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Estimating Particulate Exposure from Modern Municipal Waste Incinerators in Great Britain

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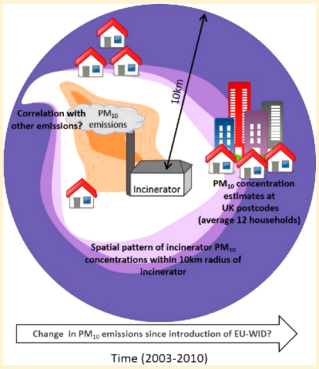
Supporting Information

ABSTRACT: Municipal Waste Incineration (MWI) is regulated through the European Union Directive on Industrial Emissions (IED), but there is ongoing public concern regarding potential hazards to health. Using dispersion modeling, we estimated spatial variability in PM_{10} concentrations arising from MWIs at postcodes (average 12 households) within 10 km of MWIs in Great Britain (GB) in 2003−2010. We also investigated change points in PM_{10} emissions in relation to introduction of EU Waste Incineration Directive (EU-WID) (subsequently transposed into IED) and correlations of PM_{10} with SO_2, NOx, heavy metals, polychlorinated dibenzo-*p*-dioxins/furan (PCDD/F), polycyclic aromatic hydrocarbon (PAH) and polychlorinated biphenyl (PCB) emissions. Yearly average modeled PM_{10} concentrations were 1.00×10^{-5} to 5.53×10^{-2} $\mu g\ m^{-3}$, a small contribution to ambient background levels which were typically $6.59-2.68 \times 10^{1}$ $\mu g\ m^{-3}$, 3−5 orders of magnitude higher. While low, concentration surfaces are likely to represent a spatial proxy of other relevant pollutants. There were statistically significant correlations between PM_{10} and heavy metal compounds (other heavy metals ($r = 0.43$, $p = <0.001$)), PAHs ($r = 0.20$, $p = 0.050$), and PCBs ($r = 0.19$, $p = 0.022$). No clear change points were detected following EU-WID implementation, possibly as incinerators were operating to EU-WID standards before the implementation date. Results will be used in an epidemiological analysis examining potential associations between MWIs and health outcomes.



INTRODUCTION

Incineration of domestic and commercial waste is increasing in Europe in response to European Union (EU) legislation to divert waste from landfill sites. Waste incinerator feedstock includes paper, food, plastics, glass, electrical appliances, and other nonhazardous materials and may vary day to day and from incinerator to incinerator.[1] Composition of combustion emissions depends on feedstock mix but potentially comprises particulate matter, sulfur dioxide (SO_2), nitrogen oxides (NOx), 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.[1,2] Limits for incinerator emissions are set by the EU. The Waste Incineration Directive (EU-WID) (2000/76/EC) was implemented in Great Britain (GB) on 28 December 2002 for new municipal waste incinerators (MWIs) and on 28 December 2005 for existing facilities. In 2010 the EU-WID was transposed into the Industrial Emissions Directive (IED) (2010/75/EU) which combined several directives; this was implemented in GB in 2013 for new MWIs and 2014 for existing facilities.[3,4]

While there is public concern regarding potential adverse health effects from MWI emissions, findings from epidemiological studies are inconsistent and inconclusive.[5] Most studies have focused on adult cancers[6−9] and to a lesser extent reproductive and child health outcomes.[5,10−14] Exposure assessment has often used simple proxies, adopting proximity to incinerator as the exposure measure.[15−18] There have been three recent studies of incinerators conducted in Italy. One used a modified risk-assessment model to estimate lung cancer risk.[20] The remaining two used a dispersion model to assess exposure to particulate matter with a diameter <10 μm (PM_{10})

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and found a higher risk of miscarriage[13] and preterm delivery[14] with increasing PM_{10} exposure (but no associations with sex ratio, multiple births, or frequency of small for gestational age[14]), where estimated PM_{10} levels from incinerators were consistent with those estimated near two British incinerators.[19]

Particulate matter/total dust emissions are monitored continuously and reported as daily means as part of the EU-WID regulations, so that dispersion of these emissions can be modeled in areas near incinerators on a daily basis (whereas heavy metals, PCDD/Fs, PAHs, and PCBs are measured periodically[3] to check compliance). We previously reported[19] methods for dispersion modeling around two MWIs. In the present study, our main aim was to model the spatial distribution of PM_{10} concentrations within 10 km of GB MWIs in operation 2003−2010 for the resident population. We were also able to look at whether there were the following:

1. Emissions above the EU-WID daily average particulate (total dust) limit value of 10 mg m^{-3} per flue.[4]
2. Correlations and associations between PM_{10} emissions and within-flue emission measurements of heavy metals, PCDD/Fs, PAHs, and PCBs (to provide information about the chemical composition of PM_{10} being emitted from flues).
3. Changes in levels of PM_{10} emissions after the implementation of the EU-WID, which reduced daily average emission limit values from 30 mg m^{-3} to 10 mg m^{-3} for particulate matter/total dust.[21]

We refer to PM_{10} rather than total suspended particulates (total dust) throughout as size fraction studies have found all particulate incinerator emissions are <10 μm diameter.[22]

MATERIALS AND METHODS

Study Area. We included all 22 MWIs in Great Britain (Figure 1) in operation between 2003 and 2010, comprising 14 existing MWIs and 8 new MWIs opening after 2003. We excluded one MWI in the Isle of Man (Richmond Hill) for which we did not have health or emissions data and three other incinerators open during this time as they were not solely MWIs (Fawley, Hampshire; Ellesmere Port, Cheshire; Peak Load Boiler, Shetland). The location of MWIs varied as to the geographical characteristics of their location and topography. A 10 km radius around each MWI was chosen as the study area as per Ashworth et al.[19]

Incinerator Data. Information on the emissions, total annual licensed throughput, the number of flues, and whether an MWI opened to or adopted EU-WID specifications were provided by the Environment Agency (EA), Natural Resources Wales (NRW), and the Scottish Environment Protection Agency (SEPA) (Table 1). Information on characteristics used in dispersion modeling including height and diameter of the MWI stack (m), exit temperature (°C), and exit velocity (m s^{-1}) per MWI per flue are reported in Supporting Information (SI) A, Table S1. Daily measured PM_{10} emissions per MWI, per flue, and per year (some originally in paper format, which were digitized and quality checked by a third party) were provided by the EA and SEPA. Non-numeric and negative values were recoded according to an algorithm agreed with the EA and SEPA (SI B, Table S2).

MWIs varied in size (Table 1, SI A) and location (Figure 1). The licensed throughput varied from 3,500 (Porthmellon) to 750,000 (Edmonton) tonnes per annum (Table 1). Populations living within 10 km of each MWI varied from 2,203 (Porthmellon) to 2,726,145 (SELCHP) (information from census 2011 data). The majority of MWIs had multiple flues

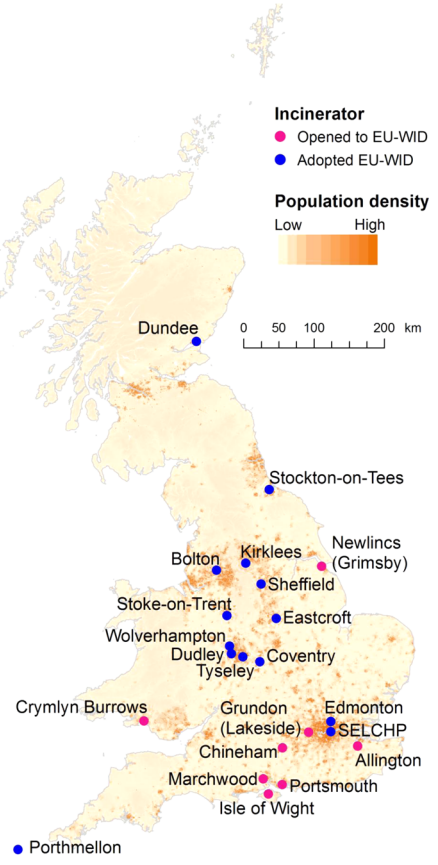

Figure 1. Location of all MWIs operating in England, Wales, and Scotland between 2003 and 2010. Base map: population density in 2001. SELCHP is an abbreviation for South East London Combined Heat and Power.

(15 MWIs; 12 with two flues, three with three flues). The number of nonoperational and missing days varied from MWI to MWI and from year to year and occurred sporadically for a few days or for longer periods (e.g., several months; Table 1 and SI C Table S3).

The availability of heavy metals, PCDD/Fs, PAHs, and PCB measurements varied. MWIs are required to complete at least two measurements per annum.[4] Typically in-flue measurements of 6−8 heavy metals and 3−4 PCDD/F, PAH, and PCBs are completed per year, per MWI, and per flue, but repeated measurements are taken if higher than limit values. Heavy metals, PCDD/Fs, PAHs, and PCBs were monitored in-flue, usually at the same time as each other over an 8 h period, using European committee standards (CEN).

Dispersion Modeling To Estimate Spatial Distribution of PM_{10} Concentrations within 10 km of GB MWIs. The Atmospheric Dispersion Modeling System Urban (ADMS-Urban) (version 2.3), utilized in previous studies characterizing emissions from MWIs,[13,14,19,23,24] was used to model ground-level PM_{10} concentrations for postcode (average 12 household per postcode) area centroids within a 10 km radius of the MWIs. ADMS-Urban is a Gaussian based dispersion model that has been widely used and extensively validated. The model characterizes the atmospheric boundary layer using the Monin-Obukhov length and boundary layer depth. It is capable of simulating the effects of plume rise and the effects of buildings and complex topography on dispersion.[25]

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Table 1. Characteristics of the 22 MWIs

MWI	licensed throughput (t year^{-1})	years of data available	population within 10 km[a]	flues	no. of operational days	no. of days of missing data	no. of nonoperational days	no. of days of emissions above the EU-WID limit[b]	concentration of highest PM_{10} emission above EU-WID limit[b] (mg m^{-3})
Opened to EU-WID Specifications									
Allington	500000	2006−2010	311,067	1	768	0	1058	14	18
				2	740	0	1086	35	27
				3	766	0	1060	57	28
Chineham	90000	2003−2010	153,411	1	1830	861	228	0	-
Crymlyn Burrows	166000	2003−2010	266,736	1	1308	63	1551	0	-
Grundon (Lakeside)	400000	2010	676,430	1	311	0	54	0	-
				2	307	0	58	0	-
Isle of Wight	38000	2009−2010	60,915	1	186	0	544	0	-
Marchwood	165000	2004−2010	389,970	1	1950	0	607	0	-
				2	1924	2	631	0	-
Newlincs (Grimsby)	56000	2004−2010	143,525	1	2127	169	261	1	19
Portsmouth	165000	2005−2010	444,963	1	959	0	308	0	-
				2	1357	0	307	0	-
Adopted EU-WID Specifications									
Bolton	128000	2003−2010	609,405	1	2362	159	6	401	26
Coventry	315000	2003−2010	399,016	1	1859	853	210	2	14
				2	1837	853	232	5	15
				3	1858	911	153	12	26
Dudley	105000	2003−2010	860,444	1	1555	1280	87	7	46
				2	1549	1306	67	8	54
Dundee	175200	2005−2010	172,002	1	1481	8	702	32	25
				2	1126	285	780	7	45
Eastcroft	160000	2003−2010	565,241	1	2136	129	657	0	-
				2	2164	107	651	0	-
Edmonton	750000	2003−2010	1,780,440	1	2799	92	31	2	11
				2	2863	14	45	0	-
Kirklees	150000	2003−2010	410,698	1	1473	1261	188	4	66
Porthmellon	3500	2003−2010	2,203	1	3809	671	1365	70	85
SELCHP[c]	420000	2003−2010	2,726,145	1	2611	31	280	2	12
				2	2534	32	326	0	-
Sheffield	225000	2003−2010	653,522	1	2512	1	409	2	19
				2	900	0	2022	0	-
Stockton-on-Tees	263000	2003−2010	388,739	1	2339	141	442	72	44
				2	2448	122	352	35	66
				3	473	10	2438	0	-
Stoke-on-Trent	210000	2003−2010	362,462	1	2143	455	324	14	17
				2	2143	455	324	13	25
Tyseley	400000	2003−2010	1,121,165	1	2412	125	385	0	-
				2	2392	122	408	0	-
Wolverhampton	110000	2003−2010	611,053	1	2405	397	120	6	19
				2	2257	546	119	3	12

[a]Taken from census 2011data. [b]Daily average particulate limit value of up to 10 mg m^{-3} per flue. [c]South East London Combined Heat and Power.

The parametrization of the dispersion model is described in Ashworth et al.[19] In brief, all MWIs were modeled as point sources. Locations were verified using site addresses, grid references, and aerial photography. The EU-WID requires that average emission values are reported after subtraction of a fixed amount (taken as 30%) to account for measurement instrument uncertainty.[4] The EU-WID also allows up to ten measured daily average values to be discarded per year if there has been measurement instrument calibration or maintenance. For the purposes of the dispersion modeling, the emissions data were therefore increased by 30% from the reported values provided (except for Isle of Wight MWI, which did not subtract 30% to account for measurement instrument uncertainty). For the purposes of assessing emissions above the EU-WID daily average, emission values with 30% subtracted were used, as this is how compliance is assessed against the EU-WID limits. Missing emissions data were imputed using the median PM_{10} value of the operational days for each year and each MWI (for justification of this approach see SI D; for counts of days with missing data per flue/year/MWI see SI C). For Coventry, Dudley, and Kirklees MWIs, data were missing for entire year(s), and therefore it was not possible to model PM_{10} dispersion for these years. Hourly meteorological data from meteorological stations within 30 km of each MWI were

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Table 2. Annual Mean Modeled PM_{10} Concentrations ($\mu g\ m^{-3}$) by MWI[a]

MWI	year							
	2003	2004	2005	2006	2007	2008	2009	2010
Allington	N	N	N	1.50×10^{-3}	2.05×10^{-3}	2.50×10^{-4}	4.20×10^{-4}	2.54×10^{-3}
Bolton	7.20×10^{-4}	7.00×10^{-4}	1.16×10^{-3}	1.34×10^{-3}	8.10×10^{-4}	4.40×10^{-4}	3.70×10^{-4}	6.60×10^{-4}
Chineham	M	M	7.20×10^{-4}	5.50×10^{-4}	3.10×10^{-4}	9.00×10^{-5}	1.10×10^{-4}	3.20×10^{-4}
Coventry	M	5.04×10^{-3}	5.53×10^{-2}[a]	2.30×10^{-3}	1.64×10^{-3}	4.73×10^{-3}	2.03×10^{-3}	6.14×10^{-3}
Crymlyn Burrows	6.00×10^{-5}	N	1.00×10^{-5}	1.10×10^{-4}	2.60×10^{-4}	2.00×10^{-4}	9.00×10^{-5}	5.00×10^{-5}
Dudley	M	M	M	1.42×10^{-3}	4.60×10^{-4}	1.18×10^{-3}	1.23×10^{-3}	6.30×10^{-4}
Dundee	N	N	3.00×10^{-5}	6.30×10^{-4}	1.03×10^{-3}	1.35×10^{-3}	1.08×10^{-3}	4.10×10^{-4}
Eastcroft	2.50×10^{-4}	1.30×10^{-4}	1.30×10^{-4}	1.40×10^{-4}	1.70×10^{-4}	2.30×10^{-4}	2.30×10^{-4}	5.00×10^{-4}
Edmonton	5.93×10^{-3}	5.47×10^{-3}	3.32×10^{-3}	3.01×10^{-3}	5.10×10^{-3}	5.59×10^{-3}	5.67×10^{-3}	5.10×10^{-3}
Grundon (Lakeside)	N	N	N	N	N	N	N	9.60×10^{-4}
Isle of Wight	N	N	N	N	N	N	4.00×10^{-5}	2.00×10^{-5}
Kirklees	M	M	M	3.00×10^{-4}	1.83×10^{-3}	2.80×10^{-4}	5.30×10^{-3}	4.50×10^{-4}
Marchwood	N	5.30×10^{-4}	6.40×10^{-4}	1.27×10^{-3}	2.15×10^{-3}	2.44×10^{-3}	6.10×10^{-4}	5.40×10^{-4}
Newlincs (Grimsby)	N	4.80×10^{-4}	4.80×10^{-4}	3.20×10^{-4}	2.00×10^{-4}	2.30×10^{-4}	1.90×10^{-4}	2.70×10^{-4}
Porthmellon	5.10×10^{-3}	2.37×10^{-3}	2.85×10^{-3}	1.50×10^{-4}	3.28×10^{-3}	4.72×10^{-3}	7.72×10^{-3}	7.43×10^{-3}
Portsmouth	N	N	3.60×10^{-4}	4.90×10^{-4}	7.60×10^{-4}	5.70×10^{-4}	8.40×10^{-4}	7.30×10^{-4}
SELCHP[c]	8.40×10^{-4}	1.78×10^{-3}	1.49×10^{-3}	1.20×10^{-3}	1.35×10^{-3}	1.32×10^{-3}	1.75×10^{-3}	3.73×10^{-3}
Sheffield	1.10×10^{-4}	1.00×10^{-4}	3.40×10^{-4}	6.00×10^{-4}	2.50×10^{-4}	1.40×10^{-4}	1.90×10^{-4}	1.40×10^{-4}
Stockton-on-Tees	2.08×10^{-3}	1.54×10^{-3}	1.47×10^{-3}[b]	3.40×10^{-4}	4.40×10^{-4}	7.40×10^{-4}	5.90×10^{-4}	9.20×10^{-4}
Stoke-on-Trent	1.44×10^{-3}	1.22×10^{-3}	2.00×10^{-5}[b]	6.00×10^{-4}	4.60×10^{-4}	4.40×10^{-4}	6.00×10^{-4}	4.30×10^{-4}
Tyseley	1.89×10^{-3}	1.04×10^{-3}	1.24×10^{-3}	1.29×10^{-3}	2.21×10^{-3}	1.26×10^{-3}	1.49×10^{-3}	1.58×10^{-3}
Wolverhampton	2.55×10^{-3}	2.05×10^{-3}	3.60×10^{-4}[b]	1.13×10^{-3}	8.00×10^{-4}	1.62×10^{-3}	1.86×10^{-3}	6.90×10^{-4}

[a]N denotes that the MWI was not in operation that year, and M denotes that there were missing data for the entire year and therefore it was not possible to model PM_{10} dispersion for these years. [b]Note that over two-thirds of the year had missing days of data (see SI C). [c]South East London Combined Heat and Power.

obtained and selected for use based on land cover (to ensure that the land type surrounding the meteorological station was representative of the MWI) and completeness of data (for choice of meteorological station and justification see SI E). The Monin-Obukhov (MO) length (the height at which turbulence is driven by buoyancy instead of wind) and surface roughness (SR) at the dispersion site were computed by ADMS-Urban based on input meteorological variables and surface information extracted from CORINE land cover data from 2000 as per Ashworth et al.[19] (SI F, Table S5 contains MO and SR values). Complex terrain was included in the dispersion modeling if more than 5% of the area surrounding a MWI (within 10 km) contained slopes of 10% or higher, which was assessed using Ordnance Survey PANORAMA data. Daily average PM_{10} concentrations were calculated at every postcode area centroid within 10 km of each MWI. Where the 10 km radius around MWIs overlapped (Edmonton and SELCHP in London; Tyseley, Dudley, and Wolverhampton in the Midlands) the modeled output concentrations were summed for each day. Our dispersion modeling was specific for MWIs and did not account for sources of PM_{10} around the incinerators.

It was not practical to include the effects of buildings within the ADMS-Urban model as the study area was large (10 km radius from each MWI). Moreover the MWI stack height was high (median = 72.5 m, max = 100 m), taller than surrounding buildings, and thus nearby buildings would not affect dispersion patterns. Instead different MO and SR lengths for each MWI were used to represent different land-uses around each MWI and effects on pollutant turbulence and dispersion (see SI F). It was not considered possible to model spatial distributions of PCDD/F, PCB, PAH, or heavy metals using flue emissions data due to the sparseness and variability of the measurements.

Correlations and Associations between PM_{10} Emissions and Other Flue Emissions. Pairwise correlation was used to evaluate correlations of heavy metal compounds, PCDD/Fs, PAHs, and PCBs and daily averages of SO_2, NOx, and PM_{10} measured during the same time period. Measurements of heavy metals for most incinerators were reported as Cd and Tl and their compounds (CdTl), Hg and its compounds (HgComp), and grouped other heavy metals (OHMs) comprising Sb, As, Cr, Pb, Co, Cu, Mn, Ni, and V (see SI G, Table S6). As the data were not normally distributed, a nonparametric Spearman's rank correlation was used. This produces a coefficient, r, which ranges from −1 to 1. Values of −1 and 1 represent perfect negative or positive correlation, respectively, whereas a value of 0 represents no correlation. A Spearman correlation p-value <0.05 was considered statistically significant. As a Spearman's rank correlation will not account for differences in MWI operations, flues, and years of data, a linear multiple regression model was used to adjust for these factors; we considered one pollutant at a time and used PM_{10}, year, flues, and MWI as predictors. Data were checked for normality (from Q-Q plots) and log transformed if necessary. We report the estimated coefficients with p-values, and the partial η^2 -which is the variance associated with an effect divided by that variance plus the error variance, to describe the proportion of variance accounted for by the variable.

Detecting Changes in Levels of PM_{10} Emissions after the Implementation of the EU-WID. Data from MWI installations operating prior to the EU-WID were investigated to determine if emissions changed before or after the implementation date and when any change took place, as timings could be used to inform epidemiological analyses investigating changes in health outcomes rates before/after EU-WID implementation. First, we conducted a descriptive analysis

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of the daily average modeled concentrations (Table 2, SI H, Figure S3) over time. Second, as MWIs had implemented technical changes at unspecified time points leading up to the EU-WID implementation date of 28 December 2005, we carried out a retrospective change point identification analysis using a Batch Change Detection (BCD) approach, which attempts to identify periods when there are changes in the time series distribution. We were unable to use time trend analysis due to gaps in the data from nonoperational days and missing data. For the BCD analysis we used monitored in-flue 2003−2010 PM_{10} emissions data for 11 of the 14 existing MWIs in operation when the EU-WID was adopted (Table 1). Dundee MWI was excluded as data were only available from 2005, Dudley MWI and Kirklees MWI were excluded as data were missing for 2003−2005, and the third flue for Stockton-on-Tees was excluded as it was only operational in 2009 and 2010 (see SI C, Table S3). Coventry MWI could only be considered from 2004 onward as data for 2003 were missing (see SI C, Table S3). Daily in-flue PM_{10} emissions data were treated as independent observations. Only days when the incinerator was classed as operational and reported daily PM_{10} in-flue concentrations were included. The date of the change point τ was estimated by a nonparametric Cramér-von Mises test (details in SI I, Equation S1). This test was chosen as it suits non-Gaussian distributed data, characterized by zero inflation and by extreme values. It tests for shifts in scale (variability measures, e.g. variance) or location (e.g. mean, median). We computed a test value for each observation and defined the change point as the maximum test value. The statistical analyses were conducted in R using the cpm package.[26]

RESULTS

Dispersion Modeling To Predict Spatial Distribution of PM_{10} Concentrations within 10 km of GB MWIs. Annual mean average modeled PM_{10} concentrations (based on daily average modeled PM_{10} concentrations) per MWI and per year ranged from 1.00×10^{-5} to 5.53×10^{-2} $\mu g\ m^{-3}$ (Table 2). Complex overlapping dispersion patterns were shown for those areas with overlapping fields from multiple MWIs (Figure 2): surfaces (a) SELCHP and Edmonton and (b) Dudley, Tyseley, and Wolverhampton. In some instances (429 days total, 0−83 days per incinerator), a modeled output value was not calculated (across all postcodes) by ADMS-Urban even though the MWI was classified as being operational ("on" or "missing"). This may occur for a number of reasons but is commonly due to data processing error (e.g., when wind speed values are very low (<0.75 m s^{-1} when measured at 10 m above ground level) or due to missing meteorological data.[27]

Emissions above the EU-WID Daily Average Particulate Limit Value. There were a small number of days with emissions above the EU-WID daily average particulate limit value in 14 of the 22 MWIs, the majority of which were <20 mg m^{-3} (Table 1; SI J Table S7). There was no distinct pattern that might indicate that there were fewer emissions above the EU-WID daily average particulate limit value after the implementation of the EU-WID or more instances of emissions >10 mg m^{-3} before it.

Correlations and Associations between PM_{10} Emissions and Other Flue Emissions. Statistically significant correlations were observed between PM_{10} and OHMs ($r = 0.43$, $p < 0.001$), PAHs ($r = 0.20$, $p = 0.050$), and PCBs ($r = 0.19$, $p = 0.022$), with borderline statistically significant correlations between PM_{10} and CdTl ($r = 0.14$, $p = 0.065$) and PCDD/F ($r = 0.15$, $p = 0.052$) (Table 3). Statistically significant estimated coefficients from the multiple linear regression were observed between PM_{10} and CdTl ($\beta = 0.264$, $p = <0.001$), OHMs ($\beta = 0.305$, $p = <0.001$), PCDD/Fs ($\beta = 0.120$, $p = 0.008$), and PCBs ($\beta = 0.084$, $p = 0.045$) (Table 4). The partial η^2 are reported as percentages, and these showed modest overall explanatory power of the model variance after adjusting for MWI, flue, and year for CdTl ($\eta^2 = 4.99$), NOx ($\eta^2 = 5.46$), and OHMs ($\eta^2 = 14.25$).

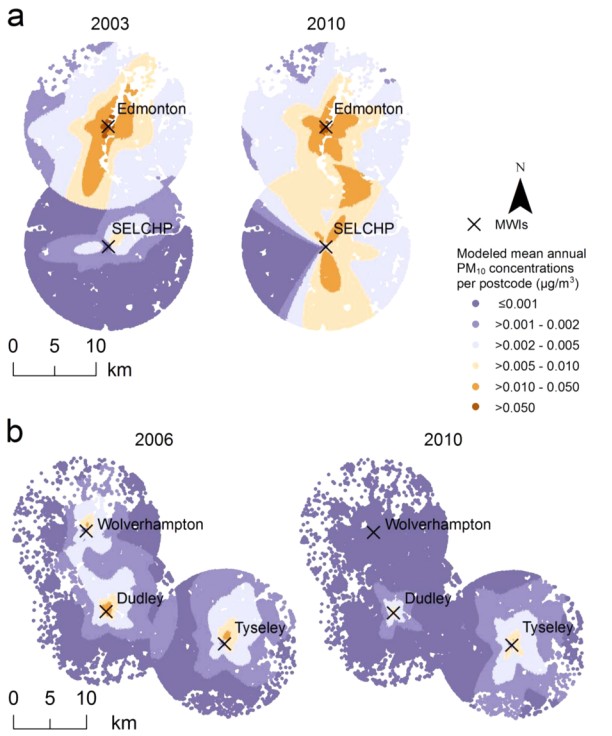

Figure 2. Exposure surfaces of the overlapping MWIs (a) SELCHP and Edmonton and (b) Dudley, Tyseley, and Wolverhampton comparing annual mean modeled PM_{10} ($\mu g/m^3$) at each postcode centroid within 10 km of each MWI in 2003 or 2006 and 2010. Note that there were no overlapping areas for (b) until 2006 as this is when Dudley MWI started operating (Table 1). SELCHP is an abbreviation for South East London Combined Heat and Power.

Table 3. Pairwise Spearman's Rank Correlation Coefficients and p-Values Comparing Monitored Pollutants[a]

	correlation (r)	p-value	no. of observations
CdTl	0.14	0.065	164
HGComp	0.11	0.181	161
OHMs	**0.43**	**<0.001**	**187**
PCDD/F	0.15	0.052	172
PAH	**0.20**	**0.050**	**100**
PCB	**0.19**	**0.022**	**147**
NOx	−0.04	0.591	197
SO_2	0.08	0.288	192

[a]Results with p-value < 0.05 are in bold.

Detecting Changes in Levels of PM_{10} Emissions Following the EU-WID. The descriptive analysis of the average modeled concentrations showed no clear pattern of a reduction in PM_{10} concentrations after the implementation of the EU-WID in MWIs adopting EU-WID specifications (Table 2, SI H, Figure S3), possibly as incinerators were already complying with EU-WID standards by the implementation date

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Table 4. Estimated Coefficients and p-Values from the Multiple Linear Regression Model, Where PM_{10} Is the Independent Variable[a]

	PM_{10} coefficient (β)	p-value	partial η^2 (%)	no. of observations
CdTl	**0.264**	**<0.001**	**4.99**	**360**
HGComp	0.051	0.218	0.63	374
OHMs	**0.305**	**<0.001**	**14.25**	**424**
PCDD/F	**0.120**	**0.008**	**0.17**	**430**
PAH	0.014	0.585	0.74	272
PCB	**0.084**	**0.045**	**0.03**	**357**
NOx	−0.096	0.619	5.46	437
SO_2	0.078	0.254	0.47	405

[a]Results with p-value < 0.05 are in bold.

which required them to fit bag filters. These descriptive findings were supported by findings in the Batch Change Detection statistical analyses (Table 5). In six of the 11 MWIs investigated, the change point occurred within a year prior or posterior to the EU-WID implementation in at least one flue (Coventry, Edmonton, Sheffield, Stockton-on-tees, Stoke-on-Trent, and Tyseley), and lower median PM_{10} emissions were detected after the change point in five of these (in seven out of 11 flues). However, in the remaining five MWIs the change point date was detected more than one year prior or posterior to the EU-WID implementation date (Bolton, Eastcroft, Porthmellon, SELCHP, and Wolverhampton), and a higher median level of PM_{10} was detected after the change point date in three of these MWIs (Eastcroft, Porthmellon, SELCHP), although still within EU-WID limits (Table 5). Figures showing monitored PM_{10} data, with the change points for each MWI included in the analysis, are presented in the Supporting Information SI K (Figure S4).

DISCUSSION

Our study of all 22 British MWIs in operation 2003−2010 indicates very low concentrations of incinerator-related PM_{10} within 10 km of the MWIs at postcode level (annual mean concentrations ranging from 1.00×10^{-5} to 5.53×10^{-2} $\mu g\ m^{-3}$). There were statistically significant correlations of PM_{10} emissions of heavy metals, PAHs, and PCBs ($r = 0.19$−0.43). A change point was detected in six of 11 MWIs adopting EU-WID specifications within a year prior or posterior to EU-WID implementation, but statistical analyses did not provide clear evidence of major changes in incinerator-related PM_{10} concentrations after implementation.

Dispersion Modeling To Estimate Concentrations of PM_{10} within 10 km of MWIs. The annual mean modeled PM_{10} concentrations from GB MWIs ranged from 1.00×10^{-5} to 5.53×10^{-2} $\mu g\ m^{-3}$ within a 10 km radius of the MWI. These contribute a small proportion of UK PM_{10} background levels, which range between 6.59 and 2.68×10^{1} $\mu g\ m^{-3}$ (annual UK means per postcode in 2010, based on modeled data).[28] As all European incinerators operate to the EU WID, this suggests that MWIs also make a small contribution to European background concentrations within 10 km of incinerators across Europe (measured ambient mean concentrations, typically in the range 2.00×10^{1}−5.00×10^{1} $\mu g\ m^{-3}$).[29] It is recognized that dispersion modeling is a simplification of reality. ADMS-Urban is a well validated, widely used dispersion model, and model errors were reduced as much as possible by using the most complete data available and by completing a series of sensitivity analyses to ensure that model inputs best represented actual conditions (see SI D, E, and F). Missing data were imputed using median values for the particular year, informed by results from a sensitivity analysis (SI D). For three incinerators in 2005, over two-thirds of the data were missing (Coventry, Stoke-on-Trent, Wolverhampton

Table 5. Dates When Changes in Emissions Were Detected in the Change Point Analysis Test Using the Cramér-Von Mises Test

MWI	flue	change point date (CPD)	no. of observations (before/after CPD)	median MWI in-flue PM_{10} measurements (mg m^{-3}) (interquartile range) (before/after CPD)
Bolton	1	17-10-2007	1751/1171	1.84 (2.00)/1.02 (1.83)
Coventry	1	24-05-2008	1606/951	2.00 (3.00)/1.00 (0.00)
	2	31-08-2008	1705/852	3.00 (4.00)/1.00 (0.00)
	3	21-05-2006	872/1685	4.00 (4.00)/2.00 (2.00)
Eastcroft	1	31-08-2009	2435/487	0.00 (0.50)/1.00 (0.00)
	2	08-07-2009	2381/541	0.00 (0.40)/1.00 (0.00)
Edmonton	1	27-01-2005	758/2164	3.20 (1.20)/2.13 (1.00)
	2	22-11-2006	1422/1500	1.80 (1.40)/2.78 (1.63)
Porthmellon	1	09-11-2007	1774/1148	2.00 (1.00)/5.00 (4.00)
SELCHP[a]	1	02-11-2009	2498/424	1.00 (1.00)/4.00 (2.00)
	2	06-08-2009	2410/512	2.00 (1.00)/4.00 (3.00)
Sheffield	1	24-01-2005	755/2167	0.10 (0.30)/0.20 (0.20)
	2	18-11-2004	688/2234	0.00 (0.10)/0.80 (0.80)
Stockton-on-Tees	1	29-09-2005	1003/1919	2.63 (3.84)/0.20 (1.00)
	2	29-09-2005	1003/1919	4.35 (2.41)/1.70 (1.50)
Stoke-on-Trent	1	28-11-2006	1428/1494	2.00 (2.00)/1.00 (1.00)
	2	21-01-2006	1117/1805	3.00 (2.00)/1.00 (0.00)
Tyseley	1	08-11-2006	1408/1514	1.10 (0.20)/1.80 (1.20)
	2	19-03-2008	1905/1017	2.50 (1.60)/1.50 (0.90)
Wolverhampton	1	30-04-2010	2312/246	4.00 (3.00)/1.00 (0.00)
	2	04-06-2004	156/2402	6.00 (2.00)/2.00 (3.00)

[a]South East London Combined Heat and Power.

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(SI C)), although annual mean modeled concentrations were still within similar ranges compared to other MWIs (Table 2). We do not know the reasons for missing data, but this may represent maintenance periods in 2005 to ensure the MWI complied with the WID implemented at the end of that year.

All MWIs used moving grate technologies except for Allington, Dundee and Newlincs (Allington and Dundee used fluidized bed technology and Newlincs used rotary kiln technology).[30] Nixon et al.[30] found that plants using fluidized bed and rotary kiln technologies had higher emissions of HCl and CO. We found no differences in PM_{10} emissions from Allington and Dundee (mean (standard deviation (SD) 2.30 (3.05), median 1.02) and Newlincs (mean (SD) 2.44 (1.27), median 2.30) compared with the remaining 19 MWIs (mean (SD) 2.10 (2.13), median 1.50).

Results for GB incinerators are consistent with two studies conducted in Italy (also operating to the EU-WID) by Candela et al.[13,14] and the previous study conducted in GB by Ashworth et al.[19] Concentration estimates were larger in Font et al. (3.00 $\times$ 10^{-2} to 1.20 $\times$ 10^{-1} μg m^{-3});[31] however, they did not measure PM_{10} directly but used tracers of heavy metals to estimate maximum ambient PM_{10} from two MWIs. The Font et al.[31] study used Cd measured during plume grounding as a quantitative tracer for PM_{10} by multiplying measured ground-level Cd concentrations by representative in-flue PM_{10} to Cd emission ratios. This approach set out to find a maximum value by assuming that all Cd was from the MWI.[31] Our findings for PM_{10} are in agreement with studies on ultrafine particles involving measurements within MWI flues and ambient air, showing that incinerators do not have significant impacts on ultrafine particles in localities near MWIs.[32]

Additional work was undertaken to confirm the plausibility of the very low modeled PM_{10} concentrations. MWI emissions were fingerprinted using daily in-flue PM_{10} to NOx concentrations, and ratios were compared to data from 15 ambient monitoring sites within 10 km of four MWIs (Edmonton, SELCHP, Tyseley, and Wolverhampton) (SI L, Table S10). Results showed that while there was some evidence of NOx and PM_{10} emissions from MWIs being detected at ground level, these were few and often could not be distinguished from other sources such as traffic (SI L, Figure S5). This supports the very low contributions of MWI PM_{10} to background concentrations in areas near MWIs in the present study.

Exposure surfaces for selected MWIs in Figure 2 that have been previously presented[19] show that incinerator-related PM_{10} concentrations were not merely a function of distance from incinerator but showed complex spatial patterns including differences between years, largely relating to differences in emission rates (including off days) and meteorology.

Consideration of Other Pollutants Emitted from MWIs. While ambient PM_{10} has been associated with adverse birth outcomes,[33] levels are much higher than those arising from MWIs emissions. Despite this, some recent epidemiological studies relating to MWIs operating to the EU-WID have found associations with adverse birth outcomes.[13,14] If these are causal associations, it is likely to be due to agents other than PM_{10} that are also emitted from incinerators such as PCDD/Fs, PAHs, and heavy metals. We were unable to model spatial distribution of these other agents directly due to sparse emissions data. Other potential incinerator emissions including polybrominated or mixed polybrominated/polychlorinated dibenzo-*p*-dioxins/furans (PBDD/Fs and PXDD/Fs) were not measured. However, it is a reasonable assumption that modeled spatial distribution of PM_{10} reflects exposure patterns of other MWI emissions. This assumption has been used in previous dispersion modeling studies, which found that heavy metals[14] had a similar deposition distribution to PM_{10}. Ranzi et al.[24] measured various pollutants including sulfur oxides, nitrogen dioxide, and heavy metals in Italy at maximum and minimum fallout points estimated by dispersion models and considered heavy metals as the tracer pollutant from MWIs. We found some support for this as we detected significant correlations for in-flue measurements between PM_{10} and heavy metals, PAHs, and PCBs, which provides some support for using PM_{10} as a tracer. While statistically significant, the amount of variance accounted for (partial η^2) was modest, which is likely due to variability in incinerator feedstock, especially differing amounts of electrical equipment. Information on feedstock mix is not recorded by MWIs.

The level of population exposure to metals and other agents from MWIs is likely to be small. Font et al.[31] compared heavy metal emission ratios with those measured at nearby ambient metal monitoring sites around six MWIs in England and found limited evidence that emissions from MWIs reached ground level.

Emissions above the EU-WID Daily Average Particulate Limit Value. Although emissions greater than the EU-WID limit of 10 mg m^{-3} were found in 14 of the 22 MWIs, these were usually <20 mg m^{-3} (SI J Table S7). These may not all represent exceedances under the EU-WID as in the event of temporary abatement failure MWIs are allowed to operate for up to 4 h at a time (maximum 60 h per flue per year) at an elevated half-hourly particulate limit value of 150 mg m^{-3} (normally 30 mg m^{-3}). If there are less than 43 half-hourly monitoring results available in a day, the daily average can be disregarded. Daily average emissions >20 mg m^{-3} were infrequent, and there were only rare occurrences >30 mg m^{-3}, which may have occurred due to "one off" changes in feedstock or failure of abatement systems. We were not provided with information on reasons for emissions greater than the EU-WID limit. However, given that mean PM_{10} concentrations estimated by the dispersion model were small (1.00 $\times$ 10^{-5} to 5.53 $\times$ 10^{-2} μg m^{-3}, a small contribution to ambient background levels which were typically 6.59−2.68 $\times$ 10^{1} μg m^{-3}), these infrequent emissions above EU-WID limits would still be expected to result in very low population exposures.

Detecting Changes in Emissions Following the EU-WID. We conducted the change point analysis for existing incinerators using the Cramér-von Mises method to account for the ordered data structure. A simpler test (e.g. a two sample *t* test to compare PM_{10} emissions before and after the EU-WID implementation) may have introduced bias due to the number of nonoperational, missing days and non-Gaussian distributed data. We assumed that a fall in emissions would be detected in existing incinerators within one year (prior or posterior) of the EU-WID implementation date, but this was only seen for five of 11 incinerators in the change point analysis. A possible explanation is that many existing MWIs may have already met (or been modified to meet) the EU-WID requirements. However, information as to whether and when each MWI adopted a new abatement system was not available. In three of the six MWIs where a change point was detected within a year prior or posterior to the EU-WID implementation date (28 December 2005), a higher mean level of PM_{10} was detected

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after the change point date in at least one flue (Edmonton, Sheffield, and Tyseley), though the increases after the change point date were small, and remained below EU-WID limits. This could be related to a number of factors including differences in the feedstock or changes in the amount of waste processed over time. Since we could not identify a clear date after which emissions fell in relation to the EU-WID in pre-existing MWIs, we conclude it is not possible to conduct before/after epidemiological studies examining the impact of the EU-WID on rates of adverse health outcomes in pre-existing MWIs. However, in MWIs opening after 28 December 2002 (n = 8 in 2003−2010) that have always operated to the EU-WID standards, it is possible to use the opening date of the incinerator as the before/after change point date.

Overall this study suggests that PM_{10} exposures related to MWI emissions in Great Britain are extremely low (annual means ranging from 1.00×10^{-5} to 5.53×10^{-2} $\mu g\ m^{-3}$) especially when compared to annual mean background concentrations (typically ranging between 2.00×10^{1} and 5.00×10^{1} $\mu g\ m^{-3}$ in Europe).[29] The results of the modeling will be used in an epidemiological analysis examining associations between MWIs and potential reproductive and other health effects.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b06478.

(A) Municipal Waste Incinerator (MWI) characteristics, (B) non-numeric and negative PM10 emissions value coding, (C) Operational, nonoperational, and missing days, (D) Sensitivity analysis: missing data imputation methods, (E) Meteorological data selection, (F) Monin-Obukhov and Surface Roughness length input values, (G) Non-continuous measurements, (H) Mean modeled PM10 concentrations (μg m-3) per MWI that adopted EU-WID specifications, (I) Change point analysis equation, (J) Emissions above the EU-WID daily average particulate limit value, (K) Change point analysis results, (L) Fingerprinting NOx to PM_{10} ratios from MWI in-flue concentrations (PDF)

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Notes

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