time trends independent of it; therefore inference may be biased if the outcome exhibits a time trend that might confound the intervention effect (Linden and Adams John, 2010). Li et al. (2013) proposed a two-stage Bayesian hierarchical model to evaluate the effect of an intervention to reduce crime, allowing for data sparsity and including control areas to account for global time trends. In the first stage, the control areas (matched to the exposed areas) were used to assess the pre-intervention trend and then the estimates were fed to a second stage, where the effect of the intervention was evaluated in the exposed areas. However, the two-stage formulation prevented the propagation of uncertainty from exposed to control areas; also, no structured local spatial dependency was included.

Here, we develop the approach used by Li et al. (2013) and propose a framework for applying the ITS approach to small area data, defining the opening of a new MWI as the intervention, and infant mortality and sex ratio in the surrounding areas as the outcome.

We consider important confounders including area-level measures of ethnicity and deprivation, both known to be associated with early life outcomes. For example, a systematic review of UK studies found that infant mortality is associated with greater area-deprivation and lower social class (Weightman et al., 2012). Infants of South Asian and Black ethnicity are reported to have higher infant mortality, partially

explained by the increased prevalence of congenital anomalies in these populations (Li et al., 2018). In addition, sex ratio (males over total) declines in association with socio-economic status, possibly because of higher stress levels with lower socio-economic status (Grech, 2017), and differences have been reported in newborn sex ratio according to paternal ethnicity (Grunebaum and Chervenak, 2016).

Our modelling framework extends the previous statistical work in the field in the following ways: (i) we include control areas matched on size and area characteristics to allow for global trends; (ii) we specify the spatial dependency between areas; (iii) we perform joint Bayesian inference of exposed and control areas which allows uncertainty to be propagated across the model; and (iv) we define an index to quantify the before-after difference in infant mortality risks comparing the MWI and control areas.

This work is part of a larger national investigation of 22 MWIs in relation to reproductive outcomes in Great Britain during 2003–10, where we estimated exposure to particulate matter from MWI emissions in Great Britain (Douglas et al., 2017) and further used both modelled emissions concentrations and distance from incinerators to examine associations with fetal outcomes (Ghosh et al., 2019).

2. Methods

2.1. Study design

We included all eight new MWIs coming into operation in England and Wales between 2003 and 2010 operating under the EU WID. Infant mortality and sex ratio data were available for the period 1996–2012, at Middle layer Super Output Area (MSOA) Census geography (average population 7500).

2.2. Incinerator (exposed) areas

We defined the exposed area for each incinerator as the set of MSOAs whose centroids lie within 10 km of a MWI (Fig. 1). We chose 10 km as the cut-off for consistency with screening criteria used for implementing the Habitats Regulations (1992/43/EEC). These regulations state that incineration plants that are within 10 km of a European Site require an assessment of their impact for short-range air emissions (Ashworth et al., 2013). For each MWI, we considered five years before and after the opening date (Table 1), except for Isle of Wight (3 years) and Grundon (Lakeside) (2 years) where opening dates were respectively 2009 and 2010, because as noted outcomes were available only to 2012.

2.3. Comparator areas

For each MWI we selected one comparator area (composed by MSOAs), as follows:

- (i) For each MWI, we identified the potential set of comparator MSOAs, as all MSOAs within the same Government Office Region as that of the MWI. For Grundon more than half of the MSOAs were in London (the rest in South East) therefore we used the London Region as reference (Table 1).
- (ii) For each MSOA in the reference region, we then constructed a 10 km circular buffer around each MSOA centroid. We excluded MSOAs within 10 km of an existing or new MWI or sharing a boundary with such an MSOA.
- (iii) For each of these potential comparator areas, we selected the same opening years as the MWI, and then constructed a vector that included:
 - annual number of live singleton births for the pre-opening period,
 - population density (Census 2011),
 - percentage of population of non-white ethnicity (Census 2011)

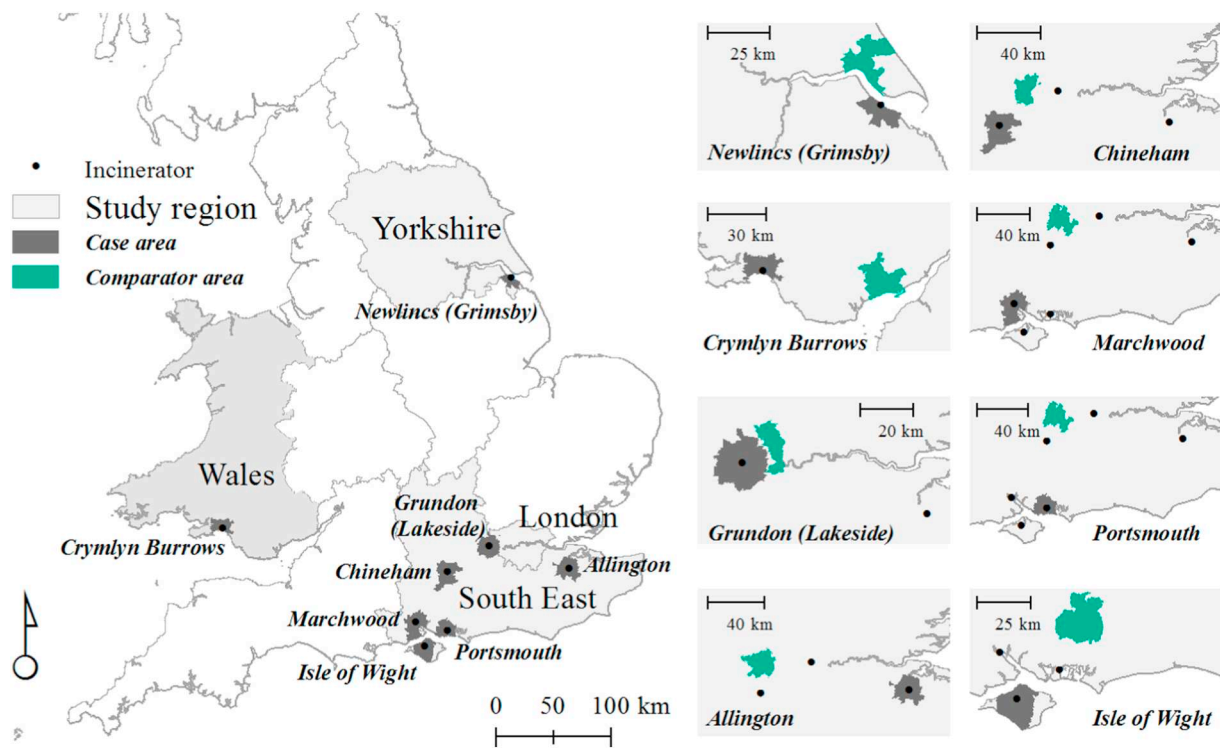


Fig. 1. Map of the study region with Municipal Waste Incinerators (MWIs), MWI areas and comparator areas.

- Carstairs deprivation index input variables: proportion of male unemployment, proportion of households without a car, proportion of overcrowded households, proportion of persons in private households with an economically active head of household in social class IV or V (partially skilled and unskilled occupations) (Carstairs and Morris, 1990).
- (iv) We computed the Euclidean multivariate distance between the MWI and comparator vectors and selected the comparator area with the smallest distance.

For the five incinerators located in the South East, we computed the comparator area for each of them independently; 28% of MSOAs were shared between control areas (see Fig. 1). We depict the steps for the choice of the comparator area (Supplementary material Fig. 1).

2.4. Outcome and confounder data

Data on infant mortality and sex ratio were obtained from the Office for National Statistics; counts of annual infant mortality were defined as the sum of deaths occurring between 0 and 365 days of life. Sex ratio is defined as the sum of the female births divided by sum of male births at area level.

We accounted for the following potential MSA-level confounding variables:

- Area-level socio-economic deprivation defined using the Carstairs 2011 score in quintiles (Carstairs and Morris, 1990)
- Population density from the 2011 Census
- Ethnicity (2011 Census) as the percentage of the MSA non-white female population ages 16–49 years old, split in three categories (Black, Asian and Other ethnicity), with cutoffs defined using the England and Wales average (12.1%) and twice the average (24.2%) (Hansell et al., 2013).
- NO₂ concentrations at 200 m × 200 m resolution, postcode head-count weighted to MSA level (Gulliver et al., 2013).

In sensitivity analyses, we investigated the use of MSA centroids within 4 km of the MWI, for consistency with previous published studies on incinerators (Candela et al., 2015; Candela et al., 2013). We also restricted the time window to include only those MWIs with data available for 5 years before/after the opening. Further, we allowed area-specific intercepts for all the MWI and comparator areas to account for possible heterogeneity of the MWIs.

2.5. Bayesian interrupted time series

We used a Bayesian hierarchical model to jointly fit the data for MWI (exposed) and comparator areas. We defined y_{ikt} as the counts of infant deaths, distributed as:

Table 1
Municipal Waste Incinerators (MWI) area characteristics.

MWI	Region	Opening years	MSOAs in 10 km MWI boundary (n)	MSOAs in 10 km comparator areas boundary (n)
Allington	South East	2006	38	35
Chineham	South East	2003	20	19
Crymlyn Burrows	Wales	2003	32	32
Grundon (Lakeside)	London	2010	77	75
Isle of Wight	South East	2009	9	8
Marchwood	South East	2004	52	43
Newlincs (Grimsby)	Yorkshire and the Humber	2004	20	21
Portsmouth	South East	2005	56	43

$$y_{ikt} \sim \text{Poisson}(\lambda_{ikt} E_{ikt})$$

where $i = 1, \dots, N$ MSOAs, t is time (1996–2012) and $k = 1, \dots, K$ MWI and E_{ikt} is the number of live singleton births. Then on the link function $\log(\lambda_{ikt})$ we specified a linear regression:

$$\log(\lambda_{ikt}) = \alpha + \beta_1 D_i + \beta_2 t + \beta_3 (t - t_{0k}) I(t > t_{0k}) + \beta_4 D_i t + \beta_5 D_i (t - t_{0k}) I(t > t_{0k}) + \sum_{i=1}^N \beta_z Z_i + b_i \tag{1}$$

where D_i indicates if the MSOA centroid lies within 10 km of an MWI ($D = 1$) or a comparator area ($D = 0$); t_{0k} ($1 \leq t_{0k} \leq t$) is the opening year for the k -th MWI and Z_i is the vector representing potential confounders at the MSOA level.

The regression coefficients are normally distributed centred on zero, with a variance equal to 10^4 . The area-specific random effects b_i are here defined as exchangeable, i.e. $b_i \sim N(0, \sigma_b^2)$, with the hyperparameter $1/\sigma_b^2 \sim \text{Gamma}(1, 0.00005)$ (Martins et al., 2013; Rue et al., 2009). We extended the specification below to allow for local spatial dependency.

In the Eq. (1) α indicates the log baseline risk for infant deaths; β_1 represents the difference in risk between MWI and comparator areas before the incinerator opened; β_2 indicates the general time trend, which we assumed was linear in this setting, but could be extended to be non-linear if appropriate; β_3 identifies a potential change in the time-trend in the comparator areas after the incinerator opening and β_4 is the coefficient describing the change in linear trend between MWI and comparator areas before the incinerator opened. Finally, β_5 indicates the change in time trend between MWI and comparator areas before-after the MWI opened. We present the model for count data using a Poisson distribution; for the log transformed sex ratio the distribution is Gaussian but the interpretation of the parameters remains the same. We extend the modelling framework in Eq. (1) to allow for spatially structured random effects (Freni-Sterrantino et al., 2018; Riebler et al., 2016; Simpson et al., 2017), appropriate when there is localised spatial dependence (Supplementary materials Appendix A).

2.6. Calculation of summary index

We constructed a summary index to describe the difference in before-after effects between MWI and comparator areas, for infant mortality:

$$\gamma = n^{EA} \left(\mu^{EA} - \mu^{EB} \frac{\mu^{CA}}{\mu^{CB}} \right)$$

and for sex ratio:

$$\gamma = \left(\mu^{EA} - \mu^{EB} \frac{\mu^{CA}}{\mu^{CB}} \right)$$

where $\mu^{CB} | \mathbf{y}, \mu^{CA} | \mathbf{y}, \mu^{EB} | \mathbf{y}, \mu^{EA} | \mathbf{y}$ ($E = \text{Exposed}, C = \text{Comparator}, A = \text{After}, B = \text{Before}$) are the posterior distributions obtained from Eq. (1) (see Supplementary material Table 1) and n^{EA} is the total number of deaths occurred in MWI areas after the opening. The $\mu^{EB} \frac{\mu^{CA}}{\mu^{CB}}$ gives an estimate of the effect of MWI opening adjusted for the comparator area trends. We report γ as the difference in deaths per 100,000 infants and as ratio difference for sex ratio.

3. Results

Overall from 1996 to 2012 there were 526,642 single live births and 2462 infant deaths recorded across the regions included in the study, across 304 and 276 MSOAs in MWI and comparator areas, respectively.

Descriptive statistics are shown in Table 2. Comparing periods before and after the opening of an MWI, infant mortality rates decreased from 4.8 to 3.9 (per 1000) in the MWI areas, 5.0 to 4.6 (per 1000) in the comparator areas. The inter-quartile ranges (IQRs) of infant mortality

were higher before opening in the MWI and comparator areas (9.62 and 9.43 per 1000 respectively), compared with after opening (IQR 7.93 and to 9.01 per 1000 respectively). We observed a similar trend for England and Wales over the same period; rates decreased from 6.0 to 4.1 (per 1000), rates decreased overall by 2.5% per year (Office for National Statistics).

For sex ratio, there was no difference between before and after MWI opening; in the exposed areas, average sex ratio was 0.96 (IQR 0.28) and 0.97 (IQR 0.26) respectively and it was 0.96 (IQR 0.25) and 0.97 (IQR 0.26) respectively in the comparator areas.

Table 3 shows the results of the regression model. The summary index γ indicates there were 8 fewer infant deaths per 100,000 live, single births (95% CI -62, 40) when comparing the before and after periods, adjusted for the global trend in the comparator areas. 27% of the variability in infant mortality was accounted for by the spatially structured random effect b (the posterior mean of the mixing parameter ϕ), and this was robust to selection of priors. The posterior estimates show that infant mortality was higher by 48% (95% CI 24, 76) for the more deprived areas and by 27% (95% CI 10, 47) for areas where the proportion of non-white ethnicity was above the national average.

For sex ratio, the summary index γ as ratio difference is -0.004 (95%CI -0.02, 0.01), and 46% of variability observed was accounted for by a spatially structured random effect.

Fig. 2 depicts the posterior mean of the linear predictors and credible intervals for the MWI and comparator areas. The lines are almost parallel, and the credible intervals overlap, indicating that the decrease in infant mortality risk is similar for both areas as is sex ratio.

By restricting the MWI boundary to 4 km in sensitivity analyses, there were 569 deaths among 113,940 live births across 59 MSOAs and 63 comparator MSOAs. We observed decreasing rates in both areas, from 5.4 to 4.9 (per 1000) before/after MWI opening for MWI areas and 5.0 to 4.5 (per 1000) in the comparator areas (Supplementary material Tables 3, 4, 5). Model estimates show the same spatial variability as observed for the 10 km analyses and a summary index γ of 6 deaths per 100,000 infants (95% CI -126, 126) in the MWI areas corrected for trends in the comparator areas. Mean sex ratio between MWI and comparator areas was the same as for the 10 km buffer and the summary index γ is 0.009 (95% CI -0.03, 0.07); spatial structured variability accounted for 21% of the variance.

Sensitivity analysis for 4 and 10 km buffers restricted to incinerators with data five years before and after the opening, did not alter the findings (Supplementary material Tables 6 and 7).

4. Discussion

We present a statistical framework that extends the ITS model to account for spatial dependence applied to the opening of MWIs. We applied the developed framework to evaluate the effect of the opening of a MWI on infant mortality and sex ratio at birth, and found no difference when comparing MWI areas with comparator areas. For infant mortality we accounted for the strong decreasing global trends seen across England and Wales over the study period.

A strength of our study is the use of all available data on MWIs and infant mortality and sex ratio from 1996 to 2012, for England and Wales, to investigate possible effects of MWI opening on these outcomes, hence avoiding selection effects (bias). Few studies to date have published results on infant mortality in relation to MWIs. Tango et al. (2004) reported no significant excess of infant deaths near 63 MWIs in Japan, but found a small statistically significant peak decline in risk with distance up to 10 km from incinerators. In addition, Dummer et al. investigated stillbirths and neonatal deaths and found no associations with incineration (MWI and crematorium) (Dummer et al., 2003).

In Ghosh et al. (2019) we previously reported neonatal and post-neonatal mortality for 2003–10 within 10 km of MWIs and found no association related to modelled emissions or distance. Here we use an alternative approach to investigate the association of infant mortality

Table 2
Descriptive statistics for MWI and comparator areas for each outcome at MSOA level, before and after MWI opening, within 10 km boundary.

	Live births	Infant deaths	Rates mean (IQR) × 1000	Sex ratio mean (IQR)
MWI (exposed) before opening	113,411	562	4.8(9.6)	0.97(0.28)
MWI (exposed) after opening	157,317	650	3.9(7.9)	0.96(0.26)
Comparator area before opening	107,844	551	5.0(9.4)	0.97(0.25)
Comparator area after opening	148,070	699	4.6(9.0)	0.97(0.26)

Table 3
Model estimates from the Bayesian hierarchical model with structured spatial random effect, and 95% credible interval for infant mortality and sex ratio (10 km boundary).

Outcome	Infant mortality	Sex ratio
Summary index γ	- 8(- 62,40) ^a	- 0.004(- 0.02,0.01)
Spatial mixing parameter (%) ϕ	27.1(3.5,68.3)	46.5(61,92.3)
Intercept α	- 4.90(- 5.19, - 4.62)	- 67.00(- 105.22, - 28.90)
Exposed vs control β_1	0.08(- 0.12,0.29)	5.02(- 24.14,34.19)
Linear time trend overall β_2	- 0.01(- 0.03,0.01)	1.68(- 1.86,5.23)
Linear time trend after opening β_3	- 0.02(- 0.07,0.02)	- 3.52(- 10.79,3.74)
Linear time trend exposed areas β_4	- 0.02(- 0.05,0.00)	- 2.80(- 7.01,1.40)
Linear time trend exposed after opening β_5	0.04(- 0.01,0.11)	4.64(- 4.43,13.72)
Carstairs quintiles - least deprived	-	-
	0.06(- 0.12,0.24)	17.27(- 3.55,38.12)
	0.12(- 0.04,0.29)	19.15(- 1.54,39.92)
	0.30(0.13,0.47)	9.01(- 12.37,30.48)
Carstairs quintiles - most deprived	0.39(0.22,0.56)	24.3(0.94,47.90)
Ethnicity [0–12.1]	-	-
[12.1–24.2]	0.24(0.09,0.38)	14.06(- 5.29,33.33)
[24.2–100]	0.41(0.22,0.59)	12.39(- 12.55,37.35)
Population density	0(0,0)	- 0.003(- 0.006,0)
NO ₂	- 0.02(- 0.03, - 0.01)	- 0.04(- 1.64,1.55)

^a Summary index γ (× 100,000).

with incinerators, taking advantage of time series to investigate potential ‘before/after’ effects.

For sex ratio, Williams et al. (1992) reported a significant excess in female births in one of three “at risk” districts compared to the Scottish average, at aggregated data level. No significant results were found with modelled incinerator generated dioxins since 1992 for one incinerator located in Taipei (Taiwan) (Lin et al., 2006); also no significant excess was found near 63 MWIs in Japan (Tango et al., 2004). In Italy Candela et al. (2013) evaluated the effect of 8 MWIs in the Emilia Romagna Region using dispersion models to estimate particulate matter (PM10) exposure, and found no significant results for sex ratio (Candela et al., 2013). Similarly, in UK, we previously reported no significant association with sex ratio (Ghosh et al., 2019), but using a dispersion model and distance in relation to 22 MWI, and not the ITS approach.

The ITS framework is well suited for policy evaluation of time-series studies in a quasi-experimental setting. Most previous studies using the ITS approach have not allowed for the possibility of analysing global trends, which is critical to interpretation of any before-after effects as these are non-randomised interventions. It offers advantages over difference in difference methods because it not only controls implicitly for baseline mean and trends but provides a model to deal with auto-correlation present in the data.

We aimed to maximize the similarity between exposed and comparator areas by including time-varying variables (i.e. live births) in the period before opening as well as socio-economic deprivation indicators. We investigated the inclusion of the MSOA rural/urban classification and NO₂ (a proxy for road traffic) as additional matching variables, but we found no association on the selected comparator areas. The fact that there was only a small posterior mean difference (and credible interval including zero) for β_5 (the parameter that indicates a potential

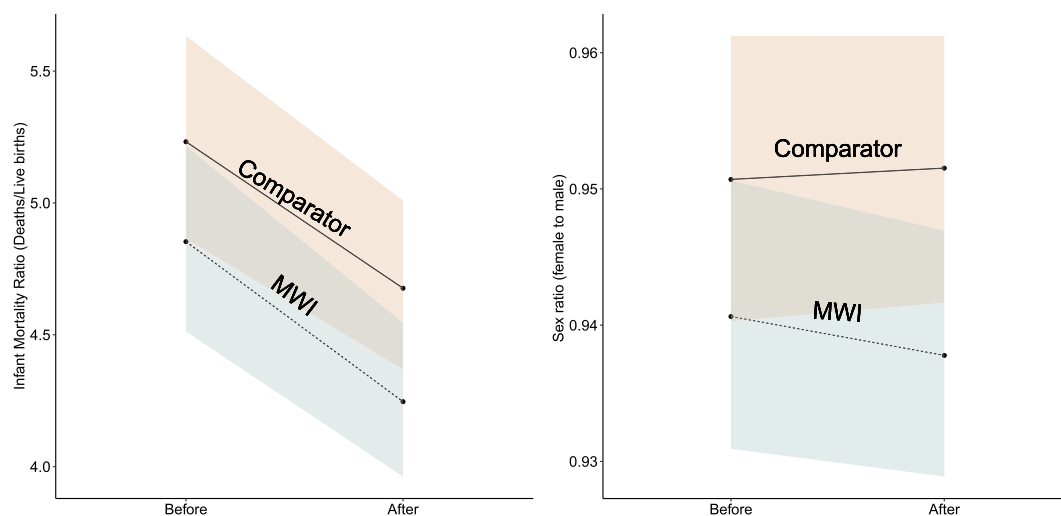


Fig. 2. Infant mortality (left) and sex ratio (right) for MWI and comparator areas before and after opening, model posterior estimates and 95% credible intervals (light blue for MWI areas, pink for comparator areas and brown for the overlapping credible intervals). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

difference in the trend before-after the incinerator opening, between MWI and comparator areas), suggests not only that there is no effect from the incinerator but also that the matching has been done appropriately. Nonetheless, despite having matched areas for socio-economic deprivation, we included spatial structured random effects to account for any potentially unmeasured confounders at small area level and an area-specific intercept to account for differences in MWI characteristics. The inclusion of a structured spatial effect resulted in a smaller Deviance Information Criterion (model selection index) compared to a spatial unstructured random effect (data not shown) and should improve inference in the ITS approach (Ning et al., 2019). Area-specific intercept did not improve the DIC, as the intercepts had similar values.

The difference before-after opening is represented as a step or shift in the intercept and slope in the regression models. We found no evidence of temporal autocorrelation or seasonality in the residuals of our model, suggesting that a linear time effect was appropriate in our data, although the proposed framework is extremely flexible and can deal with non-linearity via semi or non-parametric functions (e.g. splines or Gaussian processes) if needed.

Our approach has limitations. The official opening date for each MWI was provided by the Environment Agency but may not have coincided with the actual operating date. Douglas et al. (2017), showed that MWIs were not all operating continuously over this time period, due to repairs, upgrades, and closures. This might have resulted in some misclassification of the before-after opening period.

Other limitations include lack of gestational age for the time period 1996–2006 and lack of knowledge of maternal address before birth, as 24.4% women move during pregnancy (Hodgson et al., 2015), which meant that we could not analyse possible exposures during pregnancy, as an alternative exposure window. Furthermore, the confounders used in our analyses were obtained from the Census 2011, and we assigned them equal weight before/after MWI opening in the matching process as they were not time varying. Although the population characteristics are likely to remain similar over this relatively short period, we cannot exclude possible residual confounding.

Distance is a relatively crude measure of exposure to an MWI, but it may capture exposures not reflected in the stack emissions modelling (e.g. transport of waste to the MWI) as well as non-airborne exposure pathways. We selected a 10 km boundary around each MWI, as has been used in previous studies in the UK (Reeve et al., 2013; Elliott et al., 1996; Gouveia and Prado, 2010; Ghosh et al., 2019), also including the dispersion modelling study of MWI emissions carried out by Douglas et al. (2017) for all 22 British MWI between 2003 and 2010. In a sensitivity analysis, we also selected a 4 km boundary based on previous studies conducted in Italy (Candela et al., 2013; Candela et al., 2015; Ranzi et al., 2011), with results similar to those we observed for 10 km. Smaller buffer size of 1–3 km were not considered because of limited statistical power.

In conclusion, we did not find an association between the opening of a new MWI and changes in infant mortality trends or sex ratio at birth for 10 and 4 km buffers, using distance as proxy of exposure, after taking into account temporal trends in comparator areas and potential confounding factors.

The framework we propose, applying ITS to small areas, can readily be adapted to take account of different types of data, structures, spatial and temporal dependencies. The method may be relevant to applications that investigate a potential causal effect of an intervention or policy, taking advantage of the quasi-experimental design.

Conflict of interest

Anna Hansell declares a Greenpeace membership but has not received any money from the organisation nor has been involved in campaign, nor other relationships or activities that could appear to have influenced the submitted work. All other authors declare no conflict of interest.

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Contributors

AFS is the guarantor of this paper. All authors had full access to all of the data in the study and can take responsibility for the integrity of the data and the accuracy of the data analysis.

Data and computing code

Births and deaths data were obtained from the Office for National Statistics (ONS) National Mortality, Births and Stillbirth registers for England and Wales.

2011 Census aggregate data came from the Office for National Statistics (for England and Wales): UK Data Service (Edition: June 2016).

Health data are available from the data providers on application with appropriate ethics and governance permissions, but we do not hold data provider, ethics, or governance permissions to share these datasets with third parties. The computing code is available upon request to the corresponding author.

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Appendix A. Supplementary data

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Appendix C Wilen et al (2007) Fine Particle Emissions of Waste Incineration



FINE PARTICLE EMISSIONS OF WASTE INCINERATION

Prepared for
IEA BIOENERGY AGREEMENT - TASK 36

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Jorma Jokiniemi





PREFACE

This report is a summary report compiled for the IEA Bioenergy Agreement Task 36 – Energy Recovery from MSW. The report is based on results obtained in a 3-year research project financed by Finnish Funding Agency for Technology and Innovations (TEKES, the FINE Research Programme), VTT and companies involved in energy-to-waste businesses.

Complete results will be published in VTT publication series in 2007.

The authors

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ABBREVIATIONS

BLPI	Berner Low Pressure Impactor
C	Concentration
D ₅₀	Cut-off diameter in cyclone or impactor
dm	Difference in Mass
D _p	Particle diameter (aerodynamic diameter)
ELPI	Electric Low Pressure Impactor
ESP	Electrostatic Precipitator
FF	Fabric Filter
FTIR	Fourier Transform Infrared Spectrometer
IC	Ion Chromatography
ICP-MS	Ion Coupled Plasma Mass Spectroscopy
m	Mass (of particles)
N	Number (of particles)
NTP	Standard temperature and pressure, 0°C, 101 kPa
P	Power
<i>P</i>	Penetration of particles through filter
ppm	parts per million
SRF	Solid Recovered Fuel
Stk	Stokes number
T	Temperature
TG	Thermogravimetric analyser, thermobalance
U	Velocity of gas
UM	'Urban Mill' rejection from processing of SRF

1 INTRODUCTION

Negative health effects of fine particles have gained extensive publicity, energy production being one of the largest producers of fine particles. EU's Waste Incineration Directive (200/76/EC, WID), which come into full force in the beginning of 2006, defines clearly what kind of emissions incineration plants can release. The limits also apply to co-combustion e.g. in coal or biomass plants. The directive outlines the total particle emissions of flue gases to 10 mg/Nm³ and heavy metals regarding 12 elements, respectively. The WID does not make a stand on possible fine particle emissions; it limits the total dust emissions.

Current energy use of waste in Finland has been mainly co-firing of Solid Recovered Fuels (SRF) in biomass or coal fluidized boilers. Current waste utilization targets in Finland an EU landfill limitations calls for increasing waste utilization in Waste To Energy (WTE) applications.

Waste originating fuels contain typically higher amounts of certain elements, e.g. trace metals, which are also typically more unevenly distributed than in conventional fuels. As they are readily released to the gas phase during combustion, their concentrations in fine particles may be significant. In order to avoid large trace metal emissions, the behavior of the trace metals needs to be understood in waste combustion. The particulate cleaning devices used in WTE plants need to have high collection efficiencies in the whole fine particle range.

The aim of the project was to study formation of fine particle emissions, especially the effect of waste quality on fine particle formation and the amounts and distribution of harmful substances (like heavy metals and chlorine) in particles. The project was implemented in three parts. The first part includes incineration tests in a pilot-scale test rig. In second part, emissions at full-scale waste incineration plants and co-combustion plants were measured. The third part concentrates on studying direct dependency of fuel quality and fine particle formation with laboratory scale investigations and modelling.

2 HEALTH EFFECTS

Increasing the degree of waste utilisation sets requirements also to wastes' energy utilisation. EU's waste incineration directive, which comes into force from the beginning of 2006, defines clearly what kind of emissions incineration plants can release. Fine particles born in waste incineration and fine particle purification have been studied in Tekes' FINE technology programme. Fine particles are particles, their aerodynamic diameter of which is below 2.5 µm (PM2.5). Fine particles penetrate deep into lungs when inhaled and they have been proved detrimental for health in many studies. According to estimation of health effects conducted in CAFE programme (Matti Vainio, Kansanterveys 5/2005), PM2.5 particles shorten the average statistical life expectancy in Europe with about nine months, which is caused by premature death of over 300 000 people in a year.

Negative health effects of fine particles have gained extensive publicity, energy production being one of the largest producers of fine particles. Health effects of fine particles are especially related to incineration originated particles, such as traffic and power plants' particles. The most significant fine particle sources are small-scale combustion of wood at fireplaces in homes and sauna stoves. Fine particle emissions of small-scale combustion were studied in PIPO research project funded by Tekes and companies (University of Kuopio, final report: <http://www.uku.fi/laitokset/ifk/publications.shtml>). Emissions were also studied in power plant scale and in industrial boilers in a research project called PIHI-RAME lead by VTT and Finnish Meteorological Institute. (Jokiniemi et al., VTT Research Notes 2258/2004).

3 BOILERS, FUEL PREPARATION AND AIR POLLUTION CONTROL SYSTEM

Tests were performed in four different commercial plants and in one pilot plant. Two waste incineration plants were situated in Sweden, Figure 1 and Figure 2 . The co-combustion plant, Figure 3, and the district heating plant, Figure 4, and the pilot plant were situated in Finland. Specific performance data of the commercial plants are given in Table 1.

3.1 NORRKÖPING, HÄSSELHOLM, KAUTTUA AND SIILINJÄRVI POWER PLANTS

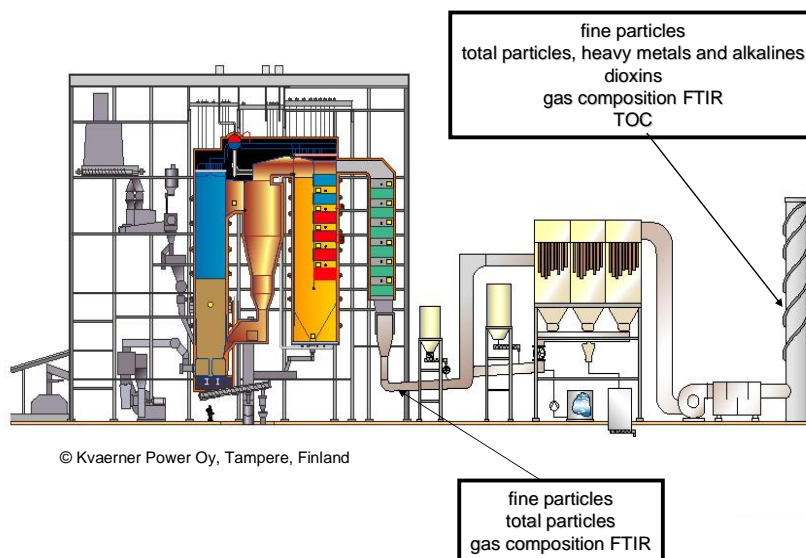


Figure 1. The Norrköping CFB boiler and performed measurements.

Plant	Norrköping	Hässleholm	Kauttua	Siilinjärvi
Owner	Sydkraft Östvärme AB	Hässleholm municipality	Fortum Heat and Power Oy	
Commissioned	2002	2003	1981	
Boiler output	75 MW	20 MW	65 MW	10 MW
Electricity output	11 MWe	1,7 MWe to own use	7 MW	-
Heat output	65 MWth	18 MWth	26 MW process steam, 4 MW district heat	10 MW
Boiler supplier	Kvaerner Power Oy	Babcock & Völund	Ahlström Pyroflow	Wärtsilä
Boiler type	Circulating fluidised bed	grate fired	Circulating fluidised bed	Rotating grate boiler
Fuel	waste; 30-50% household, 50-70% industrial	waste; 70% source separated household, 30 % presorted industrial	42% wood and construction waste wood , 30% peat, 10% Solid Recovered Fuel, 9% Coke	sawdust, bark, construction wood waste

Table 1. Data on the test plants.

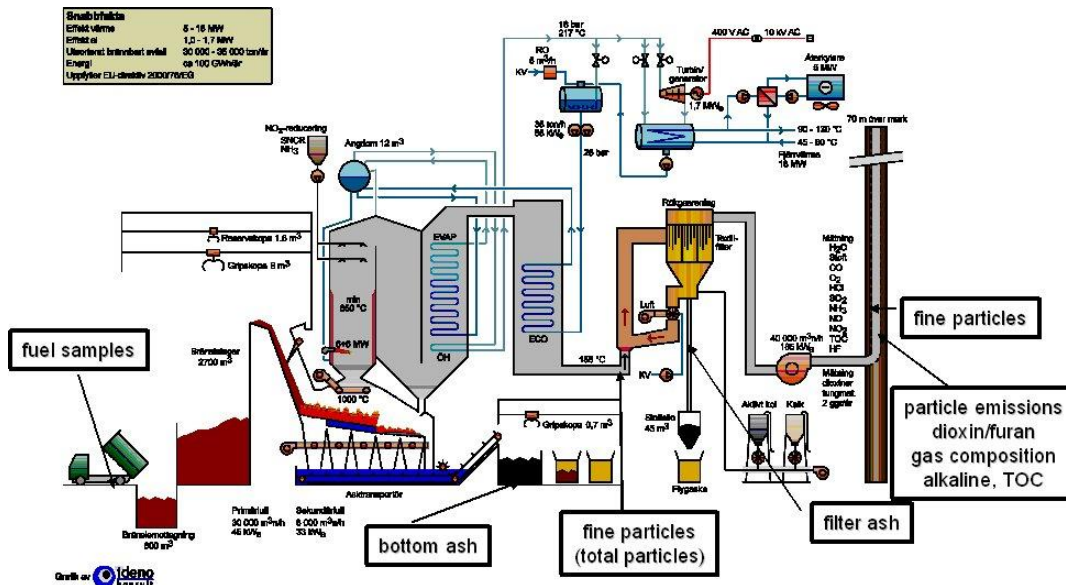


Figure 2. Flue gas cleaning for the grate boiler at the Hässleholm district heating plant.

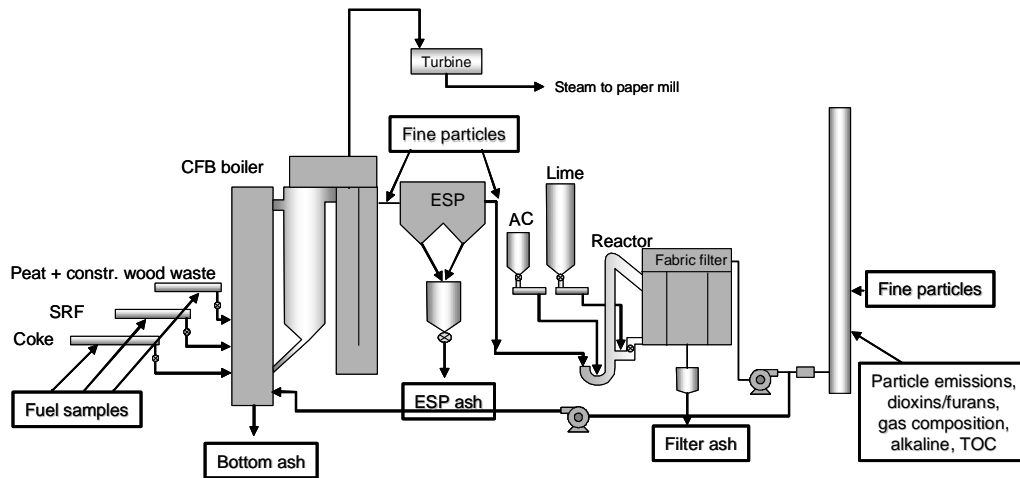


Figure 3. The Kauttua CFB boiler and performed measurements.

The flue gas cleaning systems in all three plants (Norrköping, Hässleholm, Kauttua) are essentially the same. Activated carbon was used for Hg and dioxin absorption in NID, and lime feed was used for HCl and SO₂ reduction. In Kauttua the baghouse filter is installed after the original ESP and was commissioned a few months before the time of the measurements. At Siilinjärvi the flue gas cleaning is carried out with the cyclone and the electrostatic precipitator (ESP).

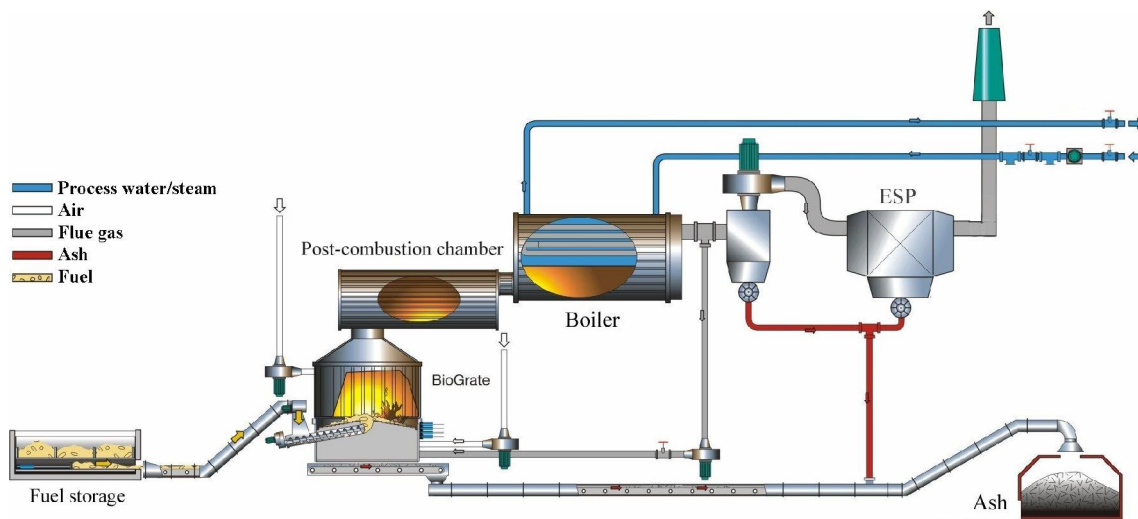


Figure 4. The Siilinjärvi process chart.

In Norrköping plant fuel is crushed in a primary shredder. After magnetic separation fuel is cut with hammer mills. The shredded fuel is stored in a barn-type intermediate storage before conveying to the plant. At the plant an Eddy Current system is used to separate non-ferrous metals from the feed. Figure 5 shows the shredded fuel as fed into the boiler.



Figure 5. The shredded fuel as fed into the boiler at the Norrköping waste-to-energy plant.

The waste in Hässleholm is delivered by trucks to the plant and unloaded into the waste bunker. The fuel is fed by a crane to an intermediate bunker, and from that directly to the grate, Figure 6. Because of an efficient source separation scheme the waste contained little organic residues.



Figure 6. Waste fuel sample from Hässleholm.

At Kauttua fuels are fed to the plant by three separate conveyers. The peat and wood residues are mixed on the courtyard. The mixture is fed to a disc screen before conveyed to the boiler. Metals are also separated by a magnet. The paper and plastic waste is fed to an on-site crusher producing a coarse fuel fraction, Figure 7. This is fed to the boiler by a separate conveyor and third conveyor is feeding the relatively small amount of crushed coal/coke to the boiler. The fuel mix during the measurements was the one mentioned in Table 1.



Figure 7. Waste wood (left, containing occasionally also paper and plastics) and SRF fractions at the Kauttua plant.

Two different fuel mixes were used in Siilinjärvi plant during tests. In the first day/set-point, the fuel was the bark and sawdust 50/50 mixture, which is normally used in the boiler. On the second day/set-point the intention was to add 30% construction wood to the bark – sawdust mixture.

3.2 KARHULA CFB PILOT PLANT

Combustion tests with five different kinds of waste fractions were performed in December 2003 at the Foster Wheeler Energia Oy's 1 MW fluidised bed pilot plant in Karhula, Finland. These waste fractions are presented in Table 2. The main objectives of the test were to study the effect of waste quality on fine particle emissions, in particular the concentrations of detrimental substances (mainly heavy metals) they contain and the distribution of the substances in different particle sizes. Important topics were also other waste related gaseous emissions like HCl and mercury.

Table 2. Fuel material used in the pilot plant tests.

Type of waste	Delivery	Composition	Abbreviation
Construction wood waste	Lassila & Tikanoja, Kerava	Construction (mainly) and demolition waste wood	Construction wood waste
Solid recovered fuel, class I	Lassila & Tikanoja, Kerava	Commercial and package waste (mainly paper, board and plastics)	SRF I
Solid recovered fuel, class III	Loimihämeen Jätehuolto, Forssa	Produced from the dry fraction of source separated household waste	SRF III
Solid recovered fuel, class I, reject after fibre removal: Urban Mill process reject	Metso (origin: Loimihämeen Jätehuolto, Forssa)	Commercial and package waste, fibres removed in a pulping process	SRF I UM
Solid recovered fuel, class III, reject after fibre removal: Urban Mill process reject	Metso (origin: Loimihämeen Jätehuolto, Forssa)	Produced from the dry fraction of source separated household waste, fibres removed in a pulping process	SRF III UM

The pilot circulating fluid bed boiler is equipped with a bag house filter with a possibility for additive injection and a scrubber. The scrubber was by-passed during the tests. The fuel amount for a 24 hour set point was estimated to be 4-5 tons, a sufficiently large and representing fuel amount considering the inhomogeneous nature of waste fuels. The combustion equipment has been proven to correspond to a real boiler and its conditions.

4 SAMPLING AND MEASUREMENTS

During the particle measurement campaigns in the power plants, the fuel sampling was arranged so that the fuel quality matched with the particle samples collected at the filter inlet. From the fuel samples, the ordinary fuel characteristics and trace element content were determined. Also, ash samples were analysed for their elemental content. Ash samples were taken both from fly and bottom ashes.

4.1 FUEL AND ASH SAMPLES

In *Siilinjärvi*, fuel samples were taken at every 30 minutes, and the sampling was matched with the time when the particle measurements were carried out for the filter inlet and outlet. Ash samples were also collected during the test. For the analysis, the ash samples taken from the multi-cyclone and electrostatic precipitator (ESP) were combined to one fly ash sample and two ashes from the grate were combined to a bottom ash sample.

In *Norrköping*, fuel samples were taken every 30 minutes when the particle measurements were carried out for the filter inlet and outlet. Ash samples were taken approximately each hour or two hours.

During the two-days measuring campaign in *Hässleholm*, fuel samples were taken twice a day by lifting a 1-2 m³ batch of waste to the floor of the receiving station, where it was divided by coning-and-quartering method and taking a 100 liter fuel sample for preparation of the final sample for analysis (Figure 8). Ash sampling was carried out so that bottom ash samples were taken from the ash containers and the fly ash samples were taken after the bag house filter by means of a sampling device.



Figure 8. Fuel sampling at Hässleholm

In *Kauttua*, the fuel samples were taken at approximately 1 hour intervals from all the fuel components: SRF, peat-wood waste mixture and coke. Ash samples were collected from bottom, ESP and filter ash.

At Siilinjärvi, for the analysis, the ash samples taken from the multi-cyclone and ESP were combined to a fly ash sample and two ashes from the grate were combined to a bottom ash sample.

4.2 GAS ANALYSIS

During the measurement campaign, the following components were measured using the following methods or standards.

O₂ was measured with electrochemical cell. Calibration was made with ambient air (20,9 %). Method is accredited VTT Processes Measurement Method PRO320202 and it follows the principle of EPA Method 3A.

H₂O, CO₂, CO, CH₄, NO, NO₂, N₂O, NH₃, SO₂, HCl and HF were measured. In Hässelholm also TOC was measured. At Siilinjärvi H₂O, CO₂, CO, NO, NO_x, SO₂ and HF were measured.

In some plants the stack gas composition was followed with FTIR, Fourier Transform Infrared Spectroscopy, which is a continuous online gas analyzer. Infrared spectroscopy is based on the ability of all gases to absorb infrared radiation with a wavelength specific to the gas. Only exceptions are noble gases and biatomic molecules with the same nuclei (N₂, O₂ etc.) which can't be measured with FTIR. The measurement result with FTIR is a spectrum, from which the components of the sample gas are determined. The quantities are calculated from the intensity of the absorption lines. The sample gas was filtered from particles and the temperature of FTIR and sample lines was kept at 150°C.

The process data were collected from the process control system.

Standard method EN 14385 was used to determine trace element concentrations in particles and in gas phase both in the stack and filter inlet. From filter inlet only Hg concentration was determined from the gas phase samples. Hg was analyzed with CVAAS, Tl with GFAAS and other trace metals with ICP-AES.

4.3 FINE PARTICLE MEASUREMENTS

Fine particle samples were taken both before and after flue gas cleaning. At Kauttua particles were also measured between ESP (Electro Static Precipitator) and fabric filter.

BLPI, Berner Low Pressure Impactor, is an eleven stage cascade impactor used to measure particle mass size distribution and mass concentration. In an impactor the sample flow runs through a thin nozzle with a certain velocity. After the nozzle the flow is forced to turn 90°. The particles larger than the cut-off diameter D_{50} impact on the particle collection substrate, while smaller particles follow the stream to the next impactor stage with a smaller cut-off diameter. The flow rate through BLPI depends only on the inlet pressure and BLPI and the sample flow temperature and is 19 l/min (NTP) at 140°C and normal pressure. The cut-off diameter on the finest stage is 0.022 μ m and on the coarsest stage 16.22 μ m. In these measurements a pre-cutter cyclone was used before impactor to collect the coarse particles usually larger than $D_p > 5\mu$ m.

The particle collection substrate material was poreless Nuclepore polycarbonate filter. The polycarbonate filter is especially good for chemical analysis of particles because it contains very little other elements but carbon. The substrates were die-cut to the desired size and shape and greased with Apiezon L ultra high -vacuum grease. After greasing the substrates were baked in a 150°C oven for 4 to 16 hours to evaporate the grease solvent before weighing. Greasing reduces the particle bouncing from the substrate when they hit it. The repeatability in weighing of the substrates is approximately $\pm 4\mu$ g per weighing. For the best results 1mg of particles should be collected on a substrate.

ELPI, Electric Low Pressure Impactor, is a twelve stage cascade impactor similar to BLPI. The main difference is that the particles are charged with a needle charger at impactor inlet. The charge is measured separately from each stage with electrometers and the particle number size distribution is calculated from the charge distribution. The particle size distribution can be monitored in real time. The lowest stage was a filter, which collected the particles smaller than 30nm. On the coarsest stage the cut-off diameter was 10 μ m.

The particles collected with BLPI were analyzed to determine their chemical composition. The water soluble anions and cations were analyzed with Ion Chromatography, IC. One quarter of each BLPI substrate was cut and dissolved in 1ml methanol and 9ml water. Pre-cyclone samples were distilled in 10ml water. If

The trace metals were analyzed with ICP-MS, ion coupled plasma-mass spectroscopy. A quarter of each BLPI substrate was cut and dissolved in 2% HF and 10% HNO₃.

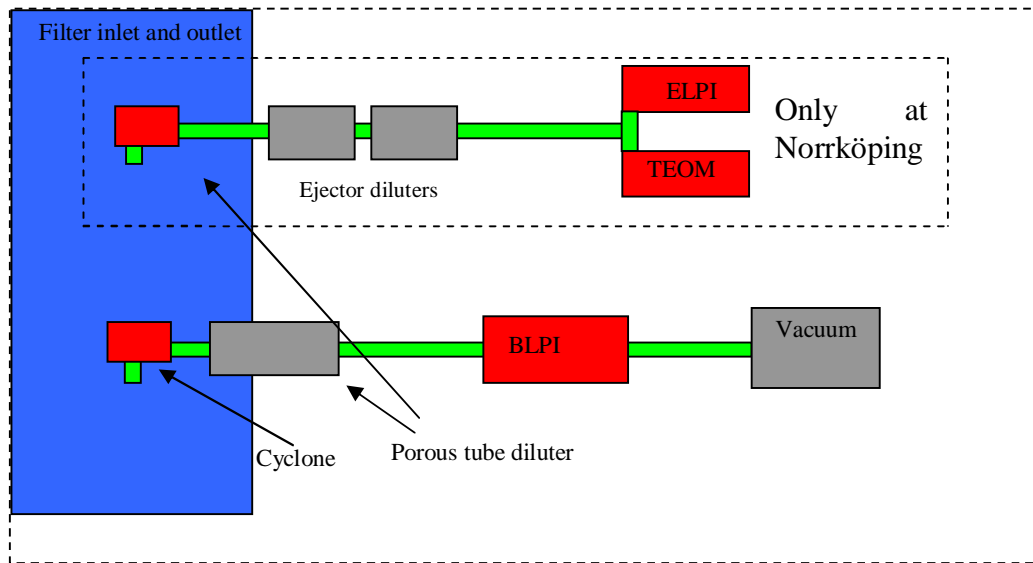


Figure 9. The experimental set-up in BLPI measurement and ELPI and TEOM measurements.

The experimental set-up for particle sampling was made as similar as possible in all field measurement sites (Figure 9). A pre-cutter cyclone was placed inside the duct to collect the coarse particles, typically particles with $D_p > 5 \mu\text{m}$, which otherwise would have had heavy losses in the sampling. After the cyclone the sample flow was diluted with a porous tube diluter, which was located partially inside the duct. BLPI was placed right next to the diluter. In BLPI measurements the dilution air and BLPI were pre-heated to 120°C - 140°C . The dilution air flow was controlled with a critical orifice. BLPI and ELPI are intrinsically critical orifices and their flow rates are known. The TEOM flow rate is adjusted with a mass flow controller. Maximum temperature was 140°C , because the Nuclepore polycarbonate substrates used to collect the particles in BLPI melt at higher temperatures.

The experimental set-up in ELPI measurements was similar to BLPI measurements except that the sample flow was quenched to 20°C - 50°C in the porous tube diluter and the sample line length between the diluter and ELPI was 1m-3m. ELPI operated at ambient temperature, typically 20°C , and. The dilution rate was monitored with a CO_2 analyzer.

All equipment inner surfaces, the pre-cyclone, the porous tube diluter, BLPI and ELPI, were made of stainless steel. The sampling lines were 1cm in diameter and either stainless steel or Tygon tubing.

At Norrköping ELPI and TEOM measurements were also performed. Set-up in Norrköping is also demonstrated in Figure 10.

At Siilinjärvi experimental set-up included BLBI measurements from both filter inlet and outlet and ELPI and TEOM measurements from filter outlet. Set-up was slightly different from other plants since the pre-cutter cyclone was placed outside the duct and the 4mm nozzle was bent 90° .



Figure 10. Fine particle measurement from duct between filter and stack at Kauttua (on left) and from the stack at Norrköping (on right).

4.4 SAMPLING AND MEASUREMENTS AT KARHULA PILOT PLANT

Figure 11 shows the Karhula pilot test rig setup and the sampling points of fine particles, gaseous emissions and solid fuels and ashes.

Fuel samples were taken during the set points with 20 minutes intervals. During the time span of the fine particle sampling the fuel samples were taken with 10 minutes interval for Construction wood waste and SRF I and with 5 minutes interval for the rest of the fuels to be able to trace possible emission peaks in the fine particle samples to the corresponding fuel sample when particle sample was taken at the inlet of the filter. Ash samples were collected at the bottom of the reactor and the boiler and in the bag house filter.

The gaseous components measured before the filter were O_2 , CO , SO_2 and NO_x . O_2 , CO , SO_2 , NO_x , CO_2 , N_2O and HCl were measured after the filter in addition to the on-line measurement of mercury. Control measurements of Hg , HCl and heavy metals of the flue gas were carried out during a SRF III set point by gas sampling and laboratory analysis.

All online measurements including process data were collected from the process control system. The feed rate of fuels and additives were controlled manually by weighing the batches.

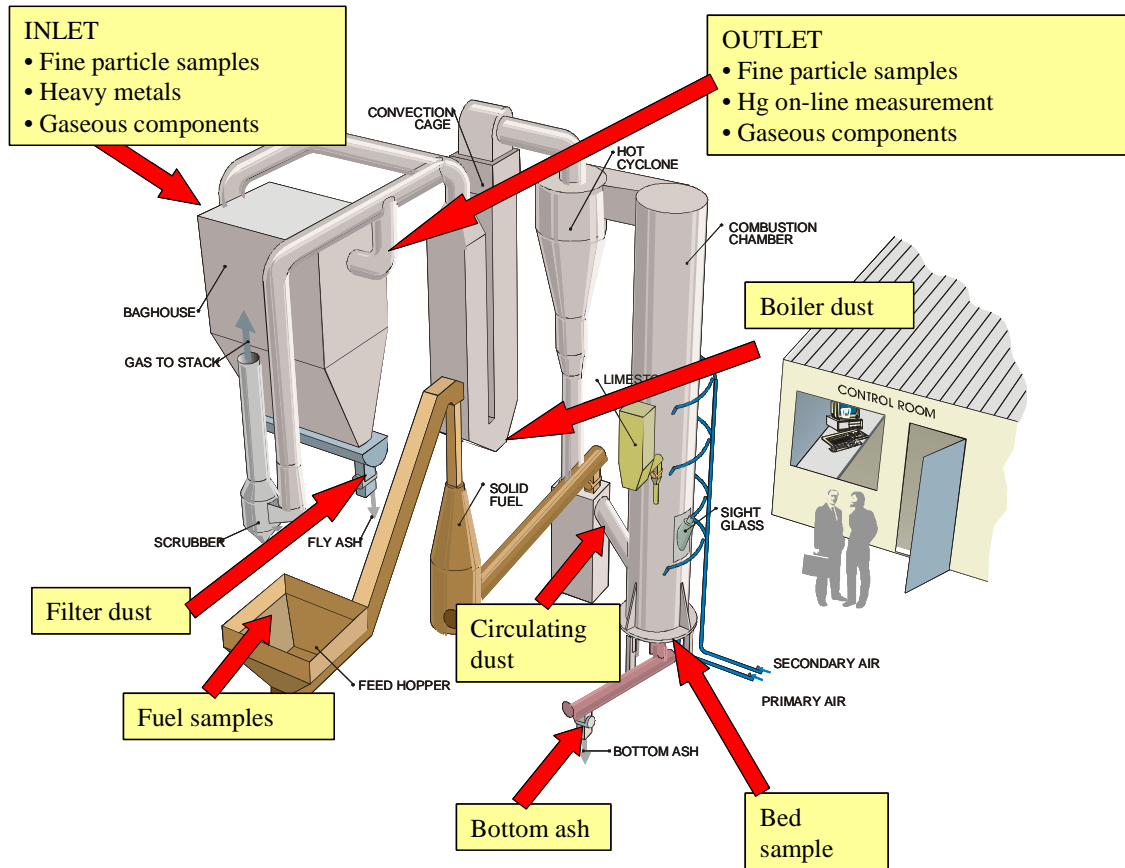


Figure 11. Figure Sampling and measurements at Karhula test rig.

4.5 FUEL ANALYSIS METHODS

For the elemental analysis, dried fuel samples were dissolved in a microwave oven with the mixture of acids ($\text{HNO}_3 + \text{H}_2\text{O}_2 + \text{HF}$). Small amount of insoluble matter remained from all the samples, but the uncertainty was assumed to be insignificant. From the solutions concentrations of As, Cd, Ni, Sb and Tl were measured by the graphite furnace atomic absorption spectrometry (GFAAS), concentrations of Co, Cr, Cu, Mn, Pb, Sn, V and Zn were measured by the inductively coupled plasma atomic emission spectrometry (ICP-AES) and concentrations of Hg were measured by the cold vapour atomic absorption spectrometry (CVAAS). Chlorine and bromine in ashes were analysed in VTT by INAA. Ash samples were analysed by The Geological Survey of Finland (GTK).

5 FUEL AND ASH COMPOSITION

5.1 FUEL ANALYSES

The average compositions of fuels are presented in Table 3, and the variation for element contents in Figure 12. The variation was determined by the difference between maximum and minimum. This variation (as relative, % on the average) in single samples during a test campaign was obtained from the pilot plant tests. The relative difference for trace elements was above 10% and for the ordinary fuel characteristics it was less than that. The variation determined for all fuels followed the same pattern as in the pilot plant test. For metals it was quite large, highest for Cu, then Cd and Ni, about 3 times the average value. The worst behaving element was copper, for example in Norrköping the Cu concentration was 130 mg/kg at its lowest and 11500 mg/kg at its highest.

Table 3. The average fuel compositions.

	dry conc.	Hässleholm Waste	Norrköping Waste	Kauttua SRF 1	Kauttua Peat-wood waste	Kauttua Coke	SRF 1	SRF 1 UM	SRF 3	SRF 3 UM
Fraction		100%	100%	~24%	~68%	~8%	100%	100%	100%	100%
Mercury (Hg)	mg/kg	0.11	0.32	<0.03	0.04	0.04	0.04	0.06	0.07	0.12
Metallic (Al) aluminium	mg/kg	3900	4200	800	2200		-	-	-	-
Arsenic (As)	mg/kg	4.3	10	0.9	74	4.1	6	17	14	7
Cadmium (Cd)	mg/kg	0.8	2.5	0.06	0.4	0.4	1.9	3.3	5.1	21.3
Cobalt (Co)	mg/kg	11	8.8	0.9	1.7	5	1.4	1.3	3.9	5.2
Chromium (Cr)	mg/kg	220	90	6.1	84	83	42	110	157	230
Copper (Cu)	mg/kg	376	4500	22	69	22	320	30	770	1590
Manganese (Mn)	mg/kg	166	260	22	84	87	36	42	230	240
Nickel (Ni)	mg/kg	144	34	3.0	7.5	40	16	8.5	32	39
Lead (Pb)	mg/kg	247	170	4.9	77	19	46	43	110	140
Antimony (Sb)	mg/kg	13	52	15	1.6	0.65	91	39	33	52
Tin (Sn)	mg/kg	45	37	1.2	3.7	1.1	16	41	12	18
Thallium (Tl)	mg/kg	<1	<1	<1	<1	<1	<2	<2	<2	<2
Vanadium (V)	mg/kg	9.6	9.4	<3	5.3	40	<5	<5	5.9	14
Zinc (Zn)	mg/kg	1160	760	30	300	68	730	270	420	620
Fluorine (F)	mg/kg	60	67	<50	<50	<50	<50	<50	63	140
Chlorine (Cl)	%, d.	0.94	1.0	0.08	0.17	0.1	0.39	0.68	1.0	1.7
Soluble Na+K	%, d.	0.5	0.6	0.07	0.07	0.01	-	-	-	-
Sulphur (S)	% d.	0.26	0.40	0.06	0.16	0.58	0.08	0.06	0.14	0.15
Nitrogen (N)	% d.	0.6	1.0	0.7	0.8	1.4	1.1	0.63	0.90	0.57
Carbon (C)	% d.	48	47	56.6	48.5	82.1	59	61	51	60
Hydrogen (H)	% d.	6.6	6.5	8.2	5.6	3.4	8.0	8.4	7.3	8.8
Volatile matter	% d.	71	72	85.9	73.6	18.3	83	85	75	78
Ash	% d.	20	18	6.9	6.8	9.9	7.3	4.1	15.6	13.7
H ₂ O	%	26	42	7.3	36.3	16	-	-	-	-
Higher heating value HHV	MJ/kg dry	21.1	20.9	26.3	19.8	31.4	26.9	28.0	23.4	29.0
Lower heating value LHV	MJ/kg dry	19.7	19.5	22.6	11.0	25.4				
Lower heating value LHV	MJ/kg ar.	14.0	10.4							

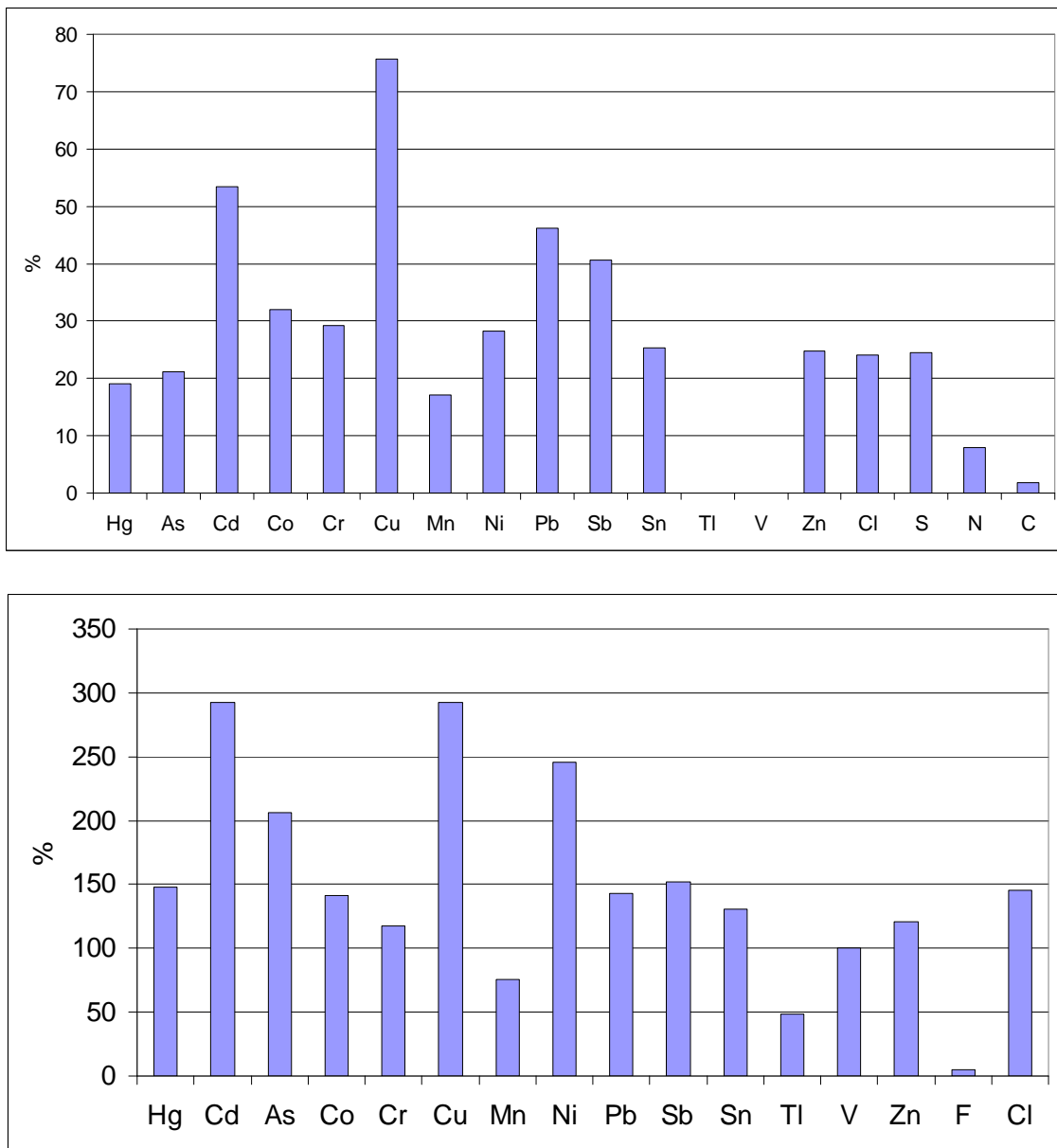


Figure 12. Variation of each element, % on average; pilot plant fuels upper graph and all fuels lower graph.

As results from Norrköping mixed fuel sample analyses, average moisture was 42%, volatile matter 72% and ash contents 18% in dry fuel. The concentration of Cu varied significantly as mentioned above, but all other element concentrations were surprisingly even between the samples. An overall estimate of the “cleanness” of the Norrköping waste fuel would rank the fuel similar or a somewhat more contaminated than the typical SRF III waste derived fuel used in the pilot tests in Karhula in December 2003.

High chlorine contents of average 70 g/kg were measured for fly ashes, from the samples taken each day. Copper content was high both in fly ash and bottom ash, average 6 g/kg. Lead content was 3.4 g/kg in fly ash and 1.4 g/kg in bottom ash.

The ash samples taken in each day at Hässelholm were combined and analyzed as well. Chlorine content in fuel was 1% in average, the main component in fly ash with 21% and only 0.4% in bottom ash. Zinc concentration in fly ash was quite high, 1.7%, in the fuel it was 0.1% and 0.4% in bottom ash. Although copper (chloride) should evaporate in relatively low temperature, more of it is left in the bottom ash than other volatile elements. Copper may exist in larger pieces, which don't stay long enough in furnace to evaporate entirely.

In the fuels, metallic aluminium content at Kauttua was also high in the peat and wood waste mixture, although visually there seemed to be plenty of aluminium in SRF. Arsenic chromium and lead contents were unexpectedly high in the peat and wood waste mixture compared to other SRF and wood waste analysis. Antimony content was high in one of the SRF samples (SRF 2). In coke, the chromium and sulphur contents were relatively high. In ashes, arsenic, cadmium, manganese, lead, antimony, thallium, vanadium zinc and fluorine contents were higher in the ESP ash than in the other ashes, while cobalt, chromium, copper, nickel and tin contents were both high in the ESP and in the bottom ash. Mercury, chlorine (Cl about 12 %) and bromine were clearly higher in the baghouse filter ash than in the others.

The 'dirtiest' fuels with the highest concentrations of the WID trace elements and Chlorine were SRF 3 UM and the waste fuel in Norrköping and Hässelholm. The 'cleanest' fuel was SRF 1 in Kauttua, even including Kauttua coke and peat-wood-construction waste wood mix.

5.2 ASH ANALYSES

Following ashes were analysed in this study:

- Pilot test SRF I bed material, circulating material and filter ash
- Pilot test SRF III bed material, circulating material, fly ash from the boiler bank and filter ash.
- Siilinjärvi sawdust + wood waste, combined multi cyclone and electrostatic precipitator (ESP) ash, bottom ash
- Norrköping filter and bottom ash
- Hässelholm filter and bottom ash
- Kauttua ESP, filter and bottom ash

The content of the following elements were determined: Hg, As, Cd, Co, Cr, Cu, Mn, Ni, Pb, Sb, Sn, Tl, V, Zn, F, Cl and Br.

According to the results, the high content of arsenic and thallium was striking in the combined ash sample collected from the multi-cyclone and electrostatic precipitator in

Siilinjärvi, although wood waste was mixed to untreated wood only 30. The As content in the feedstock was low and much lower than that in Kauttua. Mn content was also much higher in Siilinjärvi ashes (also in the bottom ash) compared to others, although the content in the feedstock was not higher than that of SRF III, SRF III UM, Norrköping or Hässleholm.

Hässleholm filter contained high amounts of Cd (159 mg/kg), Ni (400 mg/kg), Zn (17 g/kg ash), F (3 g/kg ash), Br (1.5 g/kg ash) and Cl (213 g/kg ash). The source for F can be the Teflon containing materials (or coatings) and that for Br the fire retardants. In SRF I filter ash the Br content was significantly higher (almost 1 g/kg ash) than in SRF III. This could indicate the relatively high share of plastics (e.g. a mono-fraction from industrial waste) in SRF I which contained Br based fire retardants. Pb content was in Hässleholm 2.7 g/kg, and it was high also in Norrköping 3.5 g/kg ash; it was also relatively high in Kauttua ESP ash, about 2 g/kg ash. In Kauttua the element contents were higher in ESP ashes than in filter ash, except Hg, Br and Cl. The As content was clearly highest in Kauttua ESP ash. Br content was highest in Kauttua filter ash (2 g/kg ash).

In the filter ashes, the added chemicals (lime and activated carbon) may have a diluting effect on the concentration of an element.

High contents of Cu, Co, Cr and even Ni in the bottom ashes indicated that these existed in metal form in the fuels.

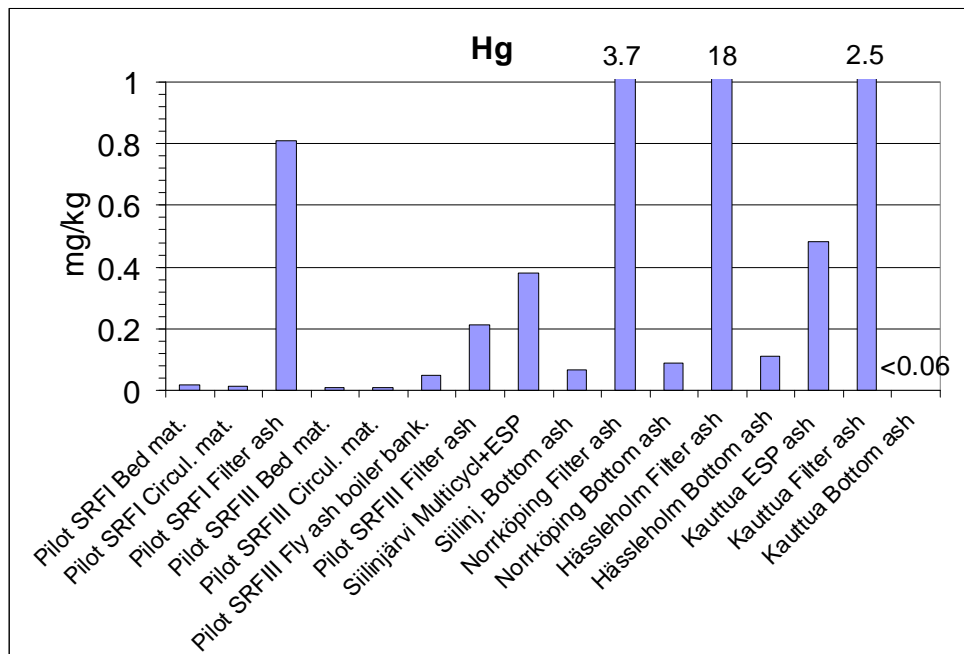


Figure 13. Mercury content in the ashes (< is below the limit).

6 PROCESS MEASUREMENTS

The process data from Kauttua and Norrköping were examined using the multivariate analytical tool Simca – P version 10.0.2.0. In Figure 14, an example of the statistical representation is shown of the Kauttua measurements. Accordingly, the processes operated relatively smoothly. Some of the points deviated from the average, for which the strongest reason was the fluctuation in the temperature and steam flow. This, however, had no effect on the results of the measurements.

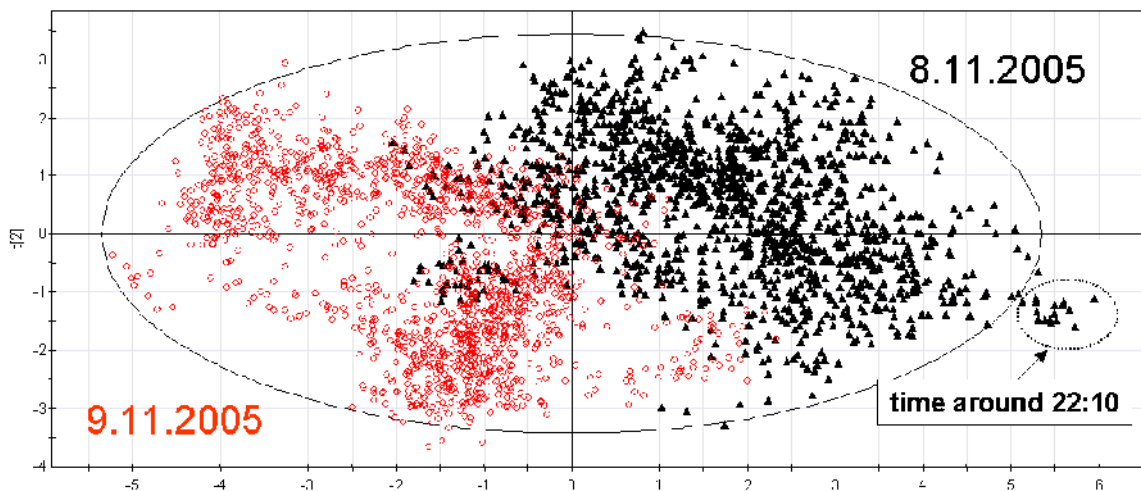


Figure 14. The score plot describing the process data of Kauttua (both boiler and flue gas) collected during the measurement campaign. The black color indicates the data collected during 8.11.2005 and the red one during 9.11.2005.

The combustion processes were running at normal performance values and was stable during the measurements. As an example bead temperatures during measurements at Norrköping are presented in Figure 15.

Process performance assessment of Hässleholm is based on daily plant reports and manually collected data from process control system. Due to the relatively warm weather conditions, 11-12 °C outside temperature, the plant was not operated at full load. Some additional cooling was used to maintain reasonable operation. The operation was, however, stable during the whole measurements campaign.

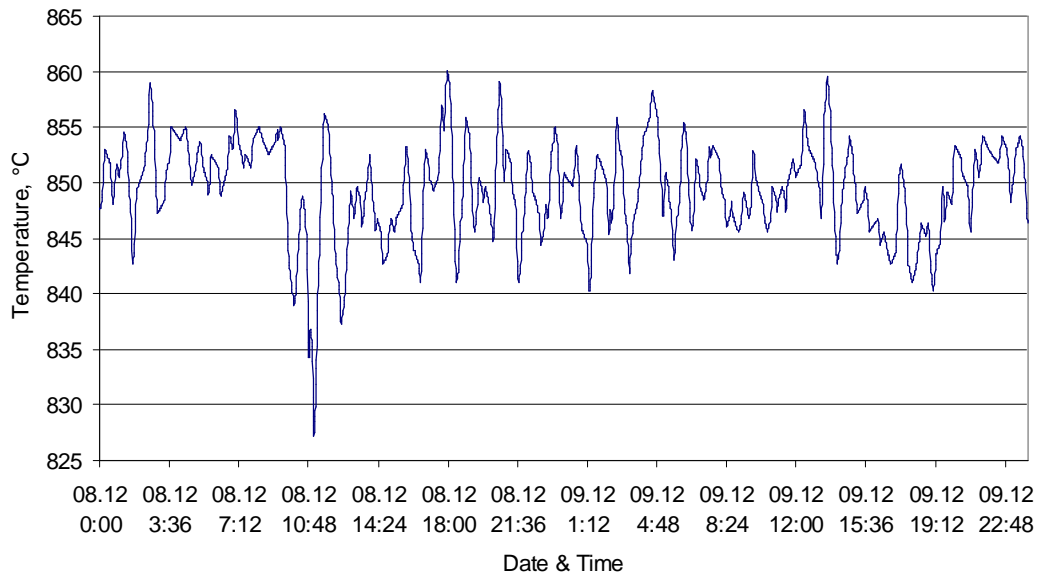


Figure 15. Variation of the bed temperature of the boiler during the measurements (Norrköping).

6.1 FLUE GAS MEASUREMENTS

Emissions of several gaseous components were measured with FTIR. Table 4 presents average gas concentrations during particle measurements. The pollution control systems in the waste combustion plants were quite effective in removing HCl and SO₂. The daily average WID limits of HCl emissions is 10 mg/m³ and SO₂ emissions 50 mg/m³. The emissions in all plants were below these limits. NO_x emissions are generally limited to either 200 or 400 mg/m³ depending on the plant nominal capacity.

Table 4. Average gas compositions during particle measurements.

	O ₂	CO ₂	H ₂ O	CO	SO ₂	HCl	NO _x
Filter inlet	[%]	[%]	[%]	[mg/Nm ³] dry 11% O ₂	[mg/Nm ³] dry 11% O ₂	[mg/Nm ³] dry 11% O ₂	[mg/Nm ³] dry 11% O ₂
Hässleholm	-	-	-	-	-	-	-
Norrköping	7	-	25	-	9	870	-
Kauttua	9	9	12	20	90	100	360
Siilinj Bio	-	-	-	-	-	-	-
Siilinj CCW	-	-	-	-	-	-	-
Filter outlet							
Hässleholm	7	11	18	10	3	6	90
Norrköping	7	10	20	7	0.1	4	40
Kauttua	9	9	10	20	30	1	340
Siilinj Bio	5	13	24	30	46	-	67
Siilinj CCW	5	13	21	15	34	-	220

Flue gas composition at Norrköping was monitored continuously with the plant instruments and checked both on the inlet and outlet side by means of VTT:s FTIR measurements. No significant differences were observed between these measurements. The HCl of the flue gas before and after the filter is shown in Figure 16. The HCl emission is well below the 10 mg/Nm³ WID threshold value. Total organic carbon TOC was both days < 1 mg/Nm³, WID threshold value is 10 mg/Nm³.

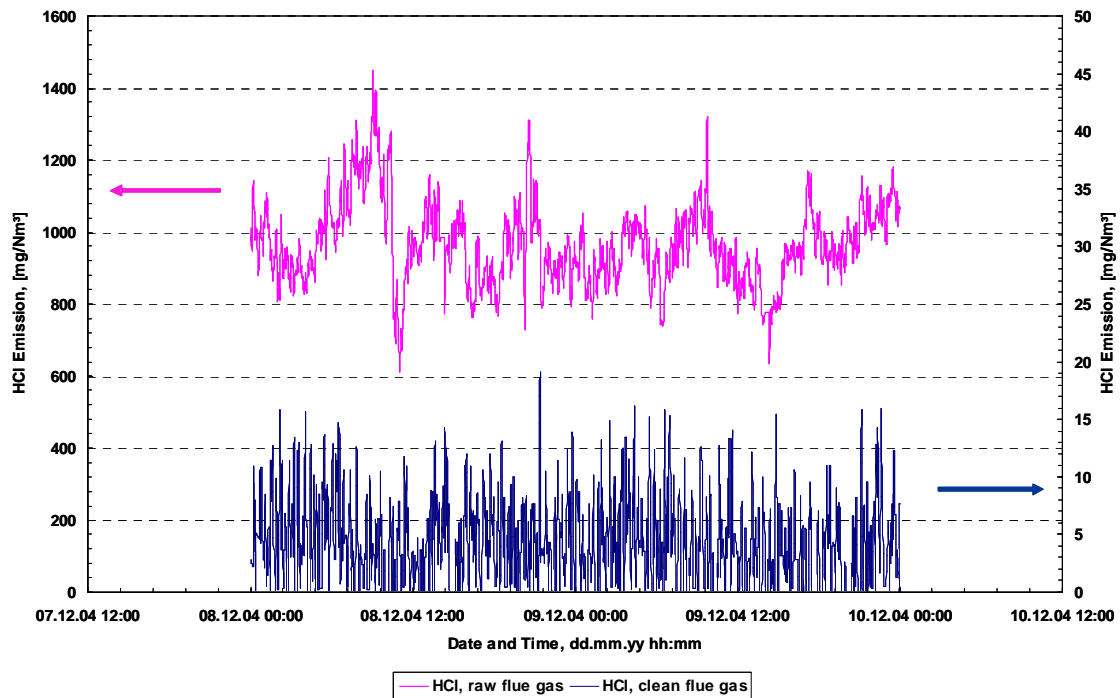


Figure 16. HCl content of the flue gas at Norrköping, process data (wet gas, ab. 4% O₂).

Various gas components were measured from filter outlet at Hässelholm. Emissions measured by VTT are well in line with the on-line plant emission measurements. As an example HCl and SO₂ emissions are presented in Figure 17.

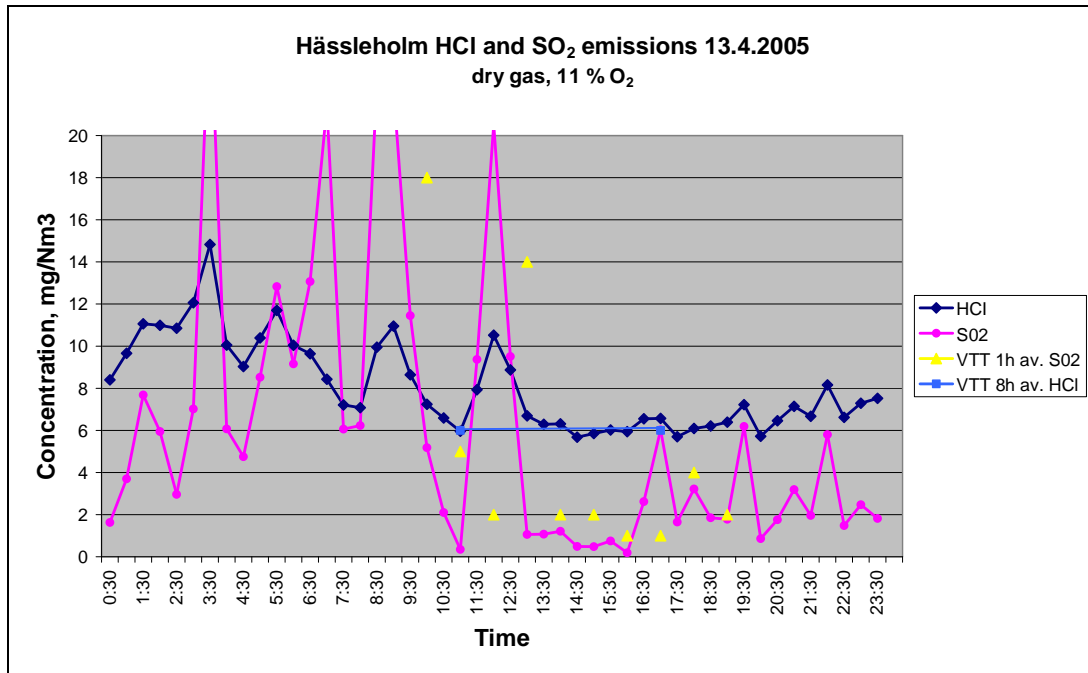


Figure 17. Hässelholm emission measurements 13.4.2006, including VTT measurements.

The HCl of the flue gas before and after the filter is shown in Figure 18, plant measurements. The yellow areas mark the time intervals of the VTT measurements. The HCl emission is well below the 10 mg/Nm³ WID threshold value. A significant increase of the HCl concentration in flue gas before the baghouse filter was observed during the first measurement day. No adequate explanation to this has been found by analyzing a restricted number of fuel samples. The most probable reason is a variation of the chlorine content of either the SRF or the peat/waste wood fuel fraction.

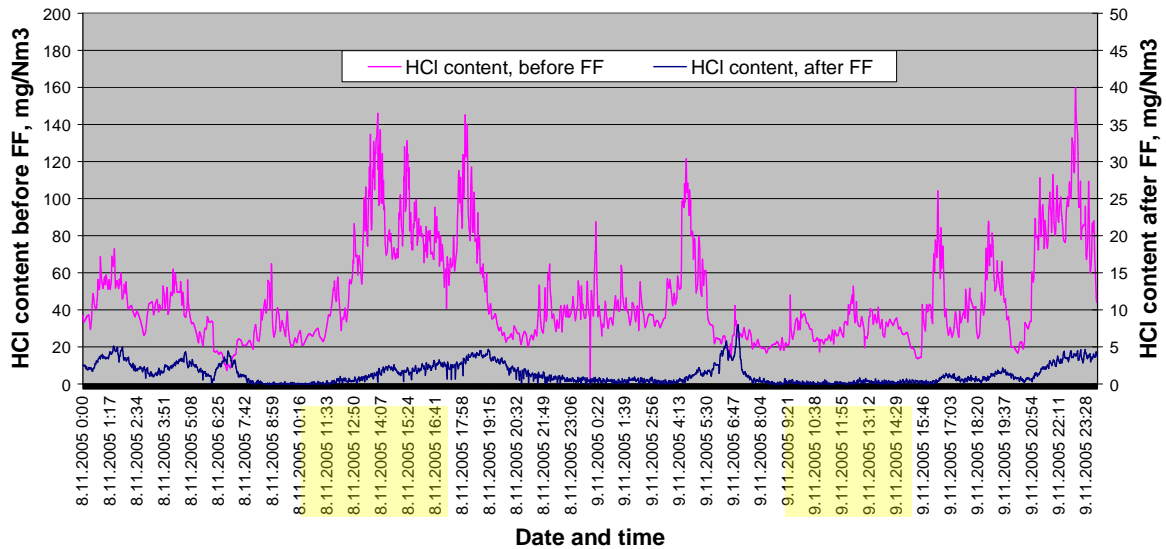


Figure 18. HCl content of the flue gas before and after the baghouse filter at Kauttua, process measurements. Reduced to 11% O₂.

6.2 RESULTS FROM KARHULA PILOT PLANT

The process data points were recorded every five seconds during the set points. This data has been used as the basis for mass and energy balance calculations, which give theoretical fuel mass flow, air flow and flue gas mass flow rates. These can be used to estimate the theoretical measurement error, which sums up uncertainties in fuel and ash analyses, and process measurements.

Table 5. Examples of measured gaseous emissions in different unit at Karhula pilot plant

Test	O ₂	CO	SO ₂ theoretical	SO ₂ measured	NO _x	HCl theoretical	HCl measured
	%, wet	ppm, wet	ppm, wet	ppm, wet	ppm, wet	ppm, wet	ppm, wet
10 CWW	6.83	41.8	31.4	40.5	135.4	34.5	26.9
20 REF I	6.77	733.0	50.7	54.1	113.5	210.3	116.6
30 REF III	7.28	227.3	71.6	34.4	117.7	733.8	383.4
40 REF I UM	8.23	380.8	56.3	41.9	99.6	387.9	231.4
45 REF I UM	7.36	421.5	87.9	33.9	70.2	530.0	213.9
50 REF III UM	6.21	294.7	31.5	19.1	95.1	315.5	143.6

The process data were examined using the multivariate analytical tool Simca – P version 10.0.2.0. With this tool all the data can be examined simultaneously. The PLS

modelling tool (Partial Least Square) was used to study the correlations between different parameters.

Compilation of setpoints

In Figure 19, all the set points are presented in the same graph. This graph was produced by Simca where the data table was examined row wise. Accordingly, the set points resembled well each other. The good resemblance can be deduced from the fact that the scattering (i.e. standard deviation) of the measured points (5 second interval) are mainly inside the elliptic circle which describes the statistical probability of 95 %.

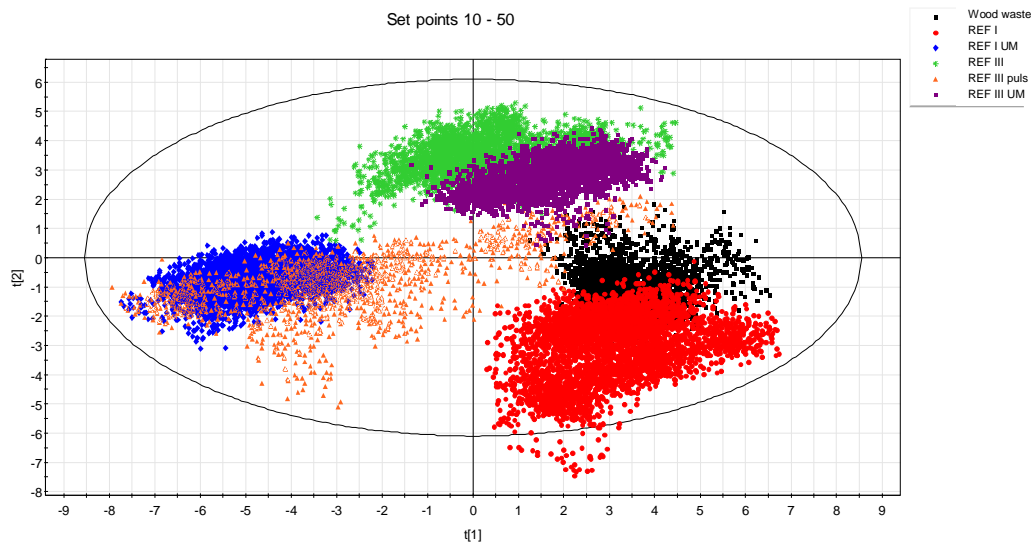


Figure 19. Compilation of the set points – the score plot describing the measured points in each set points presented by colors representing the fuels.

The feed rate was adjusted so that the bed temperature could be kept as even as possible and between 850 and 900 °C. Some fluctuation had to be accepted due to variations in the waste fuel quality and feeding properties.

Figure 20 presents the Hg measured for all the fuels. Accordingly the peak mercury contents were measured for SRF III 39 µg/m³. Typically, the gas components varied widely especially in the SRF set points. In the results, it was observed that the variation correlated well with the CO content measured in the furnace.

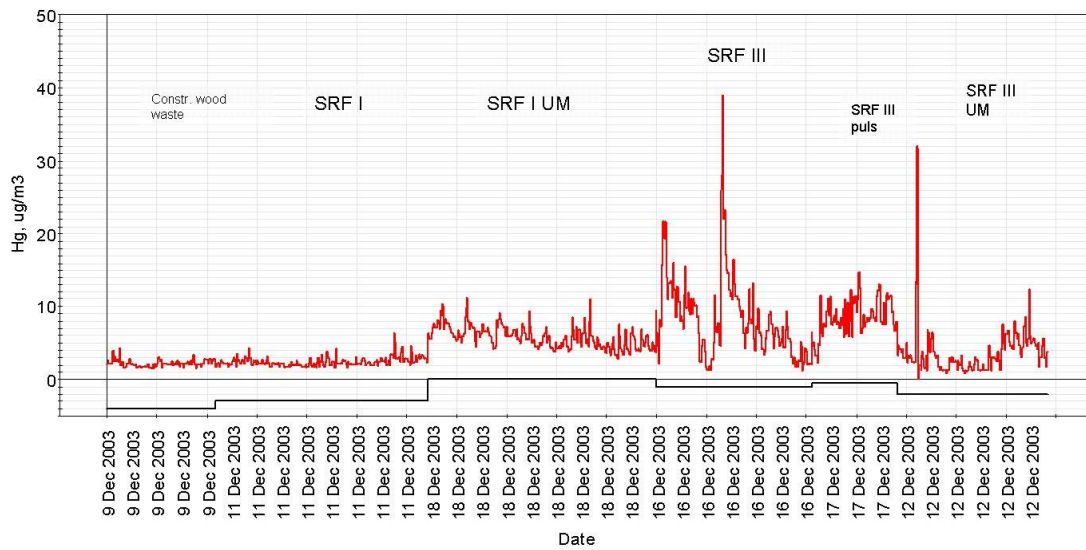


Figure 20. Mercury content measured in the flue gas after the filter in all the set points.

Fuel and ash analyses

An example of the variation in the analysis results of the single fuel samples is given in Figure 21. It can be seen that there was a quite high variation of chlorine and some metals like copper in the single fuel samples taken before the inlet particle sampling. There was even a high variation between the duplicate samples used in the analysis of the metals. In the relationship between the amount of an element fed into the furnace and the amount found in the fine particles a large variation was also found. The main reason for this variation is the inhomogeneity of waste based fuels. These results will be reported in the later stage when the leaching and electron microscopic fuel analysis are available.

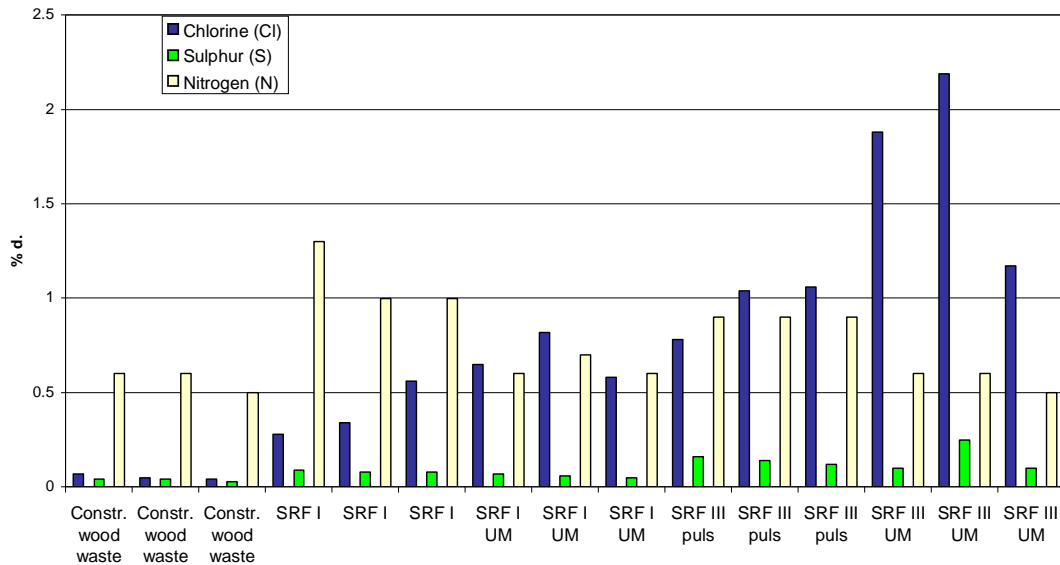


Figure 21. Variation of chlorine, sulphur and nitrogen content in the single samples

When the concentrations of the heavy metals and halogens are compared in the ash samples collected during the tests of SRF I and SRF III, the fly ash contained higher amounts of heavy metals and halogens than bed material (which contained fuel ash) and almost the same amounts than in the fuel ashes (calculated from the fuel analysis data). In the fly ashes, also halogen contents were in measurable amounts and striking was the high Cl content of 4.4 % in the fly ash of SRF III.

The laboratory studies of this project will support the elaboration of the reasons for the observations.

Lime injection into flue gas

Three set points were carried out with each waste fraction using three different limestone types: Sorbalite (containing 10 % activated coke), a commercial lime stone and a “research” limestone (Nordkalk), always in this order. Only gaseous emissions were measured during these set points.

The limestone feed was adjusted so that the HCl content of the flue gas was about or below 10 ppm. The SO₂ content usually dropped to about zero. The additives were fed by a small screw feeder placed on a balance into the flue gas line before the filter. Same feeding rate setting was used for all limestone qualities during the same set point. Between the set points the feeding was adjusted according to the procedure mentioned above.

The quantity of lime fed into the flue gases was calculated based on the weight of the additive batches added to the feeder. The feed rate varied quite substantially from set

point to set point, and also within each set point, due to the inaccurate feeding procedure. Therefore it was not possible to optimise the lime feeding with respect to emission control and no definite conclusions regarding lime quantities may be drawn.

When no additives were injected to the flue gases, the SO₂ emissions after the filter were in the range of 30-40 ppm except for SRF I UM, when the emission was about 15 ppm. Before the filter the measured SO₂ content was usually 30-35 % higher. Limestone addition to the bed did not significantly influence the SO₂ concentrations (less than 10 ppm). When lime was fed into the flue gases before the filter, the SO₂ content dropped to zero as presented in Figure 22.

The HCl was highest for the SRF III waste fraction. Comparing the chlorine content, of the SRF and SRF UM samples shows an almost doubling of the chlorine content of waste fractions which have gone through the washing (defibering) process. The washing process separated but the fibres also fines and heavy fractions like sand, stones, glass and metals. The results indicate that most of the chlorine in the waste is bound to the plastic fraction, probably in the form of PVC.

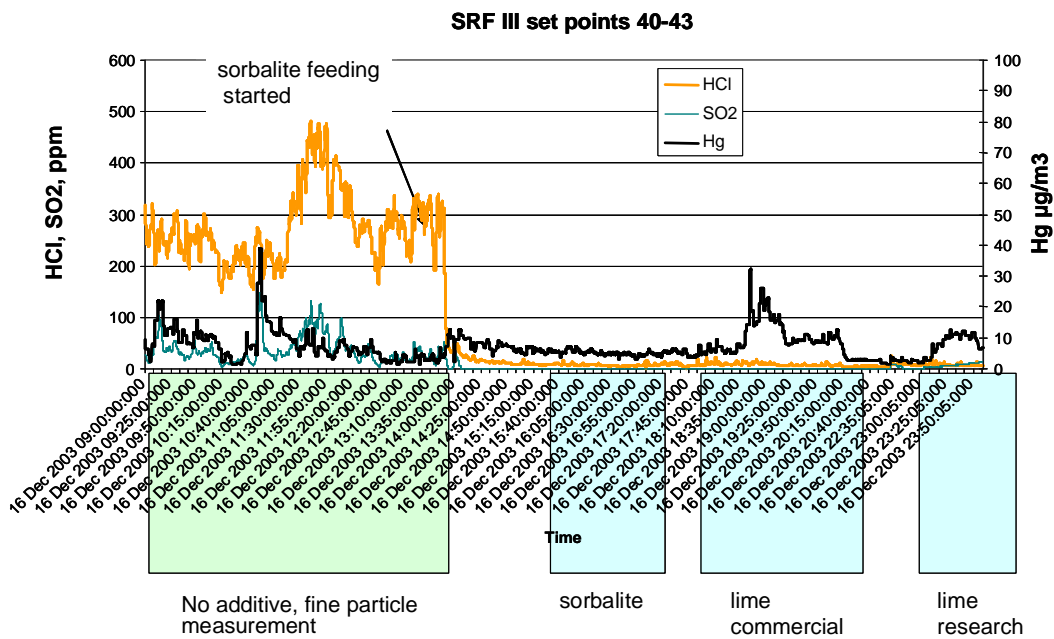


Figure 22. Hg, HCl and SO₂ content after filter measured SRF III set points with lime addition.

6.3 FINE PARTICLE CONCENTRATIONS AND SIZE DISTRIBUTIONS

6.3.1 Mass concentrations and mass size distributions

The particle mass concentration and mass size distribution were measured with BLPI from every plant and from the filter inlet and outlet. In some locations the mass concentration was also measured according to standard EN 13284-1:2001, their results are shown in Table 6 and Table 7 along with BLPI results.

Table 6. Filter inlet mass concentrations.

Filter Inlet	Mass concentration [mg/m ³] NTP, dry gas, 11% O ₂	PM 2.5 [mg/m ³] NTP, dry gas, 11% O ₂	PM 1 [mg/m ³] NTP, dry gas, 11% O ₂
Hässleholm, BLPI	1100 – 1600	1000 – 1400	1100 - 1500
Hässleholm, STD	-	-	-
Norrköping, BLPI	3800 – 5300* (10 300* – 14 800)**	430 – 490	250 - 310
Norrköping STD	4600	-	-
Kaattua BLPI	2400 - 4300	140 – 220	22 – 41
Kaattua STD	-	-	-
SRF 1, BLPI	3800	340	240
SRF 3, BLPI	3700	460	340
SRF 1 UM, BLPI	2200	210	140
SRF 3 UM, BLPI	3200	290	190

* Isokinesis overestimation corrected (Figure 23)

** Actual overestimated result due to bad isokinesis in sampling

Table 7. Filter outlet mass concentrations.

Filter Outlet	Mass concentration [mg/m ³] NTP, dry gas, 11% O ₂	PM 2.5 [mg/m ³] NTP, dry gas, 11% O ₂	PM 1 [mg/m ³] NTP, dry gas, 11% O ₂
Hässleholm, BLPI	0.13	<0.1	<0.1
Hässleholm, STD EN 13284-1:2001	0.2 – 0.4	-	-
Norrköping, BLPI	0.6	0.04, 0.08	0.03, 0.06
Norrköping STD	0.5 - 0.7	-	-
Kaattua BLPI	0.3	0.04	0.03
Kaattua STD	0.2 – 0.4	-	-
SRF 1, BLPI	0.1	0.03	0.04
SRF 3, BLPI	0.1	0.05	0.05
SRF 1 UM, BLPI	0.1	0.02	0.03
SRF 3 UM, BLPI	0.1	0.03	0.04

The two combustion techniques produced very different particle mass size distributions. The grate boiler produced almost solely fine particles $0.1\mu\text{m} < D_p < 2\mu\text{m}$, while from the fluidised bed the coarse particle mode is dominant, as seen in Figure 23. The rectangle is a histogram presentation of the pre-cyclone collected particles, and the breakline presents the mass size distribution measured with BLPI. The total mass concentration, shown in Table 6, is the sum of the surface area under these two lines. The pre-cyclone had slightly different cut-off diameters at different locations.

The pilot scale studies showed that the generated fine particle concentration depended on the fuel. The fine particle concentration was the highest with SRF 3 and it was reduced by the urban mill (UM) treatment of the fuel.

The filters in each plant were highly efficient, the fine particle concentrations at filter outlet were well below 0.1 mg/m³. Due to low concentrations extremely small particle masses were collected on the substrates and the measuring accuracy remained poor, approximately $\pm 100\%$ on each stage. For the same reason the weighed masses were below the limit of identification for the data unavailable in Figure 24. At the filter outlet the majority of the particle mass concentration was in the coarse mode and no actual fine particle mode could be detected, regardless of the combustion technique or fuel.

In Norrköping filter inlet BLPI measurements the stack flow velocity was higher than the velocity in the sampling nozzle. Therefore the measured mass concentration was too high. The overestimation can be calculated from /1/:

$$C/C_0 = 1 + (U_0/U - 1) (1 - 1 / (1 + (2 + 0.62 U/U_0)Stk)) \quad (1)$$

where C is the measured mass concentration, C_0 the real concentration, U the gas velocity in the nozzle and U_0 the flow velocity in the duct. Stk is the Stokes number of the particle, which is related to the size of the particle. The mass concentration overestimation in function of the particle size in Norrköping filter inlet measurements is presented in Figure 25. There is no correction needed in the fine particle mode, but the coarse particle concentration is overestimated up to 3.6 times too high. Norrköping mass size distributions presented in Figure 23 have been corrected according to (1).

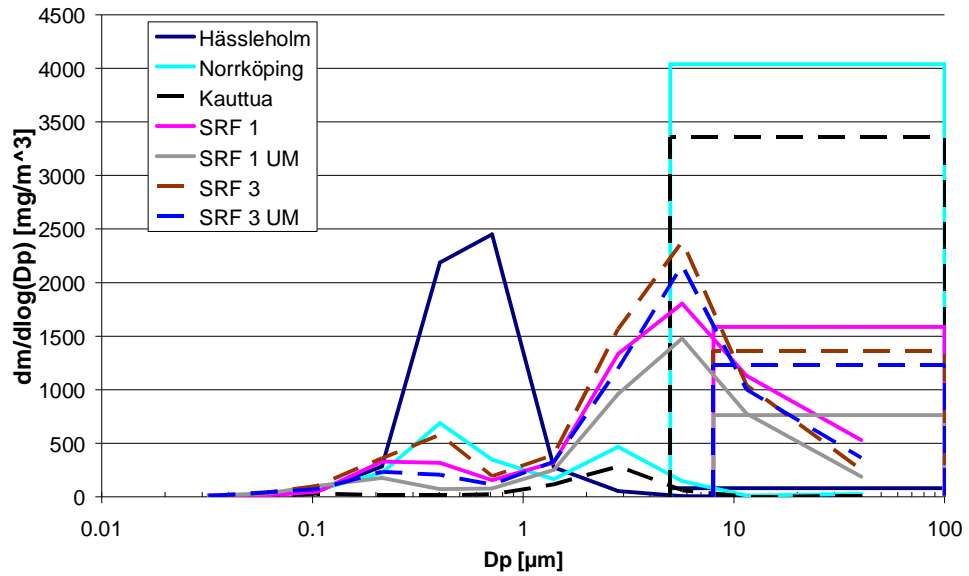


Figure 23. Mass size distributions at filter inlet. The rectangle at right is a histogram presentation of the pre-cyclone collected particles, and the break line presents the mass size distribution measured with BLPI. The total mass concentration is the sum of the surface area under these two lines.

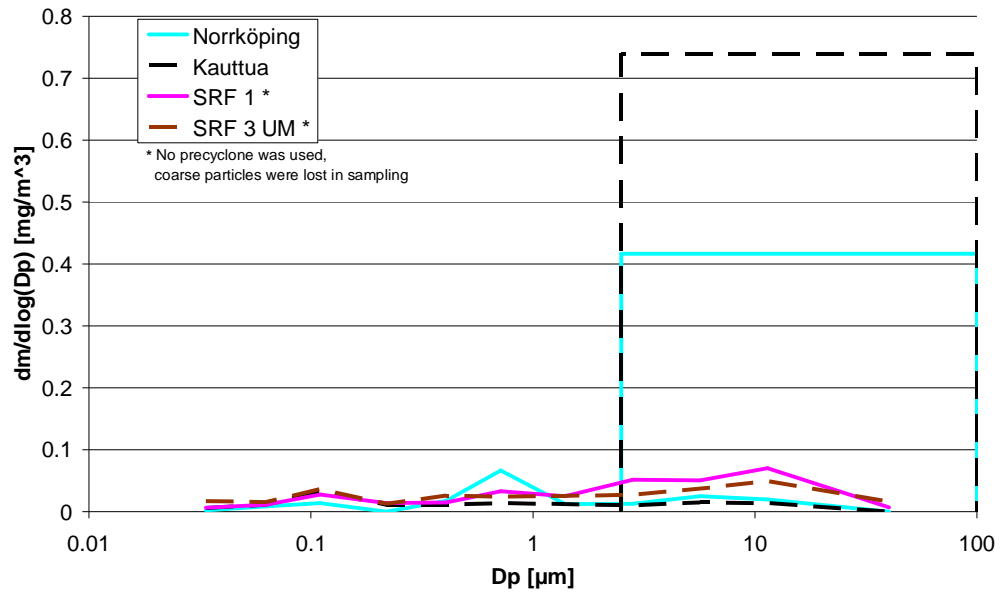


Figure 24. Mass size distributions at filter outlet. No pre-cyclone was used in the pilot plant measurements, coarse particles were lost in the sampling.

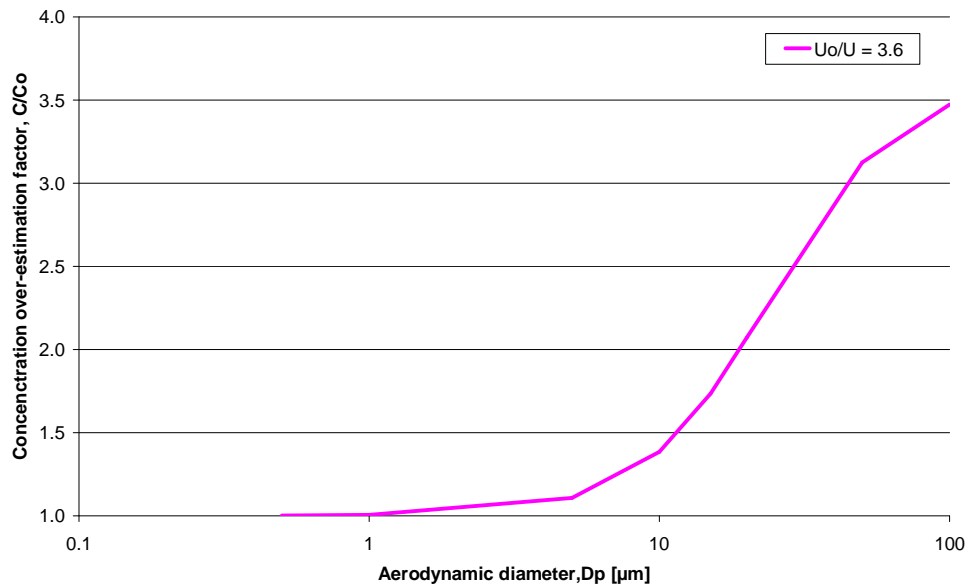


Figure 25. Concentration overestimation in function of particle size in Norrköping BLPI measurements from the filter inlet because of the bad isokinetic sampling, calculated from (1).

6.3.2 Number size distribution

The number size distribution was only measured at Norrköping. Figure 26 presents the number size distribution at the filter inlet and outlet during normal boiler operation and at the outlet during sootblowing. At the filter outlet the number concentration was very small, only ~ 20000 particles/cm³. When the sample was heavily diluted, the signal/noise ratio was low.

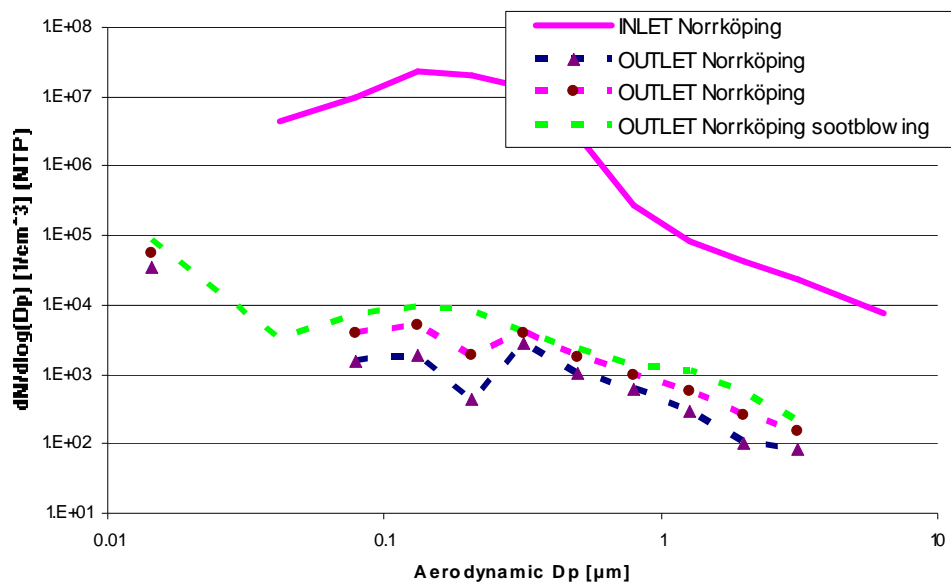


Figure 26. Number size distributions at Norrköping at the filter inlet and outlet during normal operation and at the outlet during sootblowing.

6.3.3 Elemental composition of particles

The elements regulated by EU directive and other selected elements were analyzed from particles with ICP-MS and IC. The analysis was made from BLPI collected particles to acquire the element mass concentrations and size distributions. In some locations the element mass concentration measurements were made also with a standardized method EN 14385.

The EU waste incineration threshold (WID) for emissions of Cd+Tl is $50 \mu\text{g}/\text{m}^3$ and As+Co+Cr+Cu+Mn+Ni+Pb+Sb+V $500 \mu\text{g}/\text{m}^3$. The studied emissions were significantly lower than the WID. The emissions of these elements presented in Table 8.

Some element concentrations were below the limit of identification, in which case the identification limit is added to emissions, but ‘<’ is added.

With the existing effective filters the WID limits are easily achieved, and they make the limits seem unnecessary high.

Table 8. Concentrations of WID limited elements in particle emissions.

	Particle emissions [µg/m ³]	Cd, Tl [µg/m ³]	As,Co,Cr,Cu,Mn, Ni,Pb,Sb,V [µg/m ³]	Hg Particle/gas phase [µg/m ³]
WID threshold	10 000	50	500	50
Hässleholm BLPI	100	0.001	9*	-
Hässleholm EN 14385	300	0.02	<2.2	<0.01 / 0.05
Norrköping BLPI	600	0.01	32*	-
Norrköping EN 14385	600	<1.5	<17	1.5 / 4.9
Kaattua BLPI	300	0.1	19*	-
Kaattua EN 14385	300	<0.006	<0.8	<0.004 / <0.04
Hässleholm PM1, BLPI	<100	0.02	1*	
Norrköping PM1, BLPI	50	0.005	2*	
Kaattua PM1, BLPI	30	0.001	4*	-

* Includes some Cr contamination from BLPI

The mass size distribution of manganese at filter inlets is presented in Figure 27 at filter outlets in Figure 28. Similarly the mass size distribution of lead at filter inlets is presented in Figure 29 and at filter outlets in Figure 30. Mn is concentrated mainly in coarse particles $D_p > 1\mu\text{m}$, while Pb is mainly in fine particles, $D_p < 1\mu\text{m}$. The relative abundance of a compound in the fine particles indicates that the element was readily released from the fuel to the gas phase, and that the gas-to-particle conversion mainly occurred by vapor condensation mechanism. The occurrence in the coarse mode particles indicates that the element was not released from the fuel to the gas phase during combustion, or that if it was released, the gas phase compounds reacted with the coarse mode particles by chemical surface reaction. Cl, Cd, Cu, Pb and Tl are typically volatile and thus abundant in the fine particles, while for example As, Co, Mn, Sb and V are usually almost entirely present in the coarse particle mode.

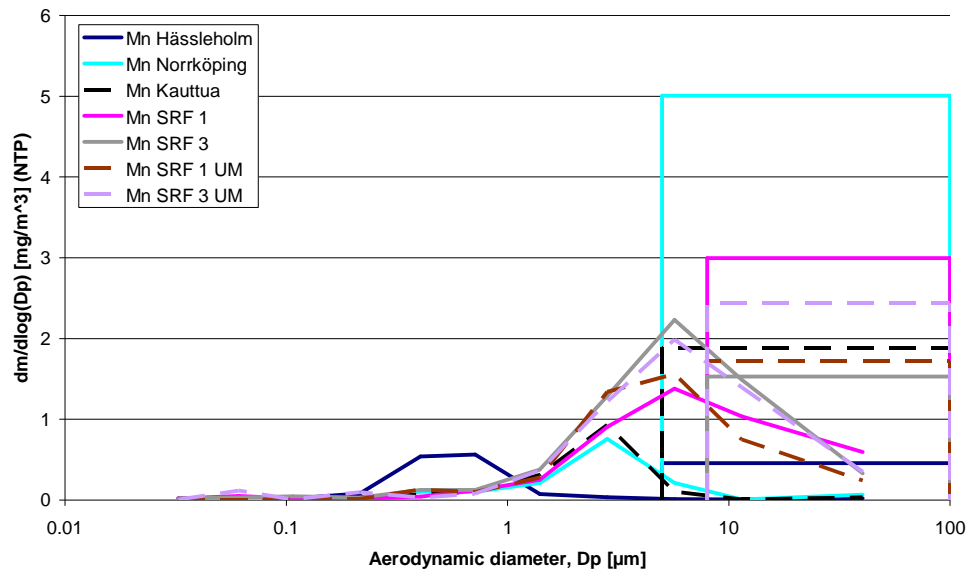


Figure 27. Manganese mass size distribution at filter inlet.

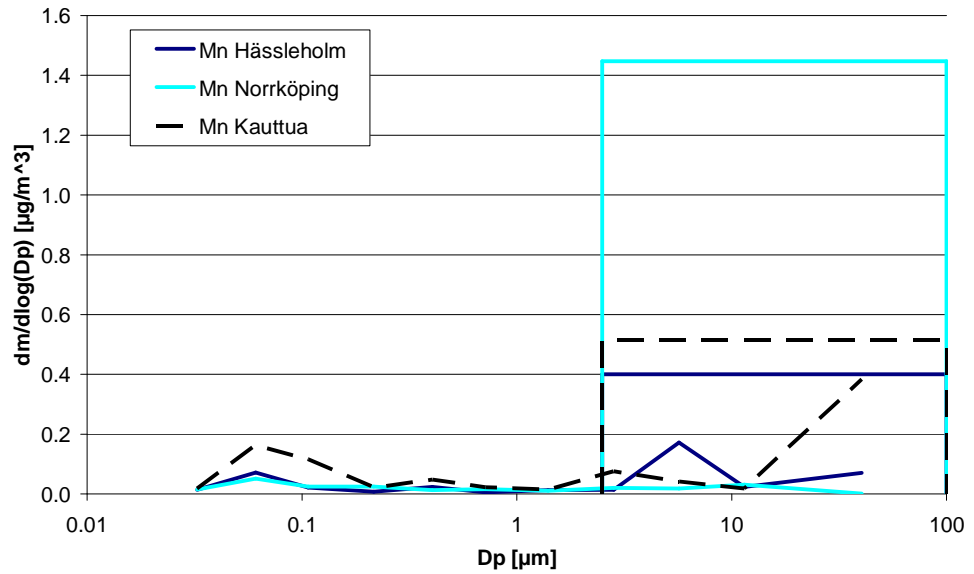


Figure 28. Manganese mass size distribution at filter outlet

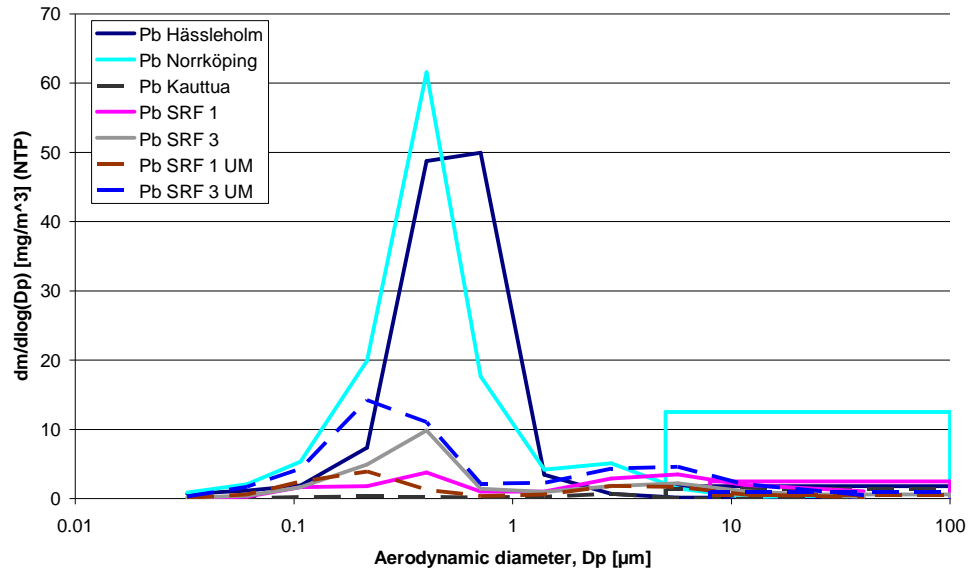


Figure 29. Lead mass size distribution at filter inlet.

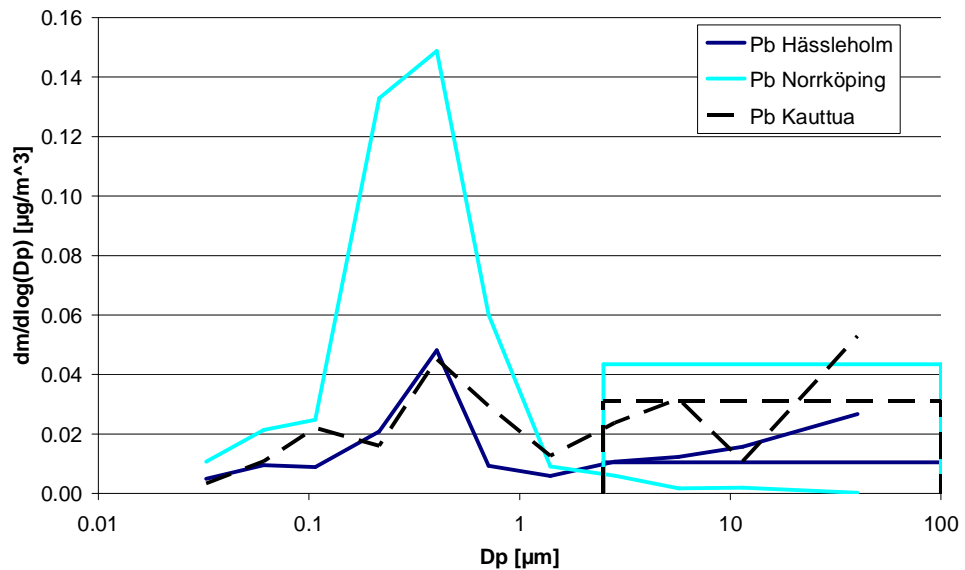


Figure 30. Lead mass size distribution at filter outlet

The pilot scale experiments indicated that the fuel only had a minor effect on the relative abundance of the elements in the fine or coarse mode particles, i.e. certain elements were found mainly in the fine or coarse mode particles for all the fuels. However, for the elements present in the fine particle mode, trends between the element concentrations in the fuel and in the fine particle mode could be found. The concentration of a certain element in fine particle mode depended on both the element and Cl concentrations in the fuel.

On the other hand the combustion technique was an important factor in the fine particle formation. In grate boilers the maximum temperatures are typically higher than in fluidised beds, which causes more elements to be more volatile. This leads to larger fine particle fraction in grate boilers. Also in fluidised bed boilers the volatile elements are found in large concentrations from the coarse particles as the vapours interact with the bed material and other coarse particles reducing the fine particle concentration. For example manganese requires higher temperature than lead or copper to significantly evaporate and is generally found solely from the coarse particle mode in the fluidised beds. However in the Hässleholm grate boiler Mn was found also in the fine particle mode.

The pilot scale studies showed that the waste quality has a significant effect on the fine particle concentration and composition. SRF 3 UM and SRF 3 fuels consisted more chlorine and volatile trace elements leading to larger fine particle concentrations

The combustion of mixed fuels, coke, peat, construction wood waste and REF in Kauttua produced significantly low concentration of fine particles. The chlorine concentration in the Kauttua fuels was low and sulphur concentration high, which result to lower evaporation. Peat consists large amounts of silicon and also aluminium, which react with the volatile trace metals further reducing the fine particle concentration. Silicon and aluminium form insoluble compounds with the trace metals as opposed to soluble metal chloride compounds. In disposal of the filter or bottom ashes the solubility is a cost factor.

The element distribution at filter outlet was very different from that of filter inlet. While the main components, of the analyzed elements, at the inlet were Cl, K, Na, SO₄, Pb, Cu and Zn, the main components at the filter outlet were Fe, Al and Cr. It was detected with electron microscopy that after fabric filters there were coarse iron oxide particles, which may be fragments of the duct walls or other structures located after the filter. With such a small particle mass concentration it is significantly increased by few collected rust fragments.

6.3.4 Efficiency of air pollution control systems

The particle penetration through the fabric filter, P , is:

$$P = 100 - \text{Collection Efficiency (\%)} = 100 * (\text{outlet mass concentration} / \text{inlet mass concentration})$$

The penetration of particles through filters was calculated from simultaneous filter inlet and outlet BLPI measurements. The penetration in function of particle size is presented in Figure 31. The fabric filters removed the fine particles so effectively, that very small masses were collected and the weighing accuracy of the filter outlet BLPI substrates remained poor. Therefore the error in the penetration for individual data points is approximately $\pm 100\%$. The efficiencies of the fabric filters designed for the waste combustion were significantly better than ESPs or two other FFs in a biomass and a coal

plant. Fabric filters don't have similar penetration window that ESPs have between particle size $0.1\mu\text{m}$ and $1\mu\text{m}$, but the efficiency seems to slightly worsen towards the finer particles.

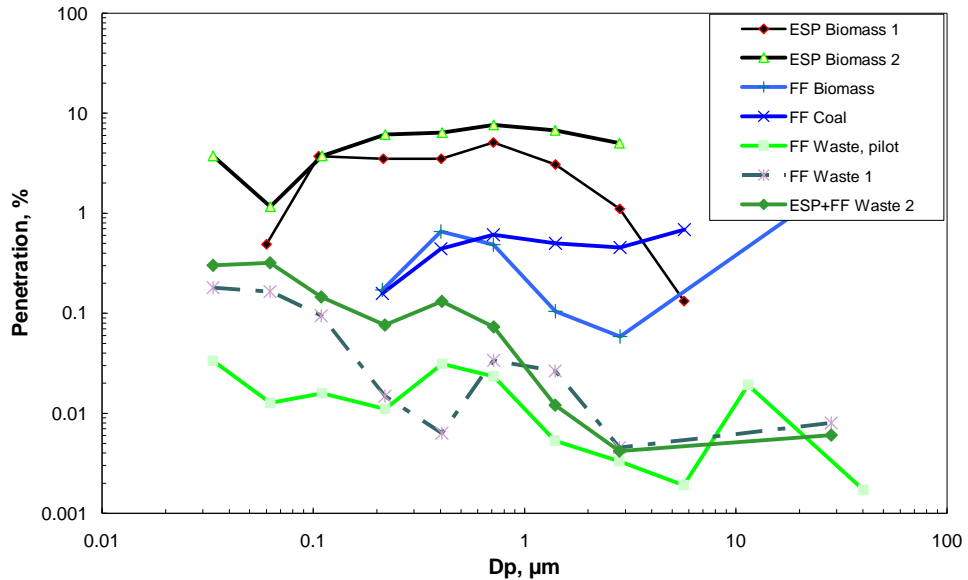


Figure 31. Filter penetration of particles in the plants measured in this project and a previous project (/2/ Hokkinen 2004)

BLPI is not capable of collecting the nano sized particles $D_p < \sim 20\mu\text{m}$. Electron microscopy revealed that particles in this size range would exist. The penetration of these nano particles through fabric filters would require further studying with devices capable of detecting them.

6.3.5 Construction Wood Waste Combustion

Construction wood waste was burned in three locations, Pilot scale plant, Siilinjärvi (70% wood and 30% wood waste) and $\sim 42\%$ mixed with other fuels (peat, coke, SRF) in Kauttua.

Compared to 'clean' bark+sawdust fuel the construction waste wood fuel, even when mixed with other fuels, consisted large quantities of Arsenic, Cadmium, Chromium, Copper, Nickel, Lead, Antimony, Tin, Vanadium and Zinc. Bark contains relatively high concentration of Manganese, mixing wood waste to bark+sawdust decreased the fuel Mn concentration because the bark proportion decreased. The fuel concentrations are presented in Table 9.

The Arsenic concentration in all three waste woods, especially in Kauttua, was remarkably high. The active ingredients in wood impregnating agents are copper, chromium and/or arsenic based. The use of arsenic has been restricted, but it is not forbidden. It should be possible to reduce Arsenic concentration in fuel by careful selection of the wood wastes. Chromium and Copper concentrations were not high in

wood wastes compared to the other waste fuels. Copper and chromium based impregnating agents do not have any restrictions.

Table 9. Characteristics of the construction wood wastes and bark+sawdust

		Siilinjärvi bark+sawdust	Siilinjärvi mixed wood waste	Pilot wood waste	Kaattua mixed wood waste
		100% bark+sawdust	70% bark+sawdust		30% peat 19% SRF, 9% coke
		0% wood waste	30% wood waste	100% wood waste	42% wood waste
Mercury (Hg)	mg/kg d	<0.04	<0.04	0.03	0.04
Arsenic (As)	mg/kg d	<0.04	6.7	14.2	74
Cadmium (Cd)	mg/kg d	0.1	0.26	0.54	0.4
Cobalt (Co)	mg/kg d	1.6	0.89	0.93	1.7
Chromium (Cr)	mg/kg d	1.0	15	29.4	84
Copper (Cu)	mg/kg d	4.5	8.5	31.5	69
Manganese (Mn)	mg/kg d	276	180	85.3	84
Nickel (Ni)	mg/kg d	1.0	3.1	4.7	7.5
Lead (Pb)	mg/kg d	1.1	14	46.3	77
Antimony (Sb)	mg/kg d	0.02	5.5	1.4	1.6
Tin (Sn)	mg/kg d	0.08	0.36	1.6	3.7
Thallium (Tl)	mg/kg d	0.06	0.04	0.02	<1
Vanadium (V)	mg/kg d	<0.1	0.66	1.5	5.3
Zinc (Zn)	mg/kg d	63	142	198	300
Bromine (Br)	mg/kg d			<50	
Fluorine (F)	mg/kg d			<50	<50
Chlorine (Cl)	% d	0.12	0.44	0.1	0.17
Soluble Na+K	% d				0.07
Sulphur (S)	% d	0.01	0.02	0.04	0.16
Nitrogen (N)	% d	0.2	0.50	0.57	0.8
Carbon (C)	% d	51	50	49.0	48.5
Hydrogen (H)	% d	6.0	6.1	5.9	5.6
Volatile matter	% d	79	78	78.7	73.6
Ash	% d	1.7	2.9	3.4	6.8
High heating value	MJ/kg d	20.55	19.99	19.66	19.8
Lower heating value	MJ/kg d	19.24	18.66		11.0

In Siilinjärvi grate boiler the addition of 30% waste wood to bark+sawdust increased the concentration of several elements in particles to 2 to 100 fold, including As, Cd, Cl, Cr, Cu, Na, Pb, Sb and Zn. The concentrations of these elements were also higher in the waste wood containing fuel. All of these elements were also volatile in boiler and were therefore enriched in the fine particle mode. However, K was the dominating element in the fine particle mode and the total mass concentration of the fine particles was higher with bark and sawdust only. Pb mass size distribution at Sillinjärvi filter inlet is presented in Figure 32 with and without 30% construction wood waste addition to bark+sawdust.

As seen in Table 10 the WID regulated heavy metal emissions were exceeded only while burning clean bark+sawdust when Mn emission alone was higher than the limit for As+Co+Cr+Cu+Mn+Ni+Pb+Sb+V. The waste incineration directive does not concern clean wood combustion however.

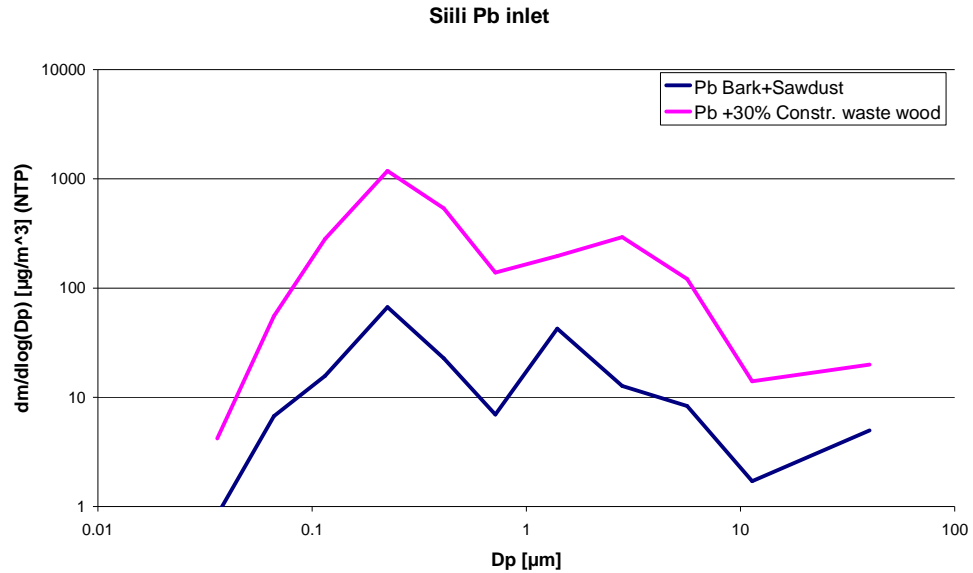


Figure 32. Pb mass size distribution at Siilinjärvi filter inlet with and without 30% construction wood waste addition to bark+sawdust

Table 10. BLPI particle mass concentrations from construction wood waste combustion

	Siilinjärvi bark+sawdust	Siilinjärvi mixed wood waste	Pilot wood waste	Kauttua mixed wood waste
	100% bark+sawdust 0% wood waste	70% bark+sawdust 30% wood waste	100% wood waste	30% peat, 19% SRF, 9% coke 42% wood waste
	ESP	ESP	FF	ESP+FF
	[mg/m ³] (NTP) 11% O ₂ , dry	[mg/m ³] (NTP) 11% O ₂ , dry	[mg/m ³] (NTP) 11% O ₂ , dry	[mg/m ³] (NTP) 11% O ₂ , dry
Filter inlet, total	280-800	230-2300	1600-3000	2400-4300
PM1	140-160	80-110	220-360	140-220
PM2.5	120-130	70-90	120-250	22-41
Filter outlet, total	60	20-130	0.07*	0.2-0.4
PM2.5	8	6-8	0.03	0.04
PM1	7	5-7	0.03	0.03
Cd+Tl	0.002	0.002	NA	0.0001
As+Co+Cr+Cu+Mn+Ni+Pb+Sb+V	1.2*	0.3*	NA	0.019*

- ~PM10, no pre-cutter cyclone could be used

** Includes some Cr contamination from measuring equipment

7 SUMMARY AND CONCLUSIONS

The pollution control systems used in the waste combustion plants were very efficient reducing the particle mass concentration approximately 99.99%. The operation of the filters is the most important factor in the particle emissions. Within the limits of measuring accuracy the mass size distributions after filters were similar and no dependence on fuel or combustion system could be detected. Some of the coarsest particles were oxidised iron pieces which are probably fragments of the duct structures, fans or filter etc. A small number of these pieces contain the majority of the emission mass concentration.

The waste quality and combustion method had significant effects on generated fine particle concentrations. Pilot studies showed that decreasing waste quality resulted in higher PM_{1.0} concentrations. In grate boiler the maximum temperature may be several hundreds of degrees higher than in fluidised bed boiler. Therefore in grate boiler volatile elements will evaporate more and also more elements will be volatile. In fluidised bed the vapours may interact with the bed material thus further reducing the fine particle concentration.

The waste fuels may contain large amounts of copper, lead and other trace metals, which concentrate in the fine particle mode as chlorides. The main components of the fine particle mode are potassium and sodium chlorides and sulphates.

The combustion of mixed fuels, coke, peat, construction wood waste and REF in Kauttua produced significantly low concentration of fine particles. The chlorine concentration in the Kauttua fuels was low and sulphur concentration high, which result to lower evaporation. Peat and coke consist large amounts of silicon and aluminium, which react with the volatile trace metals further reducing the fine particle concentration. Silicon and aluminium form insoluble compounds with the trace metals as opposed to soluble metal-chloride compounds. In disposal of the filter or bottom ashes the solubility is a cost factor and chlorides corrode the boiler structures. It could be possible to reduce the concentration of soluble metal-chloride fly ash fine particles by adding suitable additives to fuel when burning solely waste.

With the highly effective pollution control systems the particulate emissions from waste combustion are extremely low. The acceptable level for ambient outdoor PM₁₀ particle mass concentration is 50 µg/m³ as daily average according to EU directive. In spring 2006 the worst days PM₁₀ concentration at downtown Helsinki reached 150 µg/m³ /3/ because of forest- and wildfires south of Finland and dust from street sanding. PM₁₀ emissions were not measured from the waste combustion plants, but comparison to PM_{2.5}, which was 20-80 µg/m³, indicates that the particle emissions from waste combustion may almost equal the already existing Helsinki city air particle concentration.

Emissions of the EU regulated trace metals Hg, Cd+Tl and As+Co+Cr+Cu+Mn+Ni+Pb+Sb+V were all well below the WID limits. After the fabric filters the emissions were minimum of 10 times lower than the WID limits. Other

special emissions regulated by WID, like dioxins and furans, Hg, HCl, HF and TOC were all measured well below the threshold values. A summary of average measurement values is presented in Table 11.

Table 11. Average WID related measurements at the waste combustion plants.

Emission	Hässleholm (grate) mg/m ³ (11% O ₂)	Norrköping (CFB) mg/m ³ (11% O ₂)	Kaattua (CFB) mg/m ³ (11% O ₂)	WID threshold mg/m ³ (11% O ₂)
Total dust	0,45	0,58	0,27	10
Sb+As+Pb+Cr+Co +Cu+Mn+Ni+V	0,03	0,02	0,02	0,5
Cd+Tl	0,0005	0,002	0,001	0,05
Hg	0,00008	0,003	< 0,04	0,05
HCl	9,1	3,6	0,8	10
HF	< 0,032	< 0,03	< 0,012	1
Dioxins and furans	0,0048 ng/m ³	0,0076 ng/m ³	< 0,00001 ng/m ³	0,1 ng/m ³

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3. YTV homepage www.ytv.fi/eng/airquality,

**Appendix D Buonanno et al (2012) Ultrafine
particle emission from incinerators: The role of the
fabric filter**

Ultrafine particle emission from incinerators: The role of the fabric filter

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Incinerators are claimed to be responsible of particle and gaseous emissions: to this purpose Best Available Techniques (BAT) are used in the flue-gas treatment sections leading to pollutant emission lower than established threshold limit values. As regard particle emission, only a mass-based threshold limit is required by the regulatory authorities. However, in the last years the attention of medical experts moved from coarse and fine particles towards ultrafine particles (UFPs; diameter less than 0.1 μm), mainly emitted by combustion processes. According to toxicological and epidemiological studies, ultrafine particles could represent a risk for health and environment. Therefore, it is necessary to quantify particle emissions from incinerators also to perform an exposure assessment for the human populations living in their surrounding areas.

A further topic to be stressed in the UFP emission from incinerators is the particle filtration efficiency as function of different flue-gas treatment sections. In fact, it could be somehow important to know which particle filtration method is able to assure high abatement efficiency also in terms of UFPs. To this purpose, in the present work experimental results in terms of ultrafine particle emissions from several incineration plants are reported. Experimental campaigns were carried out in the period 2007–2010 by measuring UFP number distributions and total concentrations at the stack of five plants through condensation particle counters and mobility particle sizer spectrometers. Average total particle number concentrations ranging from 0.4×10^3 to 6.0×10^3 particles cm^{-3} were measured at the stack of the analyzed plants. Further experimental campaigns were performed to characterize particle levels before the fabric filters in two of the analyzed plants in order to deepen their particle reduction effect; particle concentrations higher than 1×10^7 particles cm^{-3} were measured, leading to filtration efficiency greater than 99.99%.

Implications: The main implication of the study is that the use of a fabric filter in the flue-gas treatment section of incinerators is able to guarantee very low concentrations at the stack in terms of UFPs. As regards the incineration plants, a further implication of the proposed study is that an a priori negative social response seems to be unjustified when referred to the ultrafine particle emissions.

Introduction

Particulate emission from anthropogenic sources is nowadays a relevant topic under examination of air quality and medical experts. A number of epidemiological studies were carried out to evaluate the correlation between the particulate matter (PM) and its negative health effects such as cardiovascular and breathing problems (Kreyling et al., 2006; Pope and Dockery, 2006). Moreover, toxicologists attempted to evaluate the particle properties mainly responsible of such negative effects in terms of both size and chemical composition. About the chemical composition, no definitive results were carried out to assess if particle toxicity is mainly due to the organic compounds surrounding the particles (Eiguren-Fernandez et al., 2010) or the soot core itself (Soto et al., 2008). As regard particle size, even if not unanimous results were reached, the interest of such experts is moving from mass-based concentrations (Loomis, 2000; Pope, 2000) (particulate matter with aerodynamic diameters lower than 10 PM_{10} and 2.5 μm , $\text{PM}_{2.5}$), towards submicrometer and ultrafine particles (UFPs;

i.e. particles sized about 100 nm in diameter or less as defined by the International Organization for Standardization through the ISO/TR 27628:2007), which mainly contribute to surface area and number concentrations (Hauser et al., 2001; Giechaskiel et al., 2009). Anyway, the air quality regulation is still referred to threshold limit values based on gravimetric time-integrated measurement of PM_{10} , and only proposed guideline values for $\text{PM}_{2.5}$ (U.S. Environmental Protection Agency (EPA), 1997; European Committee for Standardization, 2001; European Committee for Standardization, 2005; European Parliament and Council, 2008). To reach such air quality standards, threshold values were also introduced to regulate the emissions of anthropogenic sources as industrial plants and vehicular traffic (European Commission, 2008; European Parliament and Council, 2010). These anthropogenic activities are also recognized as the greatest emitters of UFPs (Cass et al., 2000; Morawska et al., 2008); at the moment, threshold limit values in terms of particle number concentrations are introduced for European diesel-fueled vehicles (European Commission, 2008)

and proposed for gasoline-fueled vehicles, whereas no regulating values are provided for industrial plants. Therefore, it is somehow important to characterize the emission sources as well as the evolution of particle size distribution in their proximity in order to carry out aerosol source apportionments and exposure assessment analyses in the areas characterized by high anthropogenic pressure (Buonanno et al., 2009b; Buonanno et al., 2010b).

In the waste management, incineration represents a favorable technique for reducing the waste volume and recovering its energy content to generate electricity and district heating. However, incinerators were subjected to a strong debate in Western countries about their emission of UFPs. Actually, as well as other industrial plants, only a mass-based threshold limit value is currently required in the operation of such plants as stated by the Directive 2010/75/EU (European Parliament and Council, 2010); in particular, total dust values (total amount of particle emitted in terms of mass) at the stack of the plant have to be lower than 10 and 30 mg m⁻³ on daily and half-hour basis, respectively. However, according to the scientific literature in terms of anthropogenic emissions (Airborne Particles Expert Group, 1999; Cass et al., 2000; EPA, 2000), incinerators are supposed to emit a very low amount of particles if compared to fossil fuel power plants and vehicle emissions since the Best Available Techniques (BAT) (European Commission, 2006) are used in the flue gas cleaning operations of modern plants. Actually, a few number of experimental campaigns focused on the evaluation of UFP emission from incinerators were carried out (Gomez-Moreno et al., 2003; Maghun et al., 2003; Buonanno et al., 2009a; Buonanno et al., 2010a; Buonanno et al., 2010b). Moreover, such analyses were often limited to a dimensional characterization of the aerosol emitted and/or to samplings at a downwind receptor site without evaluation of the performance of the flue-gas treatment sections. In our previous work (Buonanno et al., 2009a), measurements of fine and ultrafine particles at the stack of an incinerator were performed. However, in that case study, measurement methods not involving particle counting through condensation techniques and classification by means of particle electrical mobility methods were used.

In the present paper, an analysis of UFP emission levels from five plants is shown. In particular, four incineration plants burning wastes and a plant burning biomass were analyzed. Measurements

of total particle number and size distribution of aerosol particles were performed through a mobile system consisting of particle counters, mobility particle sizer spectrometers, thermodilution systems, as well as home-designed sampling systems. The main aim of the paper is to deepen the knowledge about fabric filter influence on ultrafine particle emission levels; therefore, aerosol particle measurements were also extended at a section located before the fabric filter for two of the incinerators under examination.

Experimental Analysis

Plant descriptions

Experimental campaigns involving aerosol particle characterization were performed in five plants showing different flue-gas treatment sections, four of which burning municipal waste or refuse-derived fuels (RDF) (Plants 1 to 4 in Table 1) and another one fed with biomass (Plant 5 in Table 1). The main purpose of these plants is the waste (or biomass) energy content recovery in a Rankine cycle power generation system. The five plants are made up of the following main sections: (i) the waste/biomass delivery area, where the delivery trucks arrive and dump the waste/biomass in a storage area (typically a bunker); (ii) the combustion and heat recovery section, consisting of a combustion chamber and an additional burner system to maintain the exhausts, for at least 2 sec, at the required minimum temperature (850 °C) (European Parliament and Council, 2010); (iii) the power generation section, constituted by a condensing turbine unit directly coupled to the generator; and (iv) the flue-gas treatment section. The exhaust treatment sections are different between the analyzed plants in terms of gas acid (dry, semidry, wet process), NO_x (selective noncatalytic reduction [SNCR], or selective catalytic reduction [SCR]), and dust removal (fabric filter, electrostatic precipitator, cyclones). In Table 1 a summary of the main characteristics of the combustion and flue-gas treatment sections of each plant is reported. Table 1 clearly shows that a fabric filter (with Polytetrafluoroethylene (PTFE) membrane) is present in each incineration plant analyzed, whereas it is not used in the biomass-fed Plant 5.

Table 1. Main characteristics of the combustion and flue-gas treatment sections of the analyzed plants

Plant	Furnace and waste characteristics	Flue-gas treatment section description
Plant 1	Grate type: moving grate Type of waste fed: refuse-derived fuel (RDF)	Semidry process made up of a SNCR, a spray absorber system (lime milk and powder activated carbons), and a fabric filter
Plant 2	Grate type: moving grate Type of waste fed: refuse-derived fuel (RDF)	Dry process made up of a SNCR, a spray absorber system (sodium bicarbonate and powder activated carbons), and a fabric filter
Plant 3	Grate type: roller-type grate Type of waste fed: municipal solid waste (MSW)	Wet process (wet scrubber) made up of a fabric filter and a SCR
Plant 4	Grate type: moving grate Type of waste fed: municipal solid waste (MSW)	Double-filtration approach: injection of lime milk before the first fabric filter and addition of sodium bicarbonate and activated carbon before the second fabric filter; NO _x reduction performed through SCR process
Plant 5	Grate type: fluidized bed reactor Type of waste fed: biomass	Wet process (wet scrubber) made up of a wet electrostatic precipitator.

Table 2. Main technical data of the analyzed plants

Plant	Waste Capacity (kg hr ⁻¹)	Average Low Heat Value (MJ kg ⁻¹)	Plant Annual Availability (hr)	Gross Electric Power (MW)	Bottom (Fly) Ashes (kg yr ⁻¹)	Reagent Consumption in Flue-Gas Treatment	Exhaust Speed at the Stack (m s ⁻¹)	Annual Mean NO _x Conc. at the stack (mg m ⁻³)	Annual Mean SO ₂ Conc. at the Stack (mg m ⁻³)	Annual Mean Dust Conc. at the Stack (mg m ⁻³)
1	10–12 × 10 ³	>15 MJ kg ⁻¹	8000	11.7	14 × 10 ⁶ (5 × 10 ⁶)	Calcium oxide CaO: 1.6 × 10 ⁶ kg yr ⁻¹ Powder activated carbons: 80 × 10 ³ kg yr ⁻¹ Urea: 160 m ³ yr ⁻¹	12	123	7.9	0.3
2	15 × 10 ³	18.2 MJ kg ⁻¹	8000	11.5	12 × 10 ⁶ (3.4 × 10 ⁶)	Urea: 270 × 10 ³ kg yr ⁻¹ Powder activated carbons: 80 × 10 ³ kg yr ⁻¹ Sodium bicarbonate NaHCO ₃ : 1.6 × 10 ⁶ kg yr ⁻¹	20	170	6	0.9
3	9–10 × 10 ³	>12 MJ kg ⁻¹	7800	6.0	17.5 × 10 ⁶ (1.2 × 10 ⁶)	Calcium hydroxide Ca(OH) ₂ : 250 × 10 ³ kg yr ⁻¹ Sodium hydroxide solution (30%): 160 × 10 ³ kg yr ⁻¹ Ammonia solution (26%): 100 m ³ yr ⁻¹	14	n.a.	n.a.	n.a.
4	12.5 × 10 ³	>12.5 MJ kg ⁻¹	8000	12	25 × 10 ⁶ (7 × 10 ⁶)	Sodium bicarbonate NaHCO ₃ : 330 × 10 ³ kg yr ⁻¹ Calcium hydroxide Ca(OH) ₂ : 2.2 × 10 ⁶ kg yr ⁻¹ Powder activated carbons: 130 × 10 ³ kg yr ⁻¹ NH ₃ solution (22%): 220 m ³ yr ⁻¹	12	n.a.	n.a.	n.a.
5	11.6 × 10 ³	>10 MJ kg ⁻¹	7500	19.5	3.8 × 10 ⁶ (3.3 × 10 ⁶)	n.a.	12	120	0.5	2.0

Note: n.a. = not available.

The main technical data of the plants are summarized in Table 2, where nominal performances in terms of annual reagent consumption are also reported.

Experimental apparatus

In order to measure total particle number concentrations and size distributions the following instruments were used:

- A condensation particle counter CPC 3775 (TSI Inc., Shoreview, MN) able to measure total particle number concentration down to 4 nm in diameter.
- A scanning mobility particle sizer spectrometer SMPS 3936 (TSI Inc., Shoreview, MN) made up of an electrostatic classifier EC 3080 (TSI Inc., Shoreview, MN), used to classify the sampled particles in different channel according to their size, and a CPC 3775 (TSI Inc. Shoreview, MN). The SMPS 3936 is able to measure particle number distribution in the range 6–800 nm and it can be also used to estimate surface area and mass distribution when a priori morphological and chemical analyses (useful to obtain information about the shape factor and the density of the particles) are carried out.
- A condensation particle counter CPC 5403 (Grimm, Ainring, Germany) able to measure total particle number concentration down to 4.5 nm in diameter.
- An electrostatic classifier “Vienna”-type DMA 55706 (Grimm) able to classify particles in the range 5.5–350 nm. It was also used in a scanning mobility particle sizer configuration when coupled with the CPC 5403.
- A thermodilution system (two-step dilution) made up of a rotating disk thermodiluter (model 379020; Matter Engineering AG, Wohlen, Switzerland) (Hüglin et al., 1997) and a thermal conditioner (model 379030; Matter Engineering AG, Wohlen, Switzerland) (Burtscher, 2005) allowing to ensure a proper sample conditioning during the measurement of number distributions and total concentrations of particles emitted by the waste incinerators.
- A thermodilution apparatus constituted by a continuous automatic dioxin monitoring system (DMS; Monitoring Systems, Bad Vöslau, Austria). It works according to the dilution method described in the BS EN 1948-1:2006 (European Committee for Standardization, 2006) and it is able to dilute sampled flue gas up to 1:10. Unlike the dioxin sampling, in our experimental analysis the dilution air was heated in order to perform a proper sample conditioning.
- A thermodilution apparatus made up of the diluter VKL-10E (Particle Technology Palas, Karlsruhe, Germany) cautiously heated up to actual flue-gas temperature in order to perform a proper sample conditioning. Dilution factors of 1:10 can be reached.

Methodology description

The experimental campaigns were carried out during the period 2007–2010. Measurements of total particle number concentrations and particle size distributions were performed at the

stack of each selected plant. Moreover, in order to deepen the fabric filter contribution to the particle abatement, measurements were carried out also at a section before the fabric filter for Plants 1 and 2. Sampling length of 120 sec was chosen for particle size distribution measurement, whereas total concentration data with a 1-sec time resolution were carried out. In Table 3 details of the experimental analyses carried out at the plants are summarized; in particular, measurement periods, measurement points, and experimental apparatus are reported.

Aerosol emitted from combustion sources (such as the incinerators) are typically hot, highly concentrated, and made up of volatile gaseous compounds that tend to condense, leading to either the formation of stable nuclei (nucleation) or the growth of existing particles (condensation). Therefore, it is necessary to properly dilute and thermally condition the aerosol; if not, particle size distributions and total concentrations could quickly undergo significant changes in the few seconds lasting between the aerosol sampling and its measurement (Hüglin et al., 1997; Burtscher, 2005; Holmes, 2007). Aerosol thermoconditioning is of great concern in UFP characterization from incinerators where precursor gases are typically produced. In fact, Buonanno et al. (2009a) observed a noteworthy correlation among particle number concentration and both NH₃ and SO₂ concentrations measured at the stack of an incinerator. In Table 3 dilution factor and thermodilution temperature are also reported for every analyzed plant. In each plant the sampling was performed through a probe that is connected to the thermodilution unit where the aerosol was kept at controlled temperature and humidity. After the thermodilution process, the aerosol was flown to particle counters or particle classifiers depending on whether particle number concentrations or size distributions were measured. In order to reduce artifacts during the measurements, an ad hoc sampling system was also designed to control condensation and nucleation processes in the tract connecting the probe (sampling point) to the thermodilution unit. Since the path experienced by the aerosol before entering in the measurement devices is quite long, a diffusion loss correction was applied to estimate the particle losses onto the inner surface of the connecting tubes. These corrections were evaluated according to the method proposed in Gormley and Kennedy (1949); further details about diffusion loss correction evaluation are reported in Buonanno et al. (2011).

Experimental campaigns of the six plants were carried out in sufficiently stable operating conditions. In Table 4 operative mean conditions and corresponding standard deviations of the main parameters are reported showing nearly steady-state operative conditions for the analyzed plants.

Results and Discussion

In this section results in terms of particle size distribution and total concentration measured at the plants analyzed through electrostatic classifiers and condensation particle counters are discussed.

Table 3. Details of the experimental analyses of the considered plants

Plant	Measurement Periods	Measurement Points	
Plant 1	October 2009	At the stack and before the fabric filter	Total particle number: Particle size distribution: Thermodilution:
Plant 2	September 2010	At the stack and before the fabric filter	Total particle number: Particle size distribution: Thermodilution:
Plant 3	February 2010	At the stack	Total particle number: Particle size distribution: Thermodilution:
Plant 4	September 2010	At the stack	Total particle number: Particle size distribution: Thermodilution:
Plant 5	May 2009	At the stack	Total particle number: Particle size distribution: Thermodilution:

Particle size distributions and total concentration values

In Figure 1 statistics of total particle concentrations measured at the stack of the analyzed plants through particle counters are reported. Average concentrations ranging from 0.4 to 6.0×10^3 particles cm^{-3} were measured in each plant and also maximum values are very low when compared to other anthropogenic sources (Cass et al., 2000; Morawska et al., 2008). Moreover, the significant difference between maximum and 3rd quartile values highlights that higher concentrations occasionally occurred.

As the measurements were carried out in nearly steady-state conditions, the wide deviations of the data with respect to the average and median concentrations can be only due to the presence of the fabric filters which are cleaned through pulse-jet (compressed air cleaning operation). In particular, the compressed air is injected as the cakes on the bag lead to high-pressure drops (pressure sensors are provided both at the outlet and at the inlet section of the filter). As for example, in Figure 2, a typical 10-min sample of total particle number concentration measured at the stack of Plant 2 is reported, it clearly shows an unsteady emission of the plant as every peak in concentration is due to a compressed-air pulse. Obviously, such dynamic behavior of emitted aerosol particles cannot be observed by the mass-integrated measurements required by law, since they are based on

the gravimetric time-integrated measurement technique (European Committee for Standardization, 2001; European Committee for Standardization, 2005).

In Figure 3 particle number distributions (fitted by using log-normal distribution functions) measured through particle mobility spectrometers at the stack of the analyzed incineration plants are reported. The data represent the particle number distributions corresponding to the highest emission periods of the plants. Plants 1 and 3 show a unimodal distribution with peak values in the range 60–100 nm, as also found by Buonanno et al. (2009a). Plants 2 and 4 show bimodal distributions, with one of the peaks in the nucleation range (about 10 nm). In particular, Plant 4 shows a second minor peak again in the nanoparticle range (about 30 nm) and no particles larger than 100 nm were measured. This behavior could be due to the presence of the double filtration approach by using two fabric filters.

Fabric filter efficiency

The importance of the fabric filter in the abatement of sub-micrometer particles was evaluated through the measurement of particle number distributions and total concentrations at a section before the fabric filter in two of the analyzed plants: Plants 1 and 2. In both of them the fabric filter represents the last device of the flue-gas treatment section (none of them uses SCR

Table 4. Operative mean condition and corresponding standard deviation for the main parameters of the analyzed plants

Parameter	Plant 1		Plant 2		Plant 3		Plant 4		Plant 5	
	Mean Value	Deviation (%)	Mean Value	Deviation (%)	Mean Value	Deviation (%)	Mean Value	Deviation (%)	Mean Value	Deviation (%)
Normalized flow rate ($\text{m}^3 \text{h}^{-1}$)	98.3×10^3	1.7%	75.8×10^3	1.2%	120.0×10^3	2.6%	100.0×10^3	1.6%	150.0×10^3	2.1%
Stack temperature ($^{\circ}\text{C}$)	135	3.0%	154	1.1%	150	3.0%	135	2.4%	40	5.2%
Combustion chamber temperature ($^{\circ}\text{C}$)	991	1.0%	1209	0.9%	1000	1.0%	980	1.1%	950	3.8%
Relative humidity (%)	15.3	6.5%	14.9	6.7%	15	7.3%	13	8.5%	65%	12.5%
O_2 in dry flue gas (%)	10.7	2.8%	8.7	1.1%	13	1.5%	10.5	2.9%	12.2	1.8%
SO_2 (mg m^{-3})	8.2	14.6%	5.1	15.7%	5	15.6%	0.5	12.0%	0.5	12.0%
NO_x (mg m^{-3})	115.0	7.5%	174.8	10.6%	50	11.2%	60.3	11.3%	120	10.7%
CO (mg m^{-3})	5.2	32.7%	5.5	98.2%	10	27.6%	30.1	29.9%	14	21.3%
Total dust (mg m^{-3})	0.68	16.2%	1	0.0%	2	34.5%	0.9	27.8%	2.0	26.3%
HCl (mg m^{-3})	4.3	7.0%	6.6	16.7%	<1	—	1.1	27.3%	0.6	15.4%

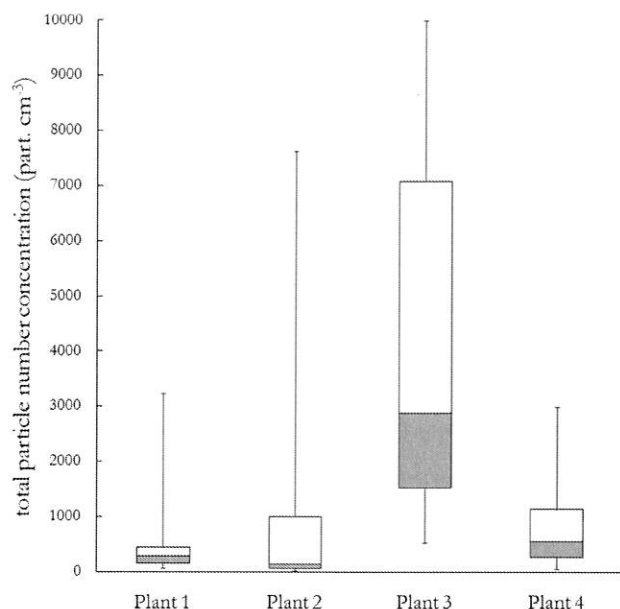


Figure 1. Statistics of total particle concentrations measured through particle counters at the stack of the analyzed incineration plants: minimum, maximum, median, 1st quartile, 3rd quartile values.

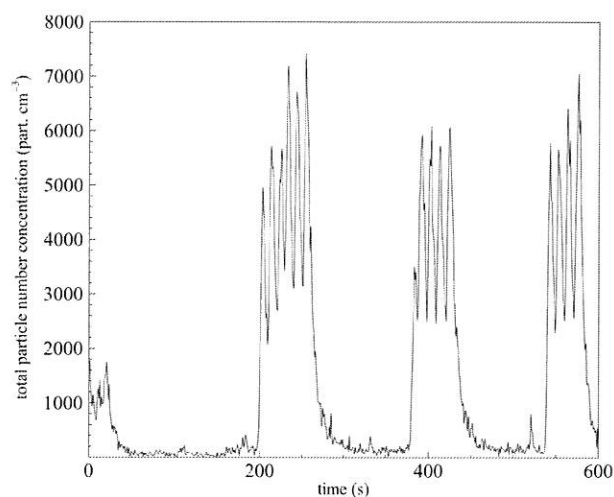


Figure 2. Example of a 10-min total particle number concentration measurement through CPC 3775 at the stack of Plant 2.

techniques to reduce NO_x or wet processes to abate acid gases). This is an essential circumstance to properly compare measurement data performed at the stack and before the fabric filter. Average total particle number concentrations before the fabric filter were measured to be equal to $2.4 \times 10^7 \pm 0.2 \times 10^7$ and $1.4 \times 10^7 \pm 0.1 \times 10^7$ particles cm^{-3} at Plants 1 and 2, respectively. Standard deviations well below 10% give evidence of the steady-state conditions during the measurement periods; it is also a further confirmation that the large deviation of the data at the stack can be only due to the fabric filter cleaning operations. In Figure 4 the comparison amongst particle number

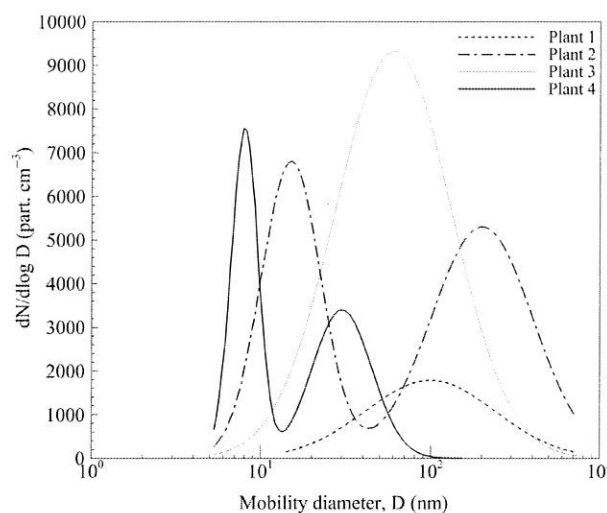


Figure 3. Particle number distributions measured through mobility particle sizers in the analyzed incineration plants. The data represent the particle number distributions corresponding to the highest emission periods of the plants.

distributions measured at the stack and before the fabric filter of the Plants 1 and 2 is reported. Logarithmic scale for $dN/d\log D$ axes needs to be used, as the total particle concentrations before and after filtration differ by 5 orders of magnitude: it gives evidence of the important contribution in the submicrometer particle reduction of the fabric filter. In this figure, particle size distributions corresponding to the section before the fabric filter represent the average distribution of the sampling time, as the particle concentrations are quite steady, otherwise, particle size distributions related to the stack represent the maximum measured distributions as also referred in Figure 3. Particle number distributions measured in both of the plants before the fabric filter show a main mode around 180–200 nm; Plant 2 also show a minor second mode at 10 nm, which was also detected at the stack. Even considering the maximum measured distributions at

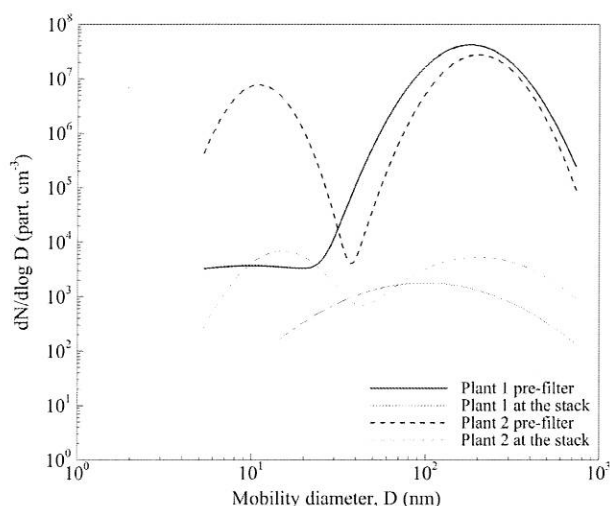


Figure 4. Particle number distributions measured through SMPS 3936 at the stack and before the fabric filter (labeled “pre-filter”) of Plants 1 and 2.

the stack, the filtration efficiencies of both the fabric filters, over the entire measurement range of the SMPS 3936 (nearly up to 1 μm), is higher than 99.99%. Therefore, the shift of the modes toward lower diameters (clearly distinguishable in Figure 4) does not involve a reduction of the global efficiency of the fabric filter. As for example, the abatement efficiency of the fabric filter for Plant 2 in the range 5–40 nm (the first mode), which is expected to be the lowest one, was estimated to be equal to 99.88%.

The importance of the fabric filter can be also recognized from the measurement carried out at the stack of Plant 5. It is fed by biomass, hence it is not a waste incinerator, but it shows a worthy flue-gas treatment section, as it is made up of Best Available Techniques (BAT) to handle the exhausts; in particular, as regard the particulate reduction, it presents an electrostatic precipitator (ESP). The average particle number distribution measured during the experimental campaign at Plant 5 is a log-normal distribution function with a mode of 90 nm, typical of wood combustion (Buonanno et al., 2010), and a standard deviation of 1.55. The corresponding average total particle number concentration was measured to be equal to 2.8×10^5 particles cm^{-3} . Even if a comparison with other plants (burning municipal waste or RDF) cannot be properly performed, the measurement results of Plant 5 roughly show that the ESP itself is not able to assure particle levels low as the ones measured in plants where a fabric filter is used.

Conclusions

An experimental analysis focused on ultrafine particle emission from incinerator plants was carried out. To this purpose, particle number distribution and total concentration measurements were performed at the stack and before the fabric filter of several incinerator plants through condensation particle counters and mobility particle sizer spectrometers.

The core finding of the work is the modest amount of UFPs emitted by the analyzed incinerators, since particle number concentrations at the stack are always lower than 1.0×10^4 particles cm^{-3} , in particular, average particle number concentrations at the stack range from 0.4 to 6.0×10^3 particles cm^{-3} .

On the contrary, average particle number concentrations before the fabric filter were measured to be equal to 1.4 and 2.4×10^7 particles cm^{-3} at Plants 1 and 2, respectively, leading to average removal efficiency of the fabric filters higher than 99.99%. In addition, measurements of the particle number distributions before and after the fabric filters show that the removal efficiency is quite constant all over the measurement range. Therefore, even if fabric filters are usually present in incinerators to meet regulatory requirements in terms of total dust at the stack (which is a mass-based threshold limit value), they assure a great filtration also in terms of UFPs.

In conclusion, the authors point out that future works will be focused on chemical and morphological analyses of emitted aerosol particles in order to perform a whole characterization of UFPs emitted by incinerators.

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**Appendix E Environment Agency internal briefing
note on UKWIN article July 2018**

Environment Agency internal briefing note on UKWIN article July 2018

What is this briefing note about?

This briefing has been put together by the Environment Agency in response to a report published by UK Without Incineration (UKWIN) on 17 July 2018 entitled “Waste Incineration and Particulate Pollution: A failure of governance”ⁱ. This briefing is primarily intended for internal Environment Agency use, but can be shared externally if required.

This briefing addresses the various points made in the UKWIN report and provides further information about the challenges around monitoring particulates at the very low concentrations found in the exhaust gases of modern municipal solid waste (MSW) incinerators (also known as energy-from-waste or EfW plants). It also provides data on the amount of particulate matter and oxides of nitrogen (NO_x) emitted from EfW plants compared to other common sources, and how we assess the impact of an EfW plant’s emissions when deciding whether to grant a permit. A list of key messages can be found at the end of the briefing.

Some explanation about different sizes of particulate matter and how it is monitored

Particulate matter (PM), also known simply as “dust”, is emitted from many different sources including cars, household wood burning and agriculture. PM is classified according to size, with the smaller particles thought to be more likely to have an impact on health. PM₁₀, for example, is all particles with a diameter of 10 micrometres *or less*, and therefore includes smaller particles such as PM_{2.5} and PM₁ etc.

There is currently no validated, commercially available equipment for continuously monitoring PM₁₀ and PM_{2.5} emissions from EfW plants. Instead, plants are required to continuously measure *total* particulate matter (TPM). TPM includes particulates of *all* sizes including PM₁₀, PM_{2.5}, PM₁ etc as well as ultrafine particles (i.e. particles with a diameter of less than 0.1 micrometres).

Equipment is available to monitor PM₁₀ and PM_{2.5} *discontinuously* i.e. by using temporary monitoring equipment to sample the exhaust gas and then working out the results in a laboratory. Indeed, all new EfW plants are required to carry out this test when they first start operating. However, the concentrations of PM in the exhaust gases of modern EfW plants are so low that it is very difficult to get an accurate result from these tests, and will remain so until new monitoring methods and technology can be developed, validated and standardised for use.

In summary, specific emissions of PM₁₀ and PM_{2.5} from EfW plants can’t be accurately measured using current technology. However, this isn’t really a problem as all EfW plants continuously measure their TPM emissions, which includes particulates of *all* sizes. If we then want to know the impact of PM₁₀ from an EfW plant under the worst-case scenario, we can simply assume that *all* of the TPM measured is PM₁₀, and the same for PM_{2.5} and so on.

How does the Environment Agency assess impacts of EfW plants on the environment and human health?

We use a number of methods, but one of the key assessments for PM₁₀, PM_{2.5} and NO_x is to compare the modelled emissions from the EfW plant with the European air quality standards for these pollutants (also taking into account the existing levels of pollution around the plant). To do that, we assume that the plant operates at its permitted limits 100% of the time (when in reality it won’t, especially for TPM where plants often operate at around 10% of their limits). For PM₁₀ and PM_{2.5} we also assume that TPM = PM₁₀ = PM_{2.5} as explained above. Making these assumptions means that we assess the worst-case scenario, which is what we then base our permitting decisions on, and we also consult Public Health England (PHE) on every application that we receive.

Do EfW plants make a big contribution to particulate matter and NO_x emissions in the UK?

The table overleaf shows estimates of the amount of pollution that was released by different example sources listed in the Government’s National Atmospheric Emissions Inventoryⁱⁱ (NAEI, which is referenced in the UKWIN report). These include figures for domestic wood burning (i.e. wood fires and stoves in people’s homes) and emissions from road transport including cars, buses and lorries.

The data shows that emissions from EfW plants make up just 0.03% / 0.05% of total UK PM₁₀ / PM_{2.5} emissions. This is compared to 5.35% / 4.96% from traffic and 22.4% / 34.3% from domestic wood burning. For NO_x the figures are 1.12% from EfW plants compared to 33.5% from traffic and 0.57% from domestic wood burning.

2016 NAEI category	PM ₁₀	PM _{2.5}	NO _x
MSW incineration	0.057 kt = 0.03%	0.057 kt = 0.05%	9.97 kt = 1.12%
Domestic wood burning	38 kt = 22.4%	37 kt = 34.3%	5.1 kt = 0.57%
Cars, buses, lorries	9.1 kt = 5.35%	5.36 kt = 4.96%	298.9 kt = 33.5%
Total UK emissions	170 kt	108 kt	893 kt

(Source: <http://naei.beis.gov.uk>; kt = kilotonne i.e. 1000 tonnes)

It is also important to understand that the overall impact of an EfW plant's emissions on human health for a given amount of PM or NO_x released will be lower than if that same amount was emitted by a car or a domestic wood fire. This is because EfW plants have tall stacks (chimneys) which help to disperse their emissions, whereas a car exhaust pipe or a chimney on a house releases its emissions much closer to ground level.

Are emissions from EfW plants causing significant health effects in England?

We consult Public Health England (PHE) on every EfW plant application that we receive and we will not issue a permit if its emissions will cause significant pollution or harm to human health. PHE has also published the following position statement on the health impact of waste incineration: *“Modern, well managed incinerators make only a small contribution to local concentrations of air pollutants. It is possible that such small additions could have an impact on health but such effects, if they exist, are likely to be very small and not detectable.”* The study of all 22 British EfW plants in operation 2003–10ⁱⁱⁱ indicates very low concentrations of incinerator-related PM₁₀ within 10 km of the plants at postcode level.

What is the Environment Agency's response to the points covered in the UKWIN report?

The following table provides a summary of our responses to the main points covered in the UKWIN report and should be read together with the information above.

Claim made or policy called for	Environment Agency response
The public have been “kept in the dark about PM ₁₀ and PM _{2.5} emissions” as there is no equipment available for their continuous monitoring.	The fact that PM ₁₀ and PM _{2.5} emissions cannot be continuously monitored does not mean that they cannot be estimated and the estimates made publically available. Indeed, this is what the NAEI does, with data available to the public going back to 1970. The 2016 data for example shows that EfW plants emitted an estimated 57 tonnes of both PM ₁₀ and PM _{2.5} , representing 0.03% and 0.05% of total UK emissions respectively. In comparison, the NAEI estimates that domestic wood burning accounted for 22% and 34% of total UK PM ₁₀ and PM _{2.5} emissions respectively.
There is a “TPM fiddle” which prevents the public from being told about TPM emissions from incinerators.	All EfW plants must continuously monitor and report TPM emissions on a quarterly basis. The results of this monitoring are placed on the public register and show that many EfW plants operate at around 10% of their emission limit for TPM.
There is a “no equipment fiddle” which allows operators to say they can't measure PM ₁₀ and PM _{2.5} when in actual fact they can measure them “by proxy”.	The method used by the NAEI is not a form of <i>measurement</i> but rather it is a conservative <i>estimate</i> of the PM ₁₀ and PM _{2.5} emissions which relies on the simple assumption that TPM = PM ₁₀ = PM _{2.5} .
Incinerator operators have been ignoring Environment Agency guidance on reporting PM ₁₀ and PM _{2.5} ; PM ₁₀ and PM _{2.5} reporting should be made mandatory and guidance should be strengthened and enforced.	As explained above, EfW operators cannot specifically measure their PM ₁₀ and PM _{2.5} emissions in an accurate way. As the UKWIN report highlights, our Pollution Inventory (PI) guidance suggests that emission factors can be used. However, these emission factors are from 2000 (when not all EfW plants were required to be fitted with bag filters) which may help explain the difference between the UKWIN figures (226.1 tonnes for England in 2017) and the NAEI data (57 tonnes for the whole of the UK in 2016). We are in the process of updating our guidance to make it clear that PM ₁₀ and PM _{2.5} emissions must be reported on the PI, as well as providing an updated method to enable operators to estimate them.
A limit value should be placed on PM ₁ emissions from incinerators if possible.	A limit on PM ₁ emissions is arguably not necessary as PM ₁ will be included in TPM emissions, and in any case, PM ₁ emissions will be taken into account when assessing an EfW plant's emissions against the air quality standards for PM ₁₀ and PM _{2.5} (which will both include PM ₁ and ultrafines as explained above).
An incineration tax should be introduced under the “polluter pays” principle and there should be a moratorium on new incinerators until this and the other policies mentioned are in place.	Whether waste incineration should be taxed or a moratorium put in place are decisions for the Government and not the Environment Agency. We will continue to consider permit applications for new EfW plants in the same way i.e. by assessing the impacts of particulates and other pollutants on the environment and human health.

Summary/key messages for a non-technical audience

- The UKWIN article is about municipal solid waste (MSW) incinerators, also known as energy-from-waste or EfW plants.
- The article talks mainly about emissions of particulate matter (PM), which is also known simply as “dust”. PM is emitted from many different sources including cars, household wood burning and agriculture.
- PM can be classed by size e.g. PM₁₀ refers to all particles with a diameter of 10 micrometres (µm) **and smaller**, and PM_{2.5} means those with diameter of 2.5 µm **and smaller**. This means that PM₁ and “ultrafine particles” (with a diameter of less than 0.1 µm) are included in PM₁₀ and PM_{2.5} measurements.
- Emissions of PM₁₀ and PM_{2.5} from modern EfW plants are so low that they cannot be accurately specifically measured using currently available technology. However, this isn’t a problem as all EfW plants continuously monitor emissions of total PM (TPM) which includes particles of **all** sizes including PM₁₀, PM_{2.5}, PM₁ and ultrafine particles.
- EfW plant operators report their continuous monitoring results (including TPM) to the Environment Agency (EA) every 3 months and these are all placed on the public register^{iv}.
- EfW plants also submit annual reports of their emissions to the EA’s Pollution Inventory (PI). The UKWIN article is critical of the fact that EfW plants do not always provide estimates of their PM₁₀ and PM_{2.5} emissions to the PI. Because of this, the EA is going to update its guidance to make it clear that estimates for these pollutants need to be submitted in the future.
- When the EA assesses applications for new EfW permits, they compare the maximum emissions from the plant against European air quality standards. For PM₁₀ and PM_{2.5} this means making a worst-case assumption that all of the EfW plant’s emissions will be either PM₁₀ or PM_{2.5}. The EA will not issue a permit for an EfW plant if its emissions will cause significant pollution or harm to human health, and it consults Public Health England (PHE) on every application it receives.
- PHE’s position is that well run and regulated modern Municipal Waste Incinerators are not a significant risk to public health. This view is based on detailed assessments of the effects of air pollutants on health and on the fact that modern and well managed Municipal Waste Incinerators make only a very small contribution to local concentrations of air pollutants.
- For more information on PHE’s position, see:
<https://www.gov.uk/government/publications/municipal-waste-incinerator-emissions-to-air-impact-on-health>
- EfW plants are an extremely small source of PM in the UK, giving rise to just 0.03% / 0.05% of total UK PM₁₀ / PM_{2.5} emissions in 2016 according to government estimates. This compares to 5.35% / 4.96% from traffic and 22.4% / 34.3% from wood fires and stoves in people’s houses.
- The other pollutant mentioned in the UKWIN article is oxides of nitrogen (NO_x). EfW plants are also a relatively small source of NO_x in the UK, giving rise to 1.12% of emissions in 2016 compared to 33.5% from traffic and 0.57% from domestic wood burning according to government estimates.

ⁱ http://ukwin.org.uk/btb/Particulate_Pollution_July_2018.pdf

ⁱⁱ <http://naei.beis.gov.uk/data/>

ⁱⁱⁱ <https://pubs.acs.org/doi/pdf/10.1021/acs.est.6b06478>

^{iv} <https://environment.data.gov.uk/public-register/view/index>