A Review of the Impact of Domestic Combustion on UK Air Quality

Mitchell, E.J.S., Cottom, J.W, Phillips, D., and Dooley, B.

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Contact:

Bruce Allen CEO, HETAS.

Newtown Trading Estate,

Unit 5 Severn House,

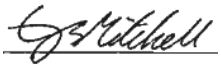
Green Lane,

Tewkesbury

GL20 8HD

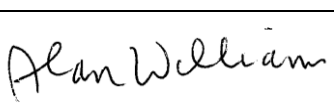
Bruce.Allen@hetas.co.uk

Authors:

Edward J. S. Mitchell PhD	
Joshua Cottom PhD	
Douglas Phillips PhD	
Ben Dooley PhD	

The authors declare no conflict of interests in the work carried out.

Reviewed by:

Professor Alan Williams CBE, FREng University of Leeds	
Professor Jenny Jones FRSC, MEI University of Leeds	

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Executive summary

Despite anthropogenic particulate matter (PM) emissions declining within the UK in recent years, the need for obtaining accurate and reliable estimations of their quantity and sources remains vitally important owing to the severe health impacts they cause. One such source is that of domestic burning, defined as the combustion of fossil fuels and biomass in residential appliances. This therefore encompasses boilers, barbeques, bonfires and solid fuel stoves, as well as many more.

In January 2019, The UK Clean Air Strategy stated that domestic burning is the main contributor to the UK's primary PM emissions, with solid-fuel stoves receiving particular scrutiny. However, the true contribution of domestic burning to air pollution remains poorly constrained and model uncertainty remains high, particularly in comparison to other emission sources.

The aim of this report is to give a high-level critical review of the contribution of domestic burning to UK air pollution with the intention to better understand the uncertainties of current estimates. This is achieved by reviewing existing literature and methodologies in relation to ambient air source apportionment methods, primary and secondary emissions, activity data, PM emission contribution estimates, associated emission factors, and the potential influence of other known emission sources.

Summary of findings

Top-down source apportionment

There are two common apportionment methods for domestic burning, aethalometers and levoglucosan; however, these cannot distinguish definitively between different biomass burning sources. The majority of aethalometers used for quantitative monitoring within the UK carry high levels of uncertainty due to large amounts of variation within the instruments default values. Although there are improved versions of aethalometers which are much less prone to interference, these have not widely been applied in the UK.

Additionally, levoglucosan, which is produced by the pyrolysis of cellulose, is often used as a marker for wood-burning stove use. However, as cellulose is found in all type of biomass, this should actually be considered a marker for biomass burning in general, with the specific source not easily identifiable. Other potential sources of biomass burning include wildfires, prescribed burning, bonfires and barbecues to name a few. Data is presented that suggests these alternate sources are common in the UK and can lead to substantial PM emissions, and therefore should be considered alongside solid-fuel stoves in top-down source apportionment studies.

Levoglucosan emission factors may also vary by a factor of 5 or more depending upon the fuel and appliance type. Despite this, a fixed ratio of 10.7 is commonly used in source apportionment studies, derived from a study using what is now a 25 year old Austrian stove. Considering all of these factors, it is believed that source

apportionment of domestic burning sources has large uncertainty and therefore it is recommended more accurate techniques be used.

Primary and Secondary Emissions

The majority of pollution sources emit PM both directly in the atmosphere (primary PM), and precursors which react in the atmosphere to form PM (secondary PM). The primary PM emissions from advanced stoves and boilers (e.g. Ecodesign compliant) can be reduced by nearly 90% when compared to an open fireplace. Additionally, the toxicity of the PM can also be reduced; which for advanced stoves, can be by as much as 3 times compared to an open fire. Advanced stoves therefore have a cumulative impact in both decreasing direct emissions and decreasing their toxicity.

For secondary PM on the other hand, the release of precursor gases (e.g. VOCs and NOx) can significantly increase the mass of PM emitted from stoves. However, efficient modern appliances can see these reduce to near zero during stable combustion conditions. Moving to the sale of only Ecodesign compliant appliances will therefore likely provide air quality benefits, as would reducing the numbers of existing open fires and older inefficient stoves.

Bottom-up source apportionment

For bottom-up source apportionment methods, data on the estimated use (activity data) is combined with emission factors to determine the total emissions over a period of time. Within the UK, the National Atmospheric Emission Inventory (NAEI) is the body responsible for this. They estimated wood burning increased from 1.8m tonnes in 2005 to 4.8m tonnes in 2017, based on data derived from the Government's *Domestic Wood Use Survey*. Applying seasonal regression, this estimate equates to 16.7 kg of wood burned per household per day during the peak heating months. This calculated wood consumption appears to be considerably out of proportion, greatly exceeding that from other countries, and when compared to other more recent industry surveys in the UK, it equates to more than double their reported value. If the recent lower estimate is applied, the total contribution of stoves to PM drops to 9.6% for PM_{10} and 14.9% for $PM_{2.5}$.

Additionally, the $PM_{2.5}$ emissions factor as reported by the NAEI, is three times higher than the limit given under new Ecodesign regulations. As there is existing evidence which demonstrates that appliances are capable of meeting the Ecodesign emissions limits, it is possible that the NAEI's emissions factors are currently overstating the impacts of domestic burning. As the UK's stove appliance types are likely to differ greatly in terms of model, age and pollution control features, further evidence is required to ensure that the emissions factors applied in the NAEI are representative.

Conclusions and recommendations

In conclusion, this report reviews the methods and uncertainties behind the contribution of domestic burning to PM emissions in the UK. Evidence presented indicates that the 38% contribution shown in the UK 2019 Clean Air Strategy, described as arising from domestic burning, has large uncertainties in many of the underlying methods and data. The cumulative effect of these uncertainties means that the 38% value is highly likely to be inaccurate. Thus, more reliable methods and further analysis is needed.

It is therefore imperative that thorough, evidence-based analysis be given to the methods, assumptions and limitations behind the calculation of the domestic burning contribution to the UK's emissions. Key recommendations and areas of future work are as follows:

[1]. Appliances & Monitoring

- **Improved Monitoring Equipment**; support the extensive uptake and utilisation of more accurate measurement techniques, including 7 (seven) wavelength aethalometers, chemical mass balance and positive matrix factorisation. Additionally, it is important that the fixed ratio methods for attributing levoglucosan concentrations are further investigated to ensure their validity when applied within the UK.
- **Review Existing Inventory Datasets**; it is vital that the existing activity data and emissions factors used are revised to properly represent the emissions associated with domestic burning.
- **Stove and Open Fire Upgrades**; replacing older, less efficient stoves and open fires with new models – that meet Ecodesign regulation – is a simple and effective method for reducing the current PM emissions associated with domestic burning.

[2]. Policy & Research Support

- **Research Support into Other Emission Sources**; the correct implementation of air quality policy decisions first requires the robust quantification of all significant sources of pollutants. It is vital that research is conducted into the use of other sources such as barbecues, fire pits, commercial cooking, wildfires and open burning.
- **Domestic Burning Emissions Expert Panel**; establish a cross-sector expert panel, helping to bring industry, academia and policymakers together, facilitating greater collaboration within the area of domestic burning.
- **Increased Funding Sources**; improve the availability of funding for local authorities to properly investigate and monitor their local air quality conditions, cooperating with researchers and industrial partners to ensure

that the correct actions are initiated. This should also extend to engagement activities within their local communities.

[3]. Public Perception

- **Knowledge Generation**; the public are currently largely uninformed on the impact of domestic burning. Ensuring that accurate evidence-based information is disseminated and made openly available to the public should take precedence.
- **Outreach Programmes**; effectively promote the 'best practices' for stove use and other domestic burning appliances. This should include correct methods for stove use and advice on wood fuel quality.

[4]. Take strategic & targeted action to reduce domestic burning emissions

- **Apply regulation to the most polluting sources**; appliances used for outdoor heating and cooking such as barbecues, chimeneas, pizza ovens, firepits and garden incinerators pose a health hazard if they continue to be unregulated. They are also an extremely inefficient use of fuel in comparison to modern Ecodesign-ready stoves and boilers.
- **Support replacement of open fires and traditional stoves**; target the 27.4% of appliances which are open fires or stoves older than 10 years.
- **Support consumers to choose the lowest emission appliances**; consumers currently have no incentive to purchase an appliance that exceeds Ecodesign limits. Providing some incentive will promote competition and the greater uptake of automatic pellet stoves and top-of-the-range wood stoves.
- **Encourage users to burn only high quality renewable fuel;** 52% of users burn wood only, transitioning the remaining users away from coal to smokeless fuels will improve air quality, and towards wood and biomass will reduce greenhouse gas emissions.

[5]. Consider the role of domestic biomass burning in climate and air quality targets

- If heat pumps are widely prioritised for domestic heating, particularly in off-gas grid properties, there may be a need for **low-cost supplementary heating during the winter months**. Modern high-efficiency low-emission biomass stoves have the potential to supply this supplementary heat demand either as a separate appliance or a hybrid heating system.
- 2030 emissions reductions obligations under the NEC Directive and revised Gothenburg Protocol are unlikely to be met without targeted action in the domestic burning sector. The same is true of achieving WHO guideline $PM_{2.5}$ concentrations.

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1. Background

1.1 Introduction

In January 2019 the UK Clean Air Strategy (DEFRA, 2019a) suggested that the "increase in burning solid fuels (wood and coal) in our homes (domestic burning) is having an impact on our air quality and now makes up the single largest contributor to our national PM emissions at 38%". Legislation established in the 1950's to combat the high level of PM emissions at the time do not necessarily align with the today's emission control requirements. Emissions from domestic burning have significantly decreased since the 1950's but we now have a better understanding regarding the impact of stove and open fire use in homes, how pollution from domestic burning can travel through the atmosphere, and the resulting health impacts on the local and wider population.

Domestic burning is defined as the combustion of fossil fuels and biomass in residential appliances, usually for heat generation. There are a number of sub-categories within the domestic burning sector with emissions attributed to stationary or mobile sources. These categories, along with the technologies typically seen in each, are shown in Figure 1 and are discussed further in 5.1. The identification number,1A4b reported in Figure 1 is derived from the Intergovernmental Panel on Climate Change (IPCC) common reporting framework (IPCC, 1996) and the identifications i and ii are in line with the National Atmospheric Emissions Inventory (NAEI) categorisation (NAEI, 2019).

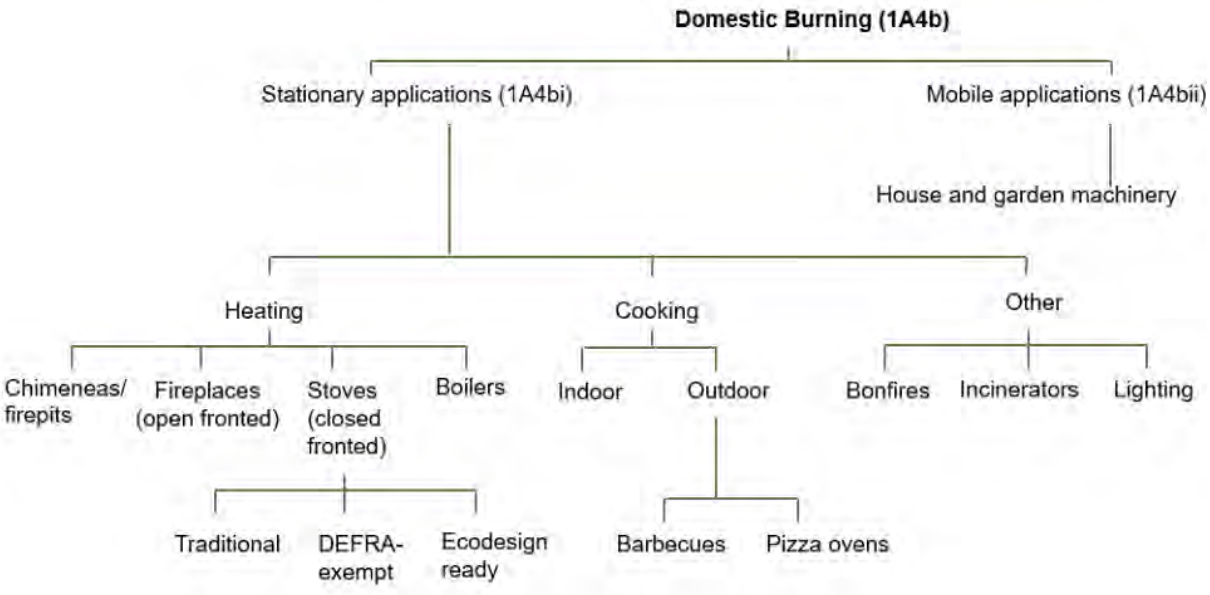


Figure 1. Domestic burning sectors in line with IPCC and NAEI notation and the associated emission sources

The focus of this report is the combustion of residential solid fuels (RSF) in the United Kingdom, a subcategory of domestic burning, and its contribution to the 38% value used by DEFRA.

The 38% value is derived from the NAEI using a bottom-up approach and is constrained by source apportionment studies of ambient concentrations (a top-down approach). Two key studies were used to inform the Clean Air Strategy, notably Font and Fuller (2017) and Stewart (2017). Font and Fuller (2017) reported that wood burning accounts for up to 31% of local urban derived $PM_{2.5}$ in London while Stewart (2017) performed a series of comparative round robin tests to determine the uncertainty in PM emission measurements.

Despite a number of recent high-profile reports, the true contribution of domestic burning to air pollution remains poorly constrained and model uncertainty remains high in comparison to other emission sources.

1.2 Air pollution and air quality in the UK

Air pollution in the UK has been declared a “public health emergency”. Both the UK High Court and the European Court of Human Rights have ruled that there is an urgent need to improve UK air quality and that current policies to address the problem are “unlawful”. Following a legal battle with activist organisation ClientEarth, the UK government is facing multimillion-euro fines unless drastic action is taken to improve air quality. The Department of Health and Social Care’s advisory Committee on the Medical Effects of Air Pollutants (COMEAP) recently estimated that long term exposure to man-made air pollution could be linked to 28,000 – 36,000 premature deaths (COMEAP, 2018), highlighting the need to drastically improve national air quality.

Figure 2 shows the trends in annual air pollution in the UK from 1970 to 2017. In recent times there has been a decline in all emissions, with some control measures impacting multiple pollutants. The main source of ammonia emissions is the agricultural sector (87% of total ammonia emissions in 2017) with additional emissions from the waste and road transport sectors. The emissions associated with ammonia decreased slightly during the 80’s and 90’s, with the largest decline seen in the late 90’s through to 2010. Since 2013, ammonia emissions have started to increase driven by the agricultural sector.

Non-methane volatile organic compound emissions peaked in 1990 after which emissions controls in vehicles (a major source in the early 90’s), the reduction in coal, the refining of petroleum fuels and stricter emissions limits in industry (solvent use) have resulted in a significant reduction.

Nitrogen oxides also peaked in 1990 but the introduction of vehicle emission controls in the late 80’s, the shift to natural gas from coal and oil in electricity production and stricter industrial combustion plant emission limits have resulted in the decline seen post 1990.

The deployment of flue gas desulphurisation (FGD) units in coal fired power stations since the 70's and the shift to gas, from sulphur containing coal, in the 90's resulted in the decline of sulphur dioxide emissions seen since 1970.

The trends in PM emissions have been steadily reducing since 1970 with PM_{10} and $PM_{2.5}$ showing the same trends. The decline has been driven by the move away from the domestic burning of coal in open fires in the 70's and 80's. The reduction in vehicle exhaust emissions due to stricter emissions standards introduced in 1996 have also contributed to the decline in total PM although the increase in traffic volume an increase in non-exhaust emissions (tyres and brakes) have partially offset the reduction. PM from industrial sources have also been reduced due to the decline of the manufacturing industry and improvements in emission controls (DEFRA, 2019b).

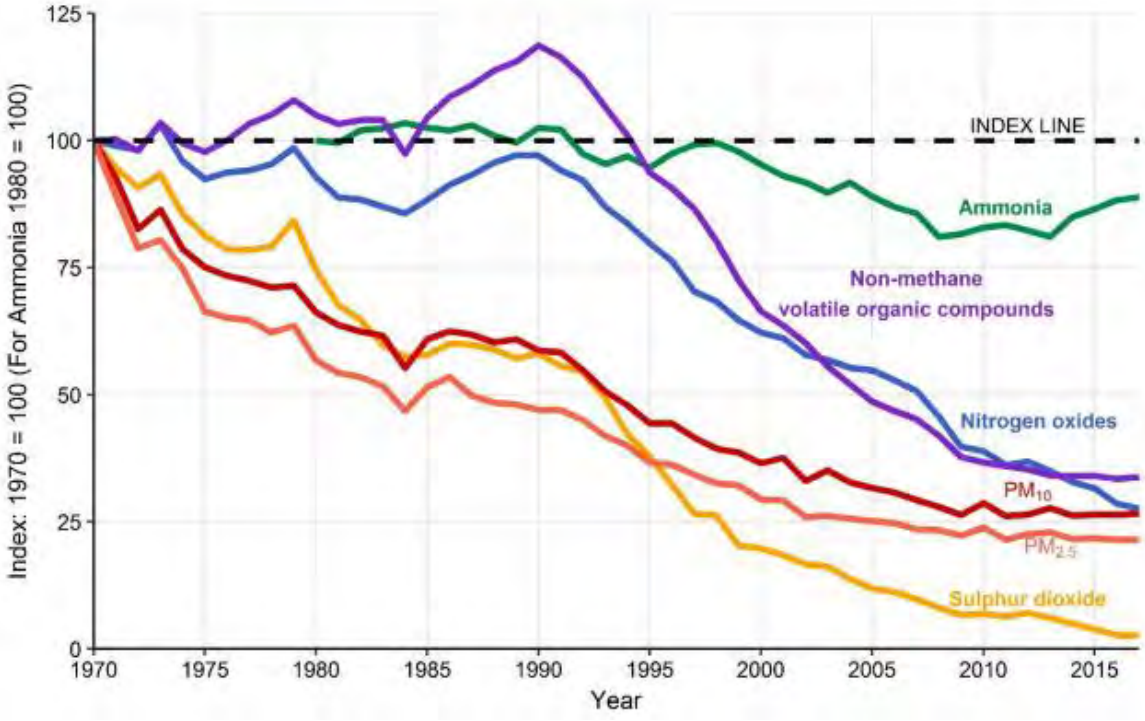


Figure 2. Trends in annual air pollutants in the UK between 1970 and 2017 (DEFRA, 2019b)

The government's Clean Air Strategy was published to "set out the case for action and to demonstrate the government's determination to improve air quality" with a new goal that considers the World Health Organisations guidelines. In relation to PM emissions, the main focus of this report, the government set additional reduction targets of 30% by 2020 and 46% by 2030 against a 2005 baseline (DEFRA, 2019a). The Clean Air Strategy suggests that 38% of current PM emissions are the result of "burning wood and coal in open fires and stoves" without consideration of the other domestic burning sources highlighted in Figure 1. As the estimated leading source of PM emissions in

the UK, and as a result of the new PM targets set, domestic burning of solid fuel is being targeted in the following ways:

- Consideration of increased powers for local authorities
- Only the cleanest Ecodesign stoves will be sold by 2022
- Prohibiting the sale of 'dirty' fuels like wet wood and high sulphur fuels
- Ensuring that new fuels derived from wastes and recycled products are safe to use
- Raising awareness and educational campaigns

1.3 Biomass, bioenergy and the decarbonisation of heat

Biomass is set to play a significant role in the UK to help meet the Committee on Climate Changes recommended target of net zero greenhouse gas (GHG) emissions by 2050, which has recently been ratified by the Government. The heating sector is a major contributor to UK emissions due to the burning of natural gas and heating oil in boilers, but is also a significant contributor to air pollution, primarily due to the burning of solid fuels in low temperature combustion applications. Estimated emissions attributable to heating accounted for 37% of total UK GHG emissions (468 $MtCO_2e$) in 2016, with space heating and cooling accounting for 17% of the total, followed by industrial processes (14%), and the remainder made up of hot water (4%) and cooking (2%) (BEIS, 2018).

Greenhouse gas emissions from heat are the single largest contributor to total UK emissions (BEIS, 2018). The decarbonisation of heat is therefore one of the greatest challenges in climate change mitigation and requires rapid and widespread deployment of renewable heating technologies in order for the UK to achieve its greenhouse gas reduction targets.

Historically, British households have relied heavily on solid fuels for space heating. In the 1950's, most homes had an open fire burning a combination of wood and coal. Emissions were generally unabated and unregulated, with the burning of poor-quality fuel such as high-sulphur coal leading to severely reduced air quality and 'great smogs' such as that in London in 1952. In response, the Clean Air Act of 1952 established smoke control areas, and together which a wide scale uptake of natural gas heating, solid fuel consumption has continued to decrease. Since 1990, solid fuel consumption has reduced by over 86% whereas gas consumption has remained steady, as shown in Figure 3.

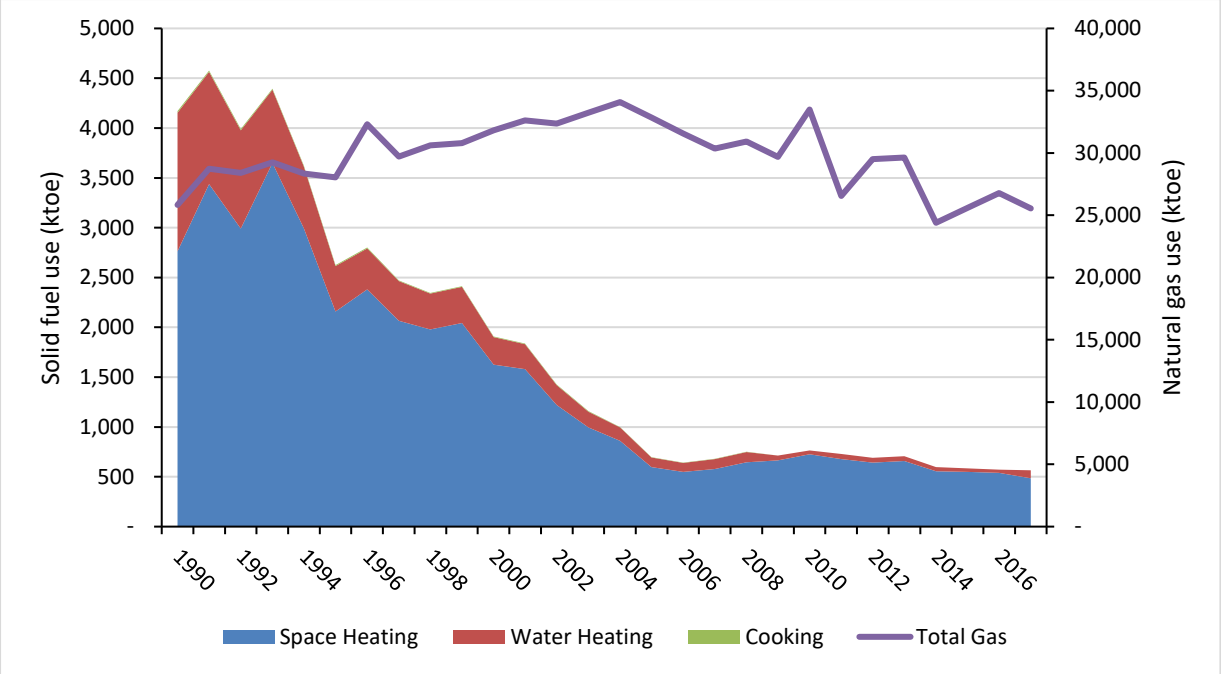


Figure 3. Domestic consumption of solid fuel and natural gas in the UK between 1990 and 2017 (DEFRA, 2019c)

Natural gas use in 2017 was a factor of five higher than solid fuel consumption and is the dominant source of greenhouse gas emissions in the domestic sector. Annual emissions vary from year to year due to weather conditions, but greenhouse gas emissions from residential combustion have reduced slightly from 78.3 $MtCO_2e$ in 1990 to 64.1 $MtCO_2e$ in 2017. The Committee on Climate Change (CCC, 2018) found that temperature-adjusted GHG emissions from the buildings sector rose in 2017 for the second consecutive year, whilst those in most other sectors reduced.

Modern bioenergy (high efficiency conversion technologies) is forecast to constitute 30% of growth in renewables over the next five years and is the “overlooked giant within renewable energy” (IEA, 2018). The term ‘modern bioenergy’ does not include residential wood burning in the IEA’s forecasts, citing environmental and sustainability issues arising from traditional biomass use in developing countries. The IEA defines modern biomass as that which “includes all biomass (such as biodiesel), with the exception of traditional biomass which include wood, charcoal, agricultural resides and animal dung used for cooking and heating.”

In the UK, traditional open fires and older simple stoves cannot be considered modern bioenergy. However, there are a number of domestic systems which it can argued do fit the criteria for modern bioenergy. Examples include log-gasification boilers and advanced stoves such as automated pellet stoves. These technologies are currently eligible for the domestic renewable heat incentive (RHI) and account for 51% (1,810 GWh) of renewable heat generated under the domestic RHI (BEIS, 2019).

1.4 Structure of report

The aim of this report is to provide a high-level critical analysis of the methodologies used to determine the contribution that domestic burning has towards total atmospheric PM emissions. The report reviews several key areas. In section 2 the source appointment methods are reviewed, this includes analysis of the PM measuring techniques, a review of a wide range of global domestic burning contribution studies, analysis of primary and secondary emissions accounting methods, and analysis of the impacts of particle toxicology.

Section 3 focuses on emissions inventories for domestic burning and includes a review of domestic burning activity estimates, a review of annual and diurnal trends in PM emissions and the reasoning behind them, and a review of the emission factors used in emission modelling.

The final section investigates alternate sources of biomass burning and their scale to better understand how they may contribute to overall PM emissions and the uncertainty they produce when considering the contribution of domestic burning on total PM levels.

The investigation in each of these areas highlights uncertainties present in the 38% value reported by DEFRA (2019a).

2. Ambient air source apportionment for domestic burning pollution sources

2.1 Source apportionment methods for wood burning

A variety of methods are available for the source apportionment of residential solid fuel smoke in atmospheric plumes. This section outlines some of the most commonly used methods and the theory behind them, building on information presented by Mitchell (2017).

2.1.1 Aethalometers

An aethalometer is an instrument that measures the absorption of specific wavelengths of light through a sampled aerosol collected on a filter. The filter is held on a spool which allows the instrument to measure black carbon concentration in ambient air in real-time.

Aethalometers are used to measure black carbon in the UK through the UK Black Carbon Network, which is operated by the National Physical Laboratory (NPL). The two-wavelength Magee AE22 is used by the network to measure black carbon and "*to indicate the presence of aromatic organic compounds such as are found in wood smoke, biomass-burning smoke, and tobacco smoke*"[1].

Figure 4. The Magee AE22 aethalometer

There are currently 14 sites across the UK in the Black Carbon Network which are summarised in Table 1.

[1] Further detail available at https://uk-air.defra.gov.uk/networks/network-info?view=ukbsn

Table 1. Monitoring sites in the UK Black Carbon Network

Site name	Site type	Surrounding area	Likelihood of RSF
Auchencorth Moss	Rural background	Farmland	No
Ballymena Ballykeel	Urban background	Middle income residential	Yes
Belfast Centre	Urban background	City centre	No
Birmingham A4540 Roadside	Urban traffic	Major roadside, lower/middle-income residential	No
Birmingham Ladywood	Urban background	Lower income residential	No
Cardiff Centre	Urban background	City centre	No
Chilbolton Observatory	Rural	Arable farmland	No
Detling	Rural background	Farmland	Possible – 1 nearby house
Glasgow High Street	Urban traffic	Major roadside, city centre	Unlikely – flats
Glasgow Townhead	Urban background	Lower income residential	No
Kilmakee Leisure Centre	Urban background	Middle income residential	Yes
London Marylebone Road	Urban traffic	Major roadside, city centre	No
London North Kensington	Urban background	Affluent residential	Yes, high
Strabane 2	Suburban background	Middle income	Yes

Two-wavelength aethalometers measure light absorbance at 880 nanometre and 370 nanometre wavelengths, and then calculate the mass of black carbon (BC) in the sample using well understood scientific principles. However, the equation used has two constants which must be assumed based on the likely air pollution sources in the sampling area:

- the mass extinction coefficient
- the Ångström exponent

The Ångström exponent is an important parameter in source apportionment studies and many experiments have been conducted to calculate this source-specific

parameter for different pollution sources, including diesel vehicles and solid fuel combustion.

The majority of fossil fuel combustion sources emit soot particles that are very dark in colour, i.e. they have a high level of black carbon and lower levels of brown carbon. The absorption Ångström exponent for these sources is close to 1.0. Soot particles with a high level of organics, known as brown carbon or tar balls, have a higher Ångström exponent, up to 6 or more. The Ångström exponent for RSF combustion sources is variable and can be anywhere within the range of 1-7, depending on fuel type and combustion conditions.

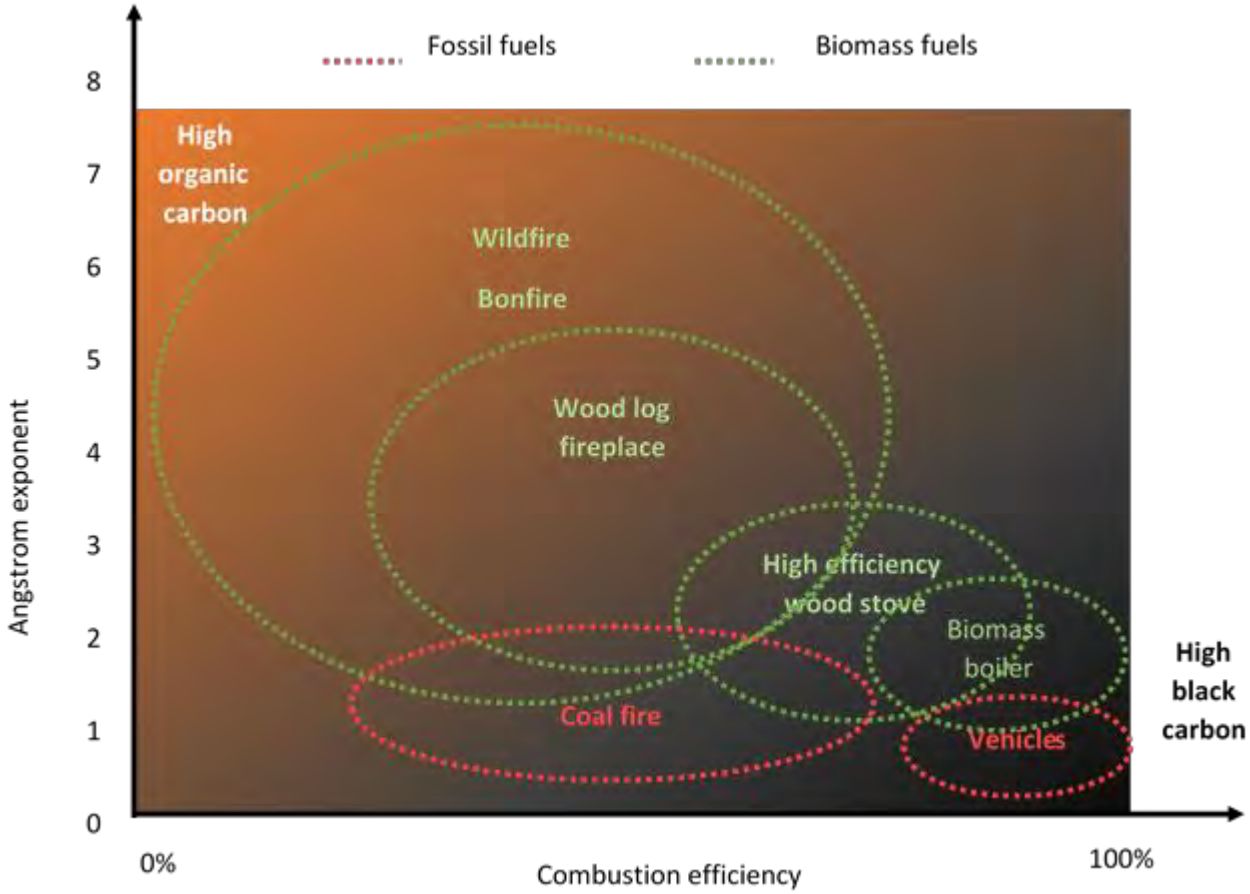


Figure 5. Impact of fuel type, combustion efficiency and organic carbon content on the Angstrom exponent used in source apportionment with aethalometers.

Font and Fuller (2017) use Ångström exponent values of 0.96 for traffic and 2.0 for wood burning. These values are supported from previous studies as an average for biomass burning (Favez et al., 2009, Sandrewi et al., 2008a, Zotter et al., 2014). Hence this method alone cannot differentiate between biomass burning sources. Values as low as 1.2 have been recorded for fresh PM and as high as 6 for wildfire smoke. Martinsson et al. (2015) showed that the Ångström exponent is dependent on the phase of combustion and stayed largely between 1.0 and 1.5 when burning birch logs

in a 9 kW stove. The value only reached 2.5 during fuel addition and during the burnout phase. Other contributors to high organic emissions include high moisture fuel, an overloaded combustion chamber and starved air supply. Soots emitted from residential burning of other solid fuels have similar Ångström exponents of 1.0-2.8 for coal (Bond et al., 2002) and up to 7.7 for peat (Pokhrel et al., 2016). Aging of smoke plumes has also been shown to increase the value of the Ångström exponent up to 8.3 (Hecobian et al., 2010). An overview of the impacts of different biomass types and aging periods on the optical properties of wildfire smoke is given in Shi et al (2019), which found that wood burning PM had a much higher fraction of black carbon than herbaceous biomass burning PM.

Qualitative source apportionment can be achieved using multi-wavelength aethalometers using the difference between BC determined at 370 nm and 880 nm. Absorption in the UV (370 nm) is interpreted as an indicator of aromatic organic compounds, or brown carbon, which is often attributed to biomass smoke. The largest differences in absorption between the 370 nm and 880 nm have been observed at evenings and weekends in the winter months, which again is usually attributed to domestic wood burning (Wang et al., 2011a, Wang et al., 2011b) (Herich et al., 2011, Sandradewi et al., 2008b, Favez et al., 2009)

More sophisticated aethalometers are now available which measure at seven wavelengths; 470 nm, 520 nm, 590 nm, 660 nm and 950 nm in addition to 370 nm and 880 nm. These instruments are used for more detailed studies of aerosol optical properties and models such as the Magee AE33 boast the ability to give real-time source apportionment for biomass burning (Arnott et al., 2005, Sandradewi et al., 2008c).

Strengths and weaknesses

The aethalometer is a useful tool to determine the concentration of absorbing particles (black carbon and brown carbon) in polluted atmospheres. The differences in absorption at different wavelengths is also very useful for studies of aerosol optical properties, which can be used in source apportionment. There are clear differences in the optical properties of PM samples taken from vehicles and PM samples taken from biogenic sources (cooking, wildfires, wood stoves, agricultural burning etc.). However, the aethalometer alone cannot differentiate between the different biogenic sources. There are several significant sources of light absorbing organic carbon (OC) in the UK atmosphere which may contribute to uncertainty in biomass burning source apportionment with aethalometers. For example, at the North Kensington air pollution monitoring site, source contributions of OC were found to be 21% vehicle exhausts, 11% food cooking and 15% wood smoke (Yin et al., 2015). Though not all of this will have been light-absorbing OC, it demonstrates that there may be other sources contributing to absorption at the lower wavelength (370 nm) which cannot be exclusively attributed to domestic burning.

There are several known issues associated with the use of aethalometers for source apportionment, including interferences from loading effects, thermal stability issues and sample matrix effects. Some studies emphasise that the ratio is mostly indicative and should not necessarily be used for empirical apportionment of wood-smoke in PM (Harrison et al., 2012). However, it can be used in conjunction with other tracers and a number of corrections to data have been proposed, such as filter loading bias (Weingartner et al., 2003, Virkkula et al., 2007, Cheng and Yang, 2015). A number of algorithms have been developed to correct for measurement bias but are usually tailored to given conditions and a given pollution source – an overview the different correction algorithms is given in Collaud Coen et al. (2010) and more recently in Saturno et al (2017).

The use of 7 wavelength aethalometers rather than the currently used 2 wavelength aethalometers may reduce uncertainty. According to Harrison et al. (2012), there are "*many facets to* [UK air quality] *data which cast doubt on whether the instrument* [2 wavelength aethalometer] *is reliably reflecting concentrations of woodsmoke*". Using a combination of a 7 wavelength aethalometer and mass spectrometer, Garg et al. (2016) was able to identify different biomass burning sources other than just wood burning, including cereal-residue burning, leaf litter burning and rubbish burning. The authors also demonstrate that flaming biomass combustion can have an Ångström value of close to 1, and PM emitted from poorly optimised vehicles can have an Ångström value in excess of 1 **which therefore renders this method unsuitable as a source apportionment method on its own**.

Li et al. (2019) compared a number of measurement techniques for biomass burning aerosols and found that results were variable due to interferences from co-emitted species and changes to the aerosol during aging. The authors recommended that calculation and correction procedures for determining the optical properties of biomass soot be further evaluated to determine their applicability this source.

The detection range of the aethalometer is 0.1 to ~ 100 μg/m^3 and has a reported accuracy of 10% (Sedlacek, 2016). The aethalometer also uses fixed default values of some aerosol optical properties such as mass absorption cross section (MAC) which are used by the instrument to calculate, for example, black carbon concentration. However, the MAC is variable depending on the particle source and the instrument manufacturers (Magee) state that users can select a value of MAC for more site-specific conversion.

The values of MAC used in the AE33 aethalometer are 18.47 m^2/g for measurements at 370 nm and 7.77 m^2/g at 880 nm. However, Tasoglou et al. (2018) showed that the variations in the OC/BC ratio from different types of biomass burning can lead to these default values of MAC being out by a factor of 2 or more.

Due to the high uncertainties in the value of MAC and the Ångström exponent between different fuel types, technology types and sampling conditions, there is a need for better understanding of the impact of variability in these parameters on source

apportionment methods and results. Additional light absorption by brown carbon in the UV is parameterised by the ratio of black carbon to organic carbon, indicating that aerosol absorptivity depends largely on burn conditions rather than fuel type (Saleh et al., 2014).

2.1.2 Carbon isotopes

Carbon isotope source apportionment is used to differentiate between biogenic/contemporary and fossil fuel derived combustion sources to carbonaceous aerosol. During growth, biomass material absorbs atmospheric CO_2 through photosynthesis with a known abundance of the carbon radioisotope ^{14}C, which decays as it ages. Therefore, a particulate sample from residential coal burning would be more depleted in ^{14}C that a particulate sample from biomass burning. Consequently, the ratio of ^{14}C abundance in a sample to that of contemporary carbon is useful in source apportionment. Other isotopes are also useful such as ^{13}C.

Using this technique, Ward et al. (2006) found that 78-82% of ^{14}C in the ambient PM of Libby, Montana, was attributable to the burning of biomass, mostly likely residential wood burning. There may however be contributions to ^{14}C from other sources such as wildfires and garden burning

Carbon isotopes may also be used for the source apportionment of other climate and health relevant pollutants contacting carbon, such as methane, CO and PAH (Currie et al., 1994a). Unlike vehicular particulate samples, wood smoke PAH has been found to be ^{13}C heavy in benz(a)anthracene relative to fluroanthene and pyrene, and also ^{13}C depleted in chrysene and benzofluoranthene. These findings by (O'Malley et al., 1994) were used to identify residential wood burning as the dominant source of PAH in St John's Harbour and Conception Bay, Newfoundland, but there was some prior indication that domestic burning may have been a major pollution source in the area.

Caution is advised for source apportionment using carbon isotopes, because the fuel conversion technology and temperature influences the ^{13}C content of the emitted compounds (McRae et al., 1999). For example, methane emitted from smouldering fires where the combustion efficiency is low (high OC, CO) was found to be depleted in ^{13}C compared to high temperature flaming combustion (high EC, CO_2) (Chanton et al., 2000). The biomass fuel type can also impact on the ^{13}C signature of emitted carbonaceous aerosol, VOC and hydrocarbons (Gensch et al., 2014).

Strengths and weaknesses

Radiocarbon analysis has been found to be a relatively precise and accurate technique in source apportionment between fossil fuel and biogenic sources. For example, it may be shown that 40% of emissions come from a biomass source but this technique cannot say exclusively what this biomass source is. Promising work has shown potential for source apportionment of biomass sources based on carbon isotope content in gaseous species such as CO and CH_4, but this is a relatively new

technique and high costs may be prohibitive to routine analysis across the UK monitoring network (Jordan et al., 2006).

2.1.3 Levoglucosan and anhydrosugars

Levoglucosan (1,6-anhydro-β-d-glucopyranose) is a water-soluble monosaccharide anhydride that is produced during the pyrolysis of carbohydrates such as starch and cellulose. Any biomass containing cellulose may be a source of levoglucosan and therefore "although levoglucosan can indicate cellulose burning emissions, it alone cannot distinguish type of biomass burnt." (Bhattarai et al., 2019).

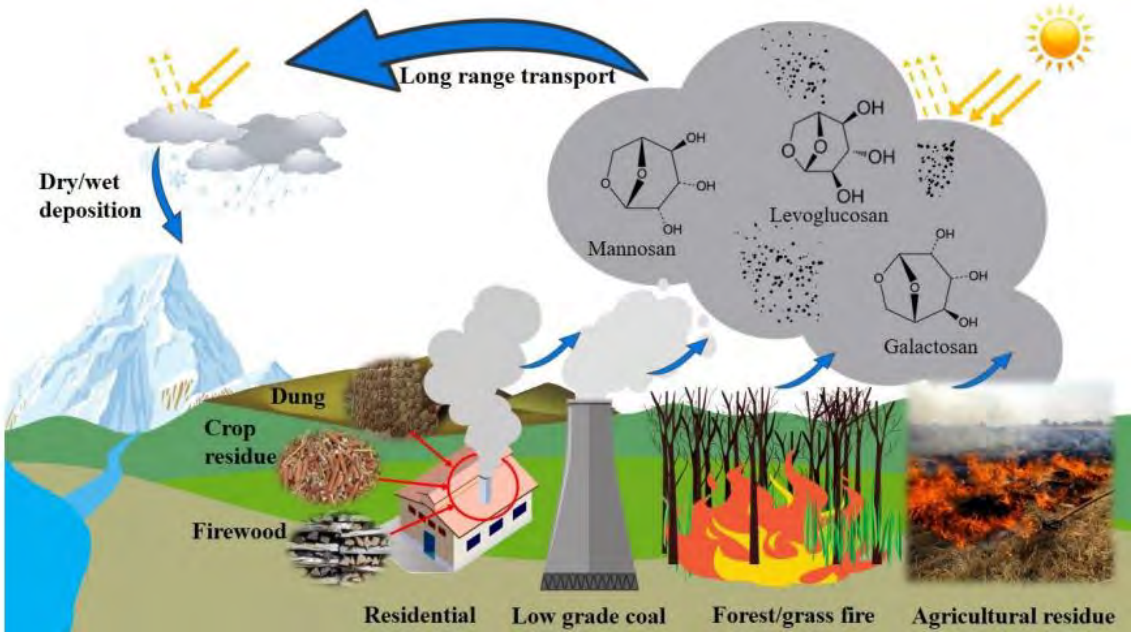


Figure 6. Levoglucosan sources and life in the atmosphere. Source: (Bhattarai et al., 2019)

Levoglucosan has been widely used as a macro-tracer for biomass burning for many years due to its relatively stable properties in the atmosphere (Bai et al., 2013, Fraser and Lakshmanan, 2000) and reasonably large emission rates (Simoneit et al., 1999, Nolte et al., 2001). Levoglucosan emissions factors for domestic wood burning appliances can vary from 1 mg kg^{-1} in efficient pellet or chip boilers to more than 5000 mg kg^{-1} in older inefficient stoves and fireplaces burning high moisture wood logs (Jordan and Seen, 2005). Hence the mass fraction of levoglucosan in PM can vary from 3% to more than 20% (Hedberg et al., 2006). Where knowledge of local wood types being burned is available, levoglucosan fractionation in total OC and PM can be used to estimate the contribution of biomass burning to aerosol loading.

Use of levoglucosan in source apportionment

In simple terms, the concentration of PM originating from biomass burning (PM_{bb}) may be calculated by multiplying the concentration of levoglucosan measured in a PM sample by a known ratio of levoglucosan to PM (F_{bb}). In the UK, it is standard practice to use a value of between 10.7 and 11.0, as used in Font and Fuller (2017), Cordell et al. (2016) and elsewhere.

The value of 10.7 for F_{bb} is derived from work by Schmidl et al (2008) and assumes a 20/70/10% split in fuel types between beech/spruce/briquettes. The authors measured PM_{10} emissions and levoglucosan emissions from a traditional Austrian tiled stove (Kachelofen) built in 1994. A sampling temperature of 30°C was used with purified dilution air, allowing condensation of the vapours. This is clearly not representative of modern Ecodesign compliant wood burning stoves and therefore it is recommended that further testing be carried out to determine the most appropriate value for the UK.

The levoglucosan content in PM varied depending on fuel type, for example it was 62% lower in PM from beech wood logs than in PM from spruce logs, both having a moisture content of 10-11%. Errors were estimated to be ±20% and the results are said to be applicable to mid-European Alpine regions. It is important to note that the factors used in this work (which has been used to inform many subsequent studies) are based on the ratio of levoglucosan measured in fresh PM collected on a filter. As described later in section 2.3.2, the mass of particles in ambient air may increase after emission due to gas-phase reactions. Since levoglucosan is relatively stable, the value of F_{bb} may therefore be lower than 11 in aged biomass smoke plumes. Bhattarai et al. (2019) state that changes in F_{bb} during aging processes may create problems in source apportionment and this should be taken into consideration. Though this may cause the value of F_{bb} to be lower, the value may also be higher for inefficient appliances such as open fires burning high moisture content wood.

Caseiro and Oliveira, (2012) used weighted-average values of F_{bb} of 10.7 for Northern European locations and 7.61 for Portugal, reflecting differences in the fuel mix in those locations. Puxbaum et al. (2007) used an average value of 7.35 (for OC rather than total PM) for a mixture of hardwood and softwood, Caseiro et al. (2009) used a weighted average of 6.1 (for OC) and 9.1 (for OM) for a fuel mix of 15% beech and 85% spruce, and Sang et al. (2013) used a value of 10.0 (OC) for a calculated fuel mix of crop residues, hardwood and softwood.

Levoglucosan stability

Estimates of levoglucosan lifetime vary from 0.7 to 26 days depending on atmospheric conditions (Bhattarai et al., 2019), but it is generally considered to be a relatively stable compound with a lifetime of at least 10 days. However, levoglucosan may react with radicals in the atmosphere and a correction for this was made by Sang et al. (2013) using a standard exponential decay rate model, allowing the ageing of biomass smoke

plumes to be taken into account. It is recommended that this approach be followed for sources of biomass burning aerosols which are located a significant distance from the sampling location.

Use of other anhydrosugars for apportionment of different biomass sources

In addition to levoglucosan, there are two other anhydrosugars that are used in source apportionment; mannosan and galactosan. The different ratios between these three compounds can be used to differentiate between different biomass burning sources (hardwood, softwood, forest fires, agricultural burning etc.).

Unlike levoglucosan which is a product of cellulose pyrolysis, mannosan and galactosan are derived from hemicellulose. The hemicellulose content of biomass varies depending on type, but it decomposes at 225-325°C whereas cellulose decomposes at 325-375°C (Zhang et al., 2011). Therefore, different ratios of levoglucosan to mannosan and galactosan can indicate different combustion temperatures, as well as fuel type.

The ratio of levoglucosan to mannosan (lev/man) and mannosan to galactosan (man/gal) can be used to distinguish between the types of biomass contributing to the aerosol loading. For example, crop residues such as cereal straws have been associated with high ratios of lev/man, whereas the ratio for softwood is much lower (Table 2). Lev/man ratios are highest for lignite and galactosan emissions are very low, indicating a loss of hemicellulose with increasing coalification (Fabbri et al., 2009). The typical ratios in ambient air are reported in Bhattarai et al. (2019).

Table 2. Average ratios of anhydrosugars and OC for different biomass types (Mitchell, 2017).

Biomass	Lev/Man	Man/Gal	OC/Lev
Crop residues	32.6 ± 19.1	1.2 ± 1.1	17.2 ± 9.2
Softwood	4.0 ± 1.0	3.9 ± 2.1	8.5 ± 6
Hard wood	21.5 ± 8.3	1.5 ± 0.3	7.1 ± 3.9
Lignite	54.0		

As shown in Table 2, there are reasonably large uncertainties associated with this method so a combination of cellulose- and hemicellulose-derived MA's with other chemical tracers is recommended, such as methoxyphenols derived from lignin pyrolysis (Gaston et al., 2016) and trace elements (Hedberg et al., 2006). Compound specific ^{13}C analysis within anhydrosugars (levoglucosan, mannosan and galactosan) has also been highlighted as a promising technique for source apportionment of fuel type and combustion conditions for biomass burning (Sang et al., 2012).

The use of levoglucosan and other anhydrosugars as tracers was recently the subject of a detailed review by Bhattarai et al. (2019). The authors surmised that "*Levoglucosan is well recognized as a tracer of BB, but the types of BB sources, e.g., forest fire and agricultural residue burning are not well differentiated. Therefore, ratios of levoglucosan with various tracers/components (not limited to OC, mannosan and galactosan) are most essential to give insights into the sources*." Bhattarai et al. (2019)

Strengths and weaknesses

Numerous studies have shown a clear correlation between increased levoglucosan and increased PM concentrations where there are known pollution sources including residential wood combustion and wildfires. In some areas, clear correlations with air temperature have also been identified (Monteiro et al., 2017) which have been attributed to the burning of wood and biomass for heating in the wintertime, though this correlation has not been observed in the UK (Cordell et al., 2016). Nevertheless, levoglucosan is a useful indicator of generic biomass burning aerosols.

However, as with other tracers, accurate quantitative source apportionment with levoglucosan is marred by the dependency of emissions factors on fuel type and burning conditions for different sources. It has been shown that levoglucosan emission factors may vary by a factor of 5 or more, resulting in large uncertainties for source apportionment (Szidat et al., 2006).

Other sources of levoglucosan

Since levoglucosan is produced by the pyrolysis of carbohydrates, it is related to inefficient and incomplete combustion. As such there may be many hundreds of sources of levoglucosan including bonfires, forest fires, grassland fires, agricultural burning, the burning of low grade coal, lignite and peat (Bhattarai et al., 2019). It is also produced from cooking and from cigarettes.

Typical cooking temperatures are not usually high enough to generate levoglucosan (<300°C), but it can be generated at lower temperatures (Larsen et al., 2003). Zhao et al. (2007) showed that levoglucosan was present in the particulate matter emitted from restaurants which fry plant-based foods. Cigarettes are also a source of levoglucosan, with emission rates reported up to 211 µg per hour (Ruprecht et al., 2017). Emission factors for coal can be up to 16 mg/kg, which equates to approx. 500 µg/MJ (Yan et al., 2018). Levoglucosan may also be released in trace amounts from the starch that binds smokeless fuel briquettes.

There is a lack of evidence around the levoglucosan emissions from the most efficient biomass burning stoves and boilers. Higher temperatures and more complete combustion lead to minimal levoglucosan emissions. Indeed, Orasche et al (2012) found that the concentrations of levoglucosan decrease significantly with increasing combustion temperature, as shown in Table 3.

Table 3. Typical levoglucosan emission factors for different types of residential wood burning appliances. Source: Orasche et al. (2012).

Appliance	Fuel type	Rated heat output	Fuel MC (%)	Condition	Efficiency (calc)	Levo-glucosan (µg/MJ)
Pellet boiler	Spruce pellets	25kW	7.7	Cold start	51	690
				Nominal load	87	140
Chip / pellet boiler	Spruce chips	30kW	23	Cold start	37	560
				Nominal load	86	83
Log wood boiler	Spruce logs	30kW	17	Cold start	22	5,500
				Nominal load	76	150
Log wood boiler	Beech logs	30kW	15	Cold start	18	6,700
				Nominal load	75	310
Pellet stove with heat exchanger	Spruce pellets	13kW	8	Cold start	27	6,500
				Nominal load	85	340
Log wood stove	Spruce logs	8kW	13	Cold start	75	11,000
				Nominal load	70	2,300
Log wood stove	Beech logs	8kW	15	Cold start	70	15,000
				Nominal load	69	1,500

Levoglucosan emission factors for several 15-33 kW pellet boilers were measured by Chandrasekaran et al. (2013) and were found to be 6-100 µg/MJ for grass pellets and between 9-130 µg/MJ for wood pellets at 75-85% efficiency.

Emission factors for wildfire burning are reported to range from 1270 ± 540 mg/kg for herbaceous biomass to 58.6 ± 35.1 mg/kg for oak woodland (Hosseini et al., 2013). Assuming a calorific value of 18 MJ/kg, this converts to 71,000 µg/MJ and 3,300 µg/MJ respectively. Hence emissions from wildfires and other inefficient sources are usually far higher than from high temperature, high efficiency modern appliances (apart from during the start-up phase).

In polluted atmospheres where there is known to be a significant amount of a certain type of biomass burning, levoglucosan is a useful means to estimate the contribution of that source to total particle loading. However, in atmospheres that are impacted by mixed sources of air pollution, levoglucosan is best used "*as a tracer for biomass combustion in general - from any cellulosic material*" (Schimdl et al., 2008).

It is recommended that the emission factors of levoglucosan for different types of domestic burning and cooking be further investigated with a view to better understanding the variation of the factor F_{bb} used to apportion PM concentrations to biomass burning. *"The apportionment methods are often based on partially known parameters like the wood consumption or the appliances used in the investigated area. Moreover, apportionment methods are affected by large uncertainties in emission factors. Indeed, levoglucosan, EC, and OC concentrations in wood smoke vary remarkably changing the wood type. In particular, the levoglucosan to PM emission*

*factor varies according to the type of wood combusted and to the appliance used as well as to burning rates, air dilution, and moisture content in the fuel" (*Piazzalunga et al., 2011).

2.1.4 Inorganics and trace elements

Inorganics and trace elements are typically used within source profiles in chemical mass balance source apportionment studies (Chow et al., 2007). The majority of inorganics are present in very low concentrations in biomass PM and the differences between wood species tend to be small, largely depending on the soil and environmental conditions where the tree was grown (Kleeman et al., 1999). Certain elements such as potassium, chlorine and zinc are abundant in biomass burning particulate matter, and high concentrations of these species in ambient PM may indicate a source of biomass burning (Hedberg et al., 2006). Potassium and chlorine each make up 2-5% of woody biomass fine particulate mass and hence are useful tracers (Reid et al., 2005a). However again they are subject to large variations in emissions factors, with higher emissions generally being correlated with higher temperature efficient combustion (Khalil and Rasmussen, 2003) and herbaceous rather than woody biomasses (Turn et al., 1997). Despite this, the Zn and K content of biomass burning particulate is not as dependent on burning conditions as levoglucosan (Hedberg et al., 2006).

Source apportionment using these species is also subject to interferences from other sources of fine particle K and Cl and from the choice of measurement method. Much of the potassium released through the combustion of biomass is in the form of water soluble inorganic salts KCl and K_2SO_4 (Sandradewi et al., 2008a). Aerosol mass spectrometers measure K+ ions generated from the volatilisation of these compounds, which is a destructive technique, and more qualitative than others due to incomplete volatilisation (Weimer et al., 2008).

Potassium is one of the most extensively used inorganic tracers. Despite potassium being ubiquitous in the environment, elevated concentrations above known background levels may be indicative of biomass burning. In affected areas, a linear correlation has been observed between levels of water soluble K in ambient PM and residential wood burning (Chow et al., 2007). Dust from crustal matter has a known K/Fe ratio and any K measured in PM in excess of that ratio may be attributed to wood smoke (Calloway et al., 1989, Currie et al., 1994b).

Although K emissions factors from softwood are generally lower than hardwood and crop residues, potassium alone is not thought to be a suitable tracer for detailed source apportionment between fuel types. Nevertheless, the ratio of potassium to levoglucosan has been shown to be useful in this regard (Urban et al., 2012). Puxbaum et al., (2007) found that a ratio below 0.2 is indicative of residential heating with wood, whereas a ratio above 0.5 is indicative of open burning of straws and grasses. However, the K/levoglucosan ratio presented by Caseiro et al. (2009) ranged from 0.59 to 1.11 which was significantly higher than values reported in the literature. The

authors argued that despite the higher values, the correlation between the two species remained strong, and higher temperature combustion in more modern appliances results in higher relative inorganic content in the particulate (fly ash potassium salts). Urban et al. (2012) also found that the ratio is lower for smouldering (0.24) versus flaming (1.01) combustion. Nonetheless, the results reflect the uncertainties associated with this method.

The ratio of potassium to elemental carbon has also been used to distinguish between fuel types, with ratios of 0.20 and 0.95 reported for herbaceous and lignocellulosic biomass PM respectively (Turn et al., 1997). In addition to potassium, herbaceous biomass particulate is characterised by high levels of Cl. Turn et al. (1997) found that chlorine emissions factors were 10 times higher for herbaceous biomass fuels than wood fuels, whereas zinc emissions factors were 25-30 times higher for wood fuels. Despite this, few studies use zinc in source apportionment as other factors are known to contribute to zinc loading, including mobile sources such as brake wear (Chow and Watson, 2002).

Certain elements may also be used to identify waste and waste wood burning in ambient particulate matter. Copper chromium arsenate (CCA) has been used as a wood preservative for many years and occasionally (and illegally) it is burned domestically as a free source of fuel. However, this can be severely detrimental to health. Measurements of ambient PM in Seattle, USA, revealed high levels of arsenic and statistical analysis revealed a Pearson correlation coefficient of 0.84 between arsenic and biomass burning, suggesting prevalent waste wood burning (Maykut et al., 2003). A similar correlation has been observed in wood burning regions of New Zealand, where annual average inorganic arsenic levels have been measured at over 3 times the national air quality guidelines limit (Ancelet et al., 2015). Arsenic was found to be largely confined to the fine particle fraction, but more research in this area is required. It should be noted that the burning of treated wood is prohibited in the UK and the vast majority of home owners are aware of this.

2.1.5 Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAH) are organic compounds consisting of multiple aromatic rings, 16 of which have been classified as priority pollutants by the US EPA due to their toxicity.

PAH are produced from a number of combustion sources and are present in solid, liquid and gaseous environmental media. Emissions are known to be high for low temperature combustion sources such as bonfires and open fireplaces (Katsoyiannis et al., 2011) and ΣPAH emissions factors of 250 ± 25 mg/kg and 43 ± 9 mg/kg have been reported for residential coal and wood burning respectively (Lee et al., 2005).

Measurements of twenty or more PAH species can be made simultaneously and the relative abundances of certain species over others can be used to determine the source (Khalili et al., 1995). In the particle phase, lower molecular weight (202-228

g/mol) species dominate the PAH profile of wood smoke irrespective of combustion conditions and fuel type (Hays et al., 2003). However, total emissions factors are much higher for higher moisture fuel and appliances with a poor air supply, and also shorter-chain PAHs such as fluorene are enriched where combustion temperatures are low (Viana et al., 2008a). Acenaphthylene, anthracene, benzo(a)pyrene, naphthalene, phenanthrene and retene are the major components of wood smoke PAH (Khalili et al., 1995). The latter is released from both hardwood and softwood, as well as coal combustion, and has been extensively studied due to its carcinogenic properties (Bari et al., 2009, Hays et al., 2003).

Retene (1-methyl-7-isopropylphenanthrene) is present in relatively high proportions in softwood smoke and has been used as a tracer for many years (Ramdahl, 1983). It is formed from resin acids typical of softwood burning (Benner et al., 1995), and is very low in hardwood particulate matter (Fine et al., 2002). It is, however, semi-volatile and may be partially lost in the atmosphere (Schauer and Cass, 2000)

Source profiles for PAH emissions are useful in receptor modelling and clear geographical and seasonal differences in the ratios of certain species have been identified depending on the pollution source. Elevated levels of fluoranthene, pyrene and benzo(a)pyrene were identified in rural areas of Croatia during the winter, thought to be due to residential heating with wood and coal (Jakovljević et al., 2016). The heating season in Harbin, China was characterised by an increase in the ratio of IcdP/(IcdP + BghiP) and a decrease in ratio of the FL/(FL+PYR), which was attributable to domestic coal burning in the region (Ma et al., 2010). Despite some interferences due to summer wildfires and photodegradation, the wood heating season is also characterised by higher ratios of BaA/(BaA + CHR) and ANT/(ANT + PHE) (Tobiszewski and Namieśnik, 2012). Detailed PAH diagnostic ratios can even be used to differentiate between biomass types.

The ratio of 1,7- to 2,6-dimethylphenanthrene (DMP) is a relatively sensitive source apportionment indicator for several fuel types (Benner et al., 1995). Values below 0.5 suggest vehicular emissions, between 0.6 and 0.7 suggest coal burning, and values between 0.7 and 0.9 suggest wood burning (Yunker et al., 2002). However, practical application of diagnostic ratios in regions with a complex mix of PAH sources and sinks has proven difficult and a prior knowledge of sources is useful (Katsoyiannis et al., 2011, Dvorská et al., 2011). Details of the diagnostic ratios of PAH compounds for cooking specific sources is given in Abdullahi et al. (2013).

Strengths and weaknesses

PAH ratios are a useful tool in source apportionment and have been used to distinguish between vehicle sources, cooking sources, coal burning and biomass burning (hardwood, softwood and grasses). There are a limited number of studies of this type in the UK, but source emission profiles are well understood and have promise for receptor modelling.

It is a relatively involved process to sample and determine PAH content in ambient air due to low concentrations which increase measurement errors near the limit of detection. Samples are also relatively easily contaminated and can degrade during long storage periods. Additionally, the ratio of PAHs is not fixed for each pollution source and can vary for biomass burning depending on fuel type, fuel moisture content and efficiency of the burn.

2.1.6 Other tracers and chemical mass balances

The most accurate source apportionment methods are thought to be modelling techniques that use a number of different chemical tracers and markers for each source to develop a source profile. Samples from ambient air are analysed and compared to different source profiles including vehicles, cooking, biomass burning etc and statistical techniques are applied to the data to look for similarities, which can be used to apportion different sources. However, these techniques require sophisticated instrumentation and analysis such as aerosol mass spectrometry and gas chromatography mass spectrometry, which can be time-consuming and costly and not routine analysis. The modelling techniques used include Positive Matrix Factorisation (PMF), Principle Component Analysis (PCA) and Chemical Mass Balance (CMB). A brief overview is given below, followed by a description of some of the key chemical tracers used.

Modelling techniques

Positive Matrix Factorisation is a commonly used technique for source apportionment and is derived from receptor modelling. Correlation matrices are developed whereby rows typically represent emissions profiles of various sources and columns represent the scores assigned to each emissions profile factor (Kim and Hopke, 2007).

PMF has been shown to have several advantages over other techniques such as principal component analysis (PCA), including non-negativity and the lack of a need for prior information on emissions sources in a given area (Gianini et al., 2012).

Several versions of PMF software are used such as PMF2 and the USEPA PMF. Comparative studies have shown that generally the different models agree well, but the wood smoke source profile was found to have one of the lowest correlations between the two models, reflecting the high uncertainty in this area (Larsen and Baker, 2003).

Chemical Mass Balance analysis (CMB) is also a receptor model that uses variance least squares regression analysis and provides a sum of linear products of source profile contributions (Friedlander, 1973, Ward et al., 2012). CMB is a fully constrained model and a prior knowledge of emissions sources and profiles is required (Chow and Watson, 2002). CMB may combine many other tracers such as anhydrosugars, trace elements and carbon isotopes and hence may be very useful in rural communities where winter wood burning is known to be a significant source of air pollution (Gianini

et al., 2013). CMB is particularly useful for source apportionment of the organic fraction of particulate matter and numerous studies have used this technique for OC, OM, VOC and individual species (Schladitz et al., 2015, Yin et al., 2015, Hellén et al., 2008, Chow and Watson, 2002). The benefits of CMB include allowing errors to be assigned to each source profile contribution, with free software available from the USEPA. Drawbacks include an inability to identify unknown emissions sources and a lack of detailed secondary aerosol effects (Viana et al., 2008b). Detailed emissions inventories are crucial for CMB, and it has been shown that fuel-specific profiles can be used to differentiate between biomass types such as hardwood and softwood (Chow et al., 2007). An example profile for general wood smoke is given in Hannigan et al., (2005).

Chemical tracers used for detailed source apportionment of biomass burning

An extensive review of organic markers for different types of biomass combustion was conducted by Simoneit (2002). It was found that differences in pyrolysis products from different parts of the biomass (cellulose, hemicellulose and lignin) made the most useful tracers. These are discussed below.

The relative proportions of pyrolysis products from the lignin types (guaiacyl-, syringyl- and anisyl-) can be used for fuel specific source apportionment in areas of mixed biomass burning (Gaston et al., 2016, Simoneit et al., 1993). Guaiacyl- lignins are typical of softwood, whereas a combination of guaiacyl- and syringyl- lignin is typical of hardwood, and anisyl- lignin is typical of herbaceous biomass (Nolte et al., 2001). Pyrolysis of guaiacyl- lignin yields guaiacol (2-methoxyphenol), eugenol (4-Allyl-2-methoxyphenol) and vanillin (4-hydroxy-3-methoxybenzaldehyde) which, together with resin acids and coniferyl alcohols, may be used for source apportionment of softwood burning (Mazzoleni et al., 2007, Nolte et al., 2001).

Pyrolysis of syringyl- lignin yields syringol (2,6-Dimethoxyphenol), syringaldehydes, syringyl organic acids and synapyl alcohols (Simpson et al., 2005). Guaiacol is emitted in approximately equal amounts from hardwood and softwood burning, but syringol emissions factors are up 250 times higher for hardwood than softwood burning (McDonald et al., 2000). Specifically, propionylsyringol and butyrylsyringol have been identified as particularly useful hardwood tracers (Schauer and Cass, 2000, Oros and Simoneit, 2001a). For softwood, coniferyl alcohol and resin acids such as abietic acid and dehydroabietic acid are particularly useful stable and non-volatile tracers (Bergauff et al., 2009, Schnelle-Kreis et al., 2007, Simoneit, 2002, Oros and Simoneit, 2001b).

Methoxyphenols are semi-volatile pyrolysis products of lignin and are present in relatively high concentrations in both the gas and particle phases of biomass smoke (Chow et al., 2007, Mazzoleni et al., 2007). Accounting for up for 21% of the fine particle mass (McDonald et al., 2000), more than thirty species of methoxyphenols have been identified for biomass burning (Hawthorne et al., 1988). Methoxyphenols

have unique tracer properties for biomass burning source apportionment and are relatively easily extracted and analysed. They are advantageous over tracers such as K, CH_3Cl, ^{14}C and anhydrosugars which may be sensitive to variable background concentrations and atmospheric degradation or require expensive and time consuming analysis (Hawthorne et al., 1988, Simpson et al., 2005, Hoffmann et al., 2007).

Nitrated phenols are a significant constituent of biomass burning secondary organic aerosol. They contribute to the light absorbing fraction of organic carbon (brown carbon) (Mohr et al., 2013) and have been used as biomass burning tracers. Compounds such as 4-methyl-5-nitrocatechol have been found to be present in higher concentrations than methoxyphenols in wood burning regions during the winter, and are evidence of aged, oxidised biomass SOA (Iinuma et al., 2010, Kahnt et al., 2013).

Traffic is believed to be the major source of nitrophenols, although wood and coal burning are known to contribute significantly (Kahnt et al., 2013). Emissions factors for nitrophenols range from 1.4 - 4.6 mg/kg for peat burning, to 12 - 31 mg/kg for softwood burning (Iinuma et al., 2007). The most abundant species in wood burning nitrophenols is believed to be 2-nitroguaiacol, although Orasche et al. (2012) found that emissions of 2,6-Dimethoxy-4-nitrophenol were 4 times higher for hardwood burning than softwood burning.

Halogenated methane compounds such as methyl bromide and methyl chloride (chloromethane, CH_3Cl) are also useful components in the wood burning source profile for CMB modelling (Edgerton et al., 1986, Turn et al., 1997). Aside from uses in industry or as a refrigerant, biomass burning is a key anthropogenic source of methyl chloride. Natural sources lead to relatively high background concentrations, but elevated concentrations have been observed in wintertime ambient air in wood burning communities (Khalil and Rasmussen, 2003, Hawthorne et al., 1988). CH_3Cl emissions factors range from 20 mg/kg for synthetic logs to 40 mg/kg for fireplace combustion of hardwood (McDonald et al., 2000).

Emissions do not vary with combustion temperature as significantly as elemental tracers (Khalil and Rasmussen, 2003), but emissions factors may be much higher for open burning or burning of high halogen fuels such as waste or agricultural residues (Lemieux et al., 2004). Many tracers for herbaceous biomass burning are the same as those of woody biomass and hence are not suitable as unique tracers. However, the relative proportions of some species such as PAH and potassium can be useful. In addition, a small number of unique tracers have been identified for herbaceous biomass burning which include anisic acid (p-methoxy-benzoic acid), triterpenoids and sterols such as campesterol (Simoneit, 2002).

Chemical tracers used for detailed source apportionment of coal burning

For peat, lignite and coal, unique tracers are dependent on the coal rank and burning conditions. The organic fraction of coal smoke is dominated by n-alkanes, n-alkenes

and aromatics such as picene (Dibenzo[a,i]phenanthrene, benzo(a)chrysene), as well as the ratio of hopane to homohopane (Oros and Simoneit, 2000). Lignite smoke is characterised by dominance of C31-hopanes, divanillyl, lignans, di- and tri-terpenoids (Simoneit, 2002). There is also an increase in phenolic emissions with decreasing coal rank.

Several studies from the Republic of Ireland offer useful information on source apportionment in regions where the RSF fuel mix is not as dominated by wood as it is in much of central Europe, North America, Australia and New Zealand. The fuel mix in Ireland consists of hardwood and softwood logs, peat turf and peat briquettes, manufactured solid fuels (petroleum coke, smokeless fuels), bituminous coals and anthracite (Mitchell et al., 2016). In addition to wood, peat and coal combustion also contribute to potassium, anhydrosugar and PAH concentrations which can hinder fuel specific source apportionment. Speciation can be achieved through additional profiles for peat and coal combustion in PMF models (Dall'Osto et al., 2014), and using PAH and monosaccharaide anhydride ratios. Levoglucosan to mannosan ratios of 3.7, 3.1 and 8.6 were found for ash wood logs, bituminous coal and peat briquettes respectively (Kourtchev et al., 2011).

Using a combination of an aerosol mass spectrometer, a 7 wavelength aethalometer and PMF, Lin et al. (2018) attributed up to 70% of PM_1 to residential solid fuel burning with peat, coal and wood in Dublin, Ireland. The measurements were taken during particularly severe air pollution episodes on 19 November 2016 and 22 January 2017. Ambient $PM_{2.5}$ concentrations were more than 12 times the WHO daily limit of 25 $\mu g/m^3$. The primary source of this pollution was found to be peat burning (46-55% of OA), followed by wood (15-17%) and coal (2-6%). Peat is a frequently used solid fuel in Ireland and in parts of Scotland but is not commonplace in most of the UK. Indeed, the authors state that "*the implications for policymaking* [in the UK] *are not clear due to poorly investigated links between solid fuel type, consumption and ultimately their contribution to air pollution*." (Lin et al., 2018).

Chemical tracers used for detailed source apportionment of cooking

The source profile for cooking is characterised by fatty acids such as palmitoleic acid and oleic acid, as well as cholesterol, aldehydes and alkanoic acids (Robinson et al., 2006), although emissions vary depending on food type and method of cooking. Frying with vegetable oils can release large amounts of organic condensable material which can confound aethalometer measurements, whereas meat cooking can release PAH, K^+ and Cl^-, which are used as indicators of wood smoke.

Several studies have been made of the emissions from charbroiling (grilling over a rack of charcoal) which is not dissimilar to barbecuing. $PM_{2.5}$ emission rates of 4.4 to 11.6 g/kg of uncooked meat have been reported (McDonald et al., 2003).

An extensive review by Abdullahi et al (2013) discussed the chemical source profiles for different types of cooking. It found that cooking processes can emit significant

amount of benzo[a]pyrene, formaldehyde, acetaldehyde and acrolein. Emissions factors for levoglucosan as high as 554 mg per gram of organic aerosol were also reported. Evidence showed that total $PM_{2.5}$ emissions from cooking could be comparable to those from vehicles in the USA.

2.2 Results of source apportionment studies in the UK and abroad

As discussed in the previous section there are a variety of methods available for the source appointment of residual solid fuel smoke in the atmosphere. Determination of the contribution of RSF to ambient levels of PM is complex due to the mixing with pollution from other sources. Typical categories used in source apportionment studies include traffic, road dust, brake wear, crustal matter, industrial sources and with regard to RSF the majority of studies refer to biomass burning (including wildfires) or residential wood combustion.

A comprehensive review of global source apportionment studies for RSF using various source appointment methods can be seen in Table A1 in the appendix, building on the findings of Mitchell (2017). There is a wide range of geographic locations, sampling station types (urban, rural etc) and source appointment methods used resulting in a range of % contributions from RSF reported. In the UK the studies suggest that the contribution to PM levels from RSF ranges from 2-45%, the lowest reported at a London urban background site in the summer and highest in West Yorkshire in Winter. The majority of the studies in the UK are based in London where the RSF % contribution ranges from 2-43%. When considering the same times of year and type of monitoring location (urban background) then the range is still 4-38% with four of the six studies reporting values below 15%. There were studies that reported the % contribution in both the winter and summer with Fuller et al (2014) reporting an 8% increase in winter (10% RSF contribution in winter) and a 12% increase in winter at another London site (23% RSF contribution in winter). Several studies (Young et al., 2015; Fuller et al., 2014) determined the annual contributions of RSF in London urban background locations and reported values ranging from 8 - 43%.

Some of the areas most affected by domestic burning include Tasmania, Australia; Thessaloniki, Greece; the Po Valley, Italy; Hastings and Alexandra, New Zealand; Roveredo, Switzerland; and Libby Montana, Rochester New York and Puget Sound Washington in the USA. In these areas, the contribution of residential wood combustion to ambient air quality has been studied in detail with reported contributions ranging from 43-96% (Table A1 in the appendices). It should be noted that without further analysis of the residential energy mix and the measurement methods used in the above studies, which is outside the scope of this report, it is not possible to compare and contrast the domestic burning contribution with that of the UK where both burning habits and source apportionment techniques are different. For example, factors such as wide scale availability of natural gas for heating in the UK may mean that there is less reliance of solid fuels for supplementary heating in comparison to other countries.

The source apportionment studies presented reveal the impact of real-world emissions on ambient air quality. Real-world emissions may be significantly higher and more variable than emissions derived under standard laboratory conditions due to user variables (Wöhler et al., 2016). The studies also show the seasonality of RSF emissions. For example, RSF was found to contribute to 28% of OC in the summer in Cork, Ireland, versus 80% in the winter (Kourtchev et al., 2011). From a policy perspective, there is therefore a need to better quantify RSF activity data and emission factors; both in-situ and under real-world conditions simulated in the laboratory

2.3 Primary and secondary particulate emissions

2.3.1 Primary emissions

When apportioning particulate matter to different sources, it is important to distinguish what size fraction the contribution refers to (PM, PM_{10} or $PM_{2.5}$) and whether the contribution refers to total emissions or to the concentration in ambient air. The former is dependent on sampling temperature as condensables can increase particle mass. An overview of the different notations is given in Table 4.

Inventories such as the NAEI in the UK and the EMEP/EEA Guidebook in Europe use emission factors and activity data for different sources to calculate the total annual emissions, which can then be apportioned from the total nationwide emissions. Therefore, the choice of PM emission factor and the composition of the particulate can have a substantial effect on the calculated contribution.

Table 4. Notations used for different fractions of particulate matter.

Notation	Description
1) PM_{solid}	Primary solid particle emissions measured at elevated temperatures to prevent semi-volatiles (e.g. tars) from condensing on the filter and adding to the mass
2) PM_{total} or TSP	Total primary emissions measured at approx. 30°C, usually by means of a dilution tunnel. $PM_{total} = PM_{solid} + PM_{condensable}$
3) $PM_{secondary}$	The mass of particles emitted, taking into account secondary particle formation in the atmosphere after mixing with ambient air and being exposed to sunlight. The increase in mass with ageing is determined by the mass enhancement factor (M_e), whereby $PM_{secondary} = PM_{total} * M_e$
3) $C_{PM,x}$	The concentration of particulate matter in ambient air which is attributable to source *X*.

PM_{solid} has been calculated in the 2016 update of the EMEP/EEA Guidebook using the method of Denier van der Gon et al. (2015). As shown in the table, $PM_{total} = PM_{solid} + PM_{condensable}$, where PM_{solid} is the amount of non-volatile matter in PM. This is derived from Table 2 of Denier van der Gon et al. (2015) which uses data from Nussbaumer (2008). A comparison of PM emission factors for selected domestic burning sources in shown in Figure 7, together with the relative proportions of solid particles and condensable matter in the emitted PM.

The reason for the difference in PM_{solid} and $PM_{condensable}$ emission factors is the filter temperature during sampling, which varies both in the field and in the laboratory. Measurements made in ambient air or using a dilution tunnel (e.g. NS3058) allow for condensation of semi-volatile condensables onto the filters which includes tars and organics. This is not the case for hot filter sampling direct in the flue (e.g. German DINPlus method). Since condensables are products of incomplete or low temperature combustion, their relative proportion varies for different technologies.

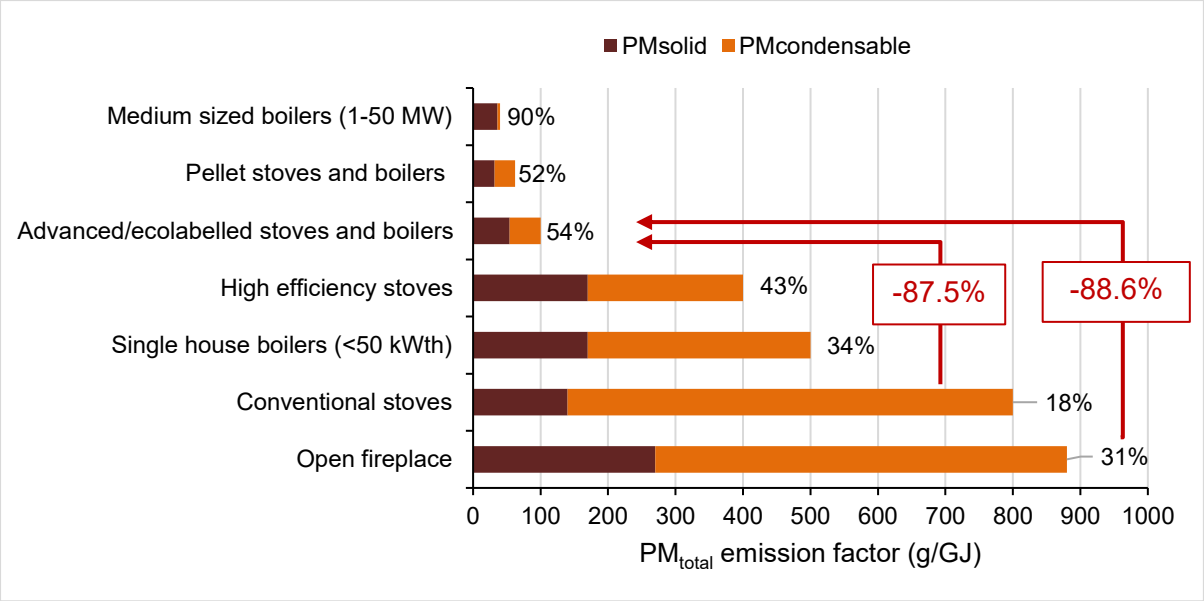


Figure 7. Solid and condensable PM_{total} emission factors by biomass technology type, according to the EMEP/EEA guidebook 2016. The solid particle content is shown as a percentage next to each bar.

As shown in Figure 7, larger scale domestic burning systems with higher efficiency not only have a lower emission factor, but also have a higher percentage of solid material in the PM. The solid material is typically elemental carbon particles and fly ash, whereas the condensable material consists of organic/brown carbon including methoxyphenols, PAH and tars which have a higher toxicity. PM from larger more efficient modern bioenergy systems can be >95% inorganic material, compared to 30-40% from older traditional log stoves (Vicente and Alves, 2018), and 1-56% for open burning (Hosseini et al., 2013). The reporting of condensable components in national

inventories is not mandatory and in recent years there has been no clear definition as to whether PM emission reporting includes or excludes the condensable component. However, in 2019 a new reporting table was included for European member states on 'Inclusion/exclusion of the condensable component from PM_{10} and $PM_{2.5}$ emission factors', and was completed by 15 member states (EEA, 2019a).

2.3.2 Secondary emissions

After emission to the atmosphere, there is a change in the size, morphology and composition of aerosol particles from all sources, including domestic burning (Cahill, 2010). Sunlight causes short-lived radicals to form in the atmosphere, which can react with a number of gas-phase pollutants to form new aerosol particles.

By injecting fresh smoke or flue gas into a sealed chamber and regulating the levels of sunlight, it has been possible to simulate smoke ageing in the atmosphere and measure the rate at which new particles are formed and the rate at which the PM mass increases. This has allowed the derivation of mass enhancement factors (M_e) for smoke from different sources, which have a different composition of gases. Certain gases have a higher propensity to form secondary organic aerosol (SOA) or secondary inorganic aerosol (SIA) than others.

A wide variety of pollution sources emit SOA and SIA precursors which can interact in the atmosphere and form new particles. An illustration of this is given in Figure 8.

Secondary inorganic aerosol

Several pollution sources emit precursors of secondary inorganic aerosol (SIA) which can contribute to particle loading in the UK atmosphere. The major constituents of SIA are ammonium nitrate and ammonium sulphate; formed by the reaction of ammonia with NOx and SO_2. Ammonia emissions are reasonably well quantified and mostly originate from the agricultural sector (88%), whereas road transport is the primary source of NOx (34%) and energy generation is the primary source of SO_2 (37%). The Clean Air Act states that domestic burning is also a major source of SO_2 emissions at 22%, largely due to the burning of high sulphur coal and smokeless fuel. Despite there being a limit of 2% sulphur content for fuels burned in Smoke Control Areas, some fuels burned outside these areas may have a sulphur content greater than 2%. However, actions set in place by the Government following the Clean Air Strategy are expected to extend to 2% limit to cover the whole of the UK. The sulphur content of wood and biomass is typically less than 0.1%, hence SO_2 emissions are very low. There is some evidence to suggest that domestic burning of coal and biomass can contribute to ammonia emissions (Li et al., 2016) but the total contribution is thought to be very small in comparison to agricultural emissions. Ammonia emissions are also related to combustion efficiency with more advanced and efficient appliances emitting less than 13% of the ammonia emissions compared to traditional stoves (Li et al., 2016).

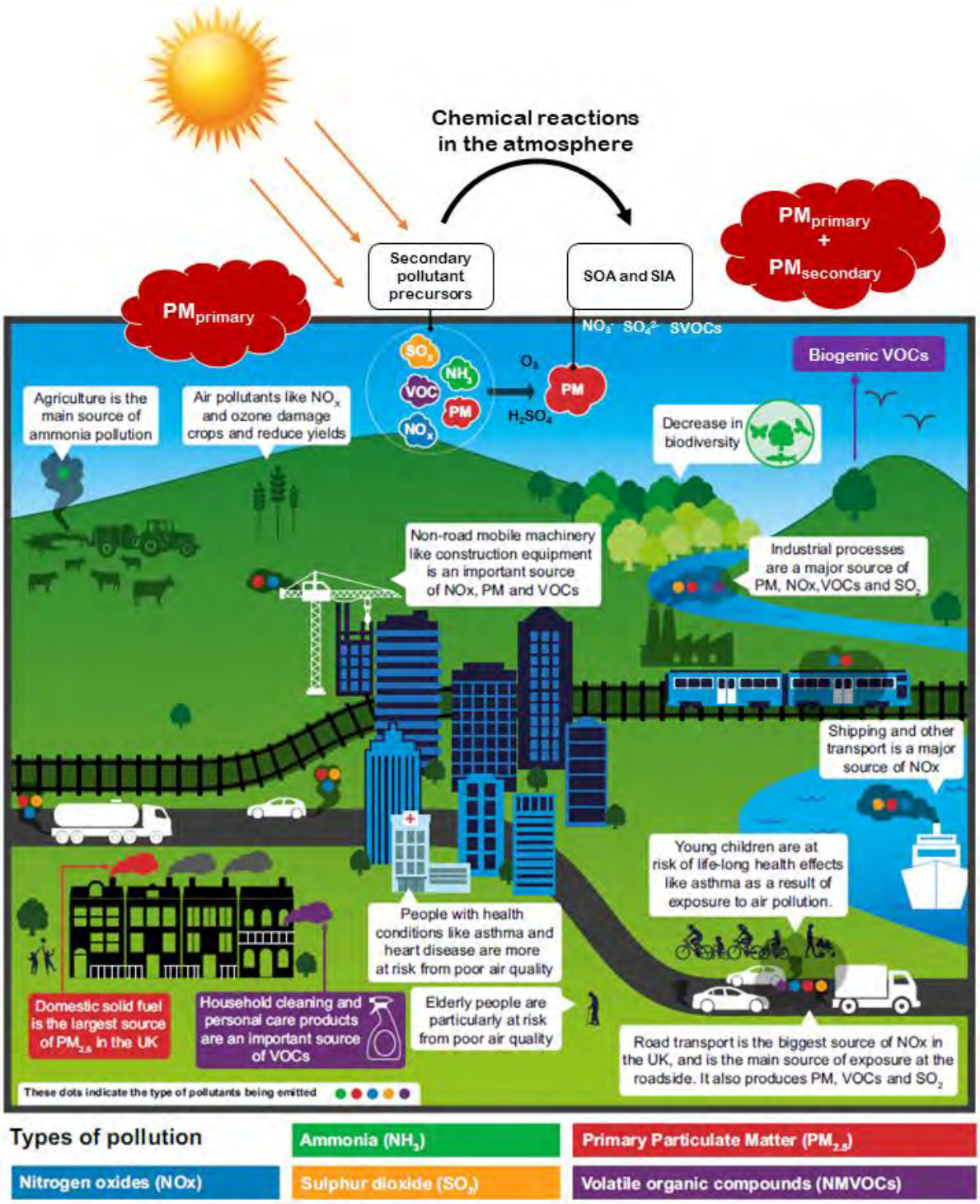


Figure 8. Illustration of primary and secondary pollutant emissions. Image adapted from the Clean Air Strategy (DEFRA, 2019a).

Secondary organic aerosol

SOA are formed as gas phase volatile organic compounds (VOCs) undergo chemical transformations to less volatile compounds, before condensing and nucleating and becoming part of the solid particulate phase (Seinfeld and Pandis, 2006). It follows

that primary organic aerosols (POA) are defined as the organic vapour compounds which are present in the hot flue gas of domestic burning sources, which have not yet condensed to the particulate phase. However, distinguishing between POA and SOA in ambient air is complicated by the fact that organic vapours can form part of an internally mixed soot particle and therefore be considered primary (Reid et al., 2005b, Seinfeld and Pandis, 2006).

The formation mechanisms of SOA are complex and dependent on the VOC mix, meteorological conditions and co-emitted species such as NOx. Details of formation mechanisms are described elsewhere (Dusek, 2000, Gelencser, 2004, Seinfeld and Pandis, 2006, Yee et al., 2013). Precursor VOCs may be emitted from a plethora of sources; both natural, including wildfires and direct emissions from living vegetation, and anthropogenic, including paints, motor vehicles, industry and domestic burning.

For domestic biomass burning sources, oxidation products of phenol, naphthalene and benzene comprise up to 80% of observed SOA (Bruns et al., 2016), with further contributions from aldehydes and lignin pyrolysis products syringol and guaiacol (Yee et al., 2013). The key VOCs emitted from coal combustion include alkenes such as octane and decene which oxidise to heptanal/nonanal and heptanoic acid/nonanoic acid (Gelencser, 2004). Important VOCs emitted from domestic burning include benzene, toluene and formaldehyde which are harmful to health as well as being precursors of SOA. As a smoke plume ages, SOA formation increases the PM mass and number concentrations as well as the OC:EC ratio over time.

Cooking processes are a significant contributor to VOCs and organic aerosol mass in urban areas, with clear evidence of SOA formation from cooking with vegetable oils (Liu et al, 2017). Despite there being high emissions of fatty acids, oils and other organics, the contribution of cooking sources to total UK particulate loading is highly uncertain. In London and Manchester, it was found that the average contributions to primary organic aerosol for traffic, cooking and solid fuels (assumed domestic burning) were 40%, 34% and 26% respectively (Allan et al., 2010). In urban environments, residential and commercial cooking contributes 17-19% of total organic aerosol (Kelly et al., 2018).

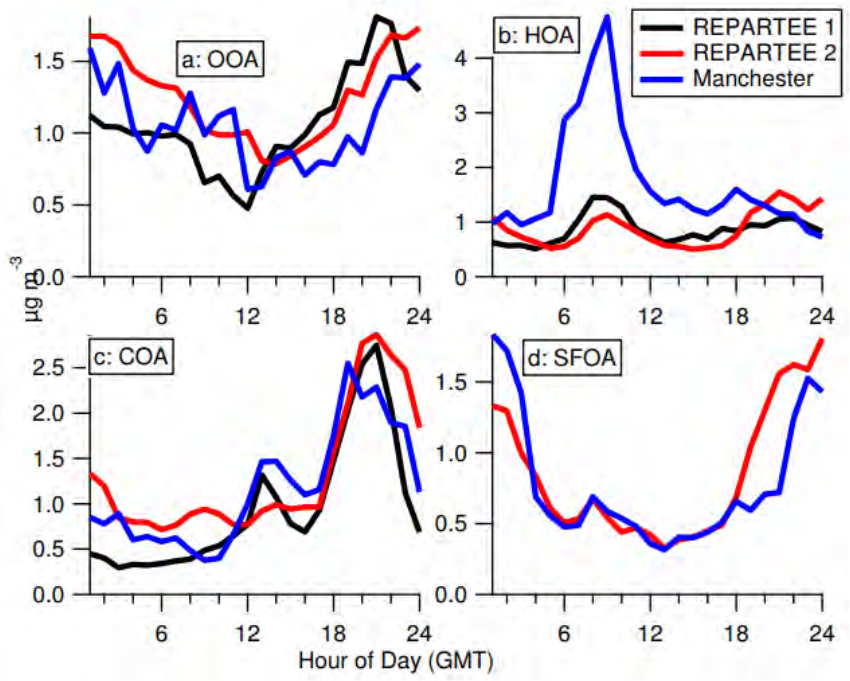
SOA formation profiles

Secondary particulate formation is influenced by diurnal trends in emissions and atmospheric chemistry, given the varying height of the boundary layer and the varying levels of sunlight throughout the day. Wintertime low temperatures favour partitioning of semi-volatile VOCs into the aerosol phase, and stable meteorological conditions favour the accumulation of precursors.

Anticyclonic conditions in winter have often been associated with high pollution episodes that are directly attributable to domestic burning. Indeed, stable air masses with low wind speeds allow for poor mixing and a build-up of air pollutants from a number of sources. It is during these conditions that an inversion layer can form,

whereby there is a reverse of the normal decrease of air temperature with altitude. Measurements made during these conditions in areas where domestic burning is commonly used for heating have shown that the total contribution to ambient PM levels can be very high (see for example Lin et al., 2018). Such periods can last from 1-2 days to 2 weeks or more in the UK, though there is less dependence on solid fuels than other countries such as Ireland, New Zealand and Alpine regions. When stable conditions occur in the summer, outside of the heating season, severe air quality episodes are most commonly attributed to vehicle sources.

During anticyclonic conditions in winter, air pollution can be imported from long-range sources and the lower temperatures favour the formation of ammonium nitrate SIA, as well as semi-volatile SOA, which could cause higher concentrations at night (McFiggans et al., 2015). There are diurnal cycles in the emissions of SOA precursors from cooking, road vehicles and domestic burning as shown in Figure 9. The solid fuel figure includes all potential sources of wood, coal, smokeless fuel and biomass combustion.



OOA = Oxygenated Organic Aerosol
HOA = Hydrocarbon-Like Organic Aerosol
COA = Cooking organic aerosol
SFOA = Solid fuel organic aerosol

Figure 9. Diurnal variation of primary organic aerosol from different sources in London and Manchester. Source: Allan et al. (2010).

Castro et al. (1999) found that the contribution of secondary organic aerosol to total PM was 17% in Birmingham in the wintertime. In the San Joaquin Valley, California, which is known to have a high level of wood burning for heating, the contribution of

wood smoke to total SOA in the winter was 16%, with the major contributors being solvent use and vehicles (Chen et al., 2010). In Zurich, however, biomass burning was found to be the main source of both total organic aerosol and SOA in the wintertime, with influences from cooking and cigarette smoke (Qi et al., 2019).

Contribution of domestic burning to SOA precursors in the UK

A comprehensive review of the sources and formation of secondary particulate matter in the UK was carried by McFiggans et al. (2015). It highlighted the lack of knowledge of SOA formation and contributing factors and the need for collaborative research in this area, including the characterisation and quantification of VOC emissions from domestic and commercial cooking and the burning of biomass for heating.

According to the Clean Air Strategy, the largest anthropogenic sources of VOC emissions in the UK are industrial processes (22%), household cleaning and personal care products (18%) and agriculture (14%). The contributions of domestic burning and transport are 5% each, and biomass burning is thought to have a sporadic short-term effect on UK air quality. The Strategy aims to reduce VOC emissions by 39% by 2030.

A comparison of the total nationwide emissions of key SOA precursor gases is given in Table 5, together with the relative contribution of domestic burning sources.

Table 5. Total UK emissions of known precursors of secondary organic aerosols for the year 2017 and the contribution of domestic burning. Data sourced from the NAEI (May 2019).

Precursor species	Total UK emissions	Domestic burning emissions		of which biomass (wood)	
	(kt)	(kt)	% total	(kt)	% total
Nitrogen oxides NOx	874	33	3.8%	5.4	0.6%
Sulphur dioxide SO_2	173	41	23.7%	0.85	0.5%
Ammonia NH_3	283	2.3	0.8%	2.29	0.8%
NMVOCs*	807	44	5.5%	33	4.1%
of which Benzene	12	5.8	48.3%	4.9	40.8%

*NMVOC = non-methane volatile organic compound

The largest contribution of domestic burning to any precursor gas is to SO_2 emissions at 24%, however the contribution of domestic wood/biomass burning is just 0.5%. On average, domestic burning sources contribute to 5.5% of NMVOC emissions, of which 4.1% is from wood or biomass burning. Benzene is clearly a significant component of domestic wood burning NMVOC emissions in the NAEI, accounting for nearly half of

the total national emissions. Nevertheless, due to the low contribution of domestic wood burning to total NOx, SO_2, NH_3 and NMVOC emissions, it is likely that the contribution of domestic wood burning to SOA and SIA loading is low.

Additionally, VOC emissions from domestic burning sources are highly variable depending on combustion efficiency, temperature and fuel type. The NMVOC emission factors in the NAEI are 6.4 g/kg for domestic wood burning and 14 g/kg for domestic coal burning for 2017. Emission factors for domestic wood burning in the EMEP/EEA inventory are 600 g/GJ for open fires (~11 g/kg) and 250 g/GJ for advanced/ecolabelled stoves (~4.5 g/kg). These values are similar to Ozgen et al., (2014) who found NMVOC emission factors as high as 18 g/kg for an open fire and as low as 0.17 g/kg for a pellet stove (Ozgen et al., 2014). VOC emission factors for cooking sources can be as high as 190 g/hour from large restaurants and as low as 11 g/hour from fast food restaurants (Wang et al., 2018). Emission factors of more than 50 g/kg were reported for forest fires (Akagi et al., 2011).

Uncertainty is very high in this area due to the numerous sources of SOA precursors and the various mechanisms by which SOA can be formed. "*Domestic burning of biomass for heating is a very understudied area, and little is known about emissions from materials and burners used in the UK. Extrapolating from wildfires, however, would suggest that the impact of changes in domestic burning will be sensed through the Automatic Hydrocarbon Network, via the growth in ambient benzene concentrations*." (McFiggans et al., 2015). Note that according to the NAEI, benzene emissions have reduced steadily from 21 kilotonnes in 2000 to 12 kilotonnes in 2017.

Due to the high uncertainty, it is recommended that further work be undertaken to better understand the contributing factors to SOA and SIA in the UK in order to determine the relative contributions of both anthropogenic and biogenic emissions.

2.3.3 Mass enhancement factors

The vast majority of air pollution sources emit primary particles as well as precursors of secondary particles. As a result of this, the total mass of particulate matter in a given volume of air will increase over time as gas-phase precursors react and form aerosols which are then captured in the solid phase. There are also natural sinks through particle deposition. As described above, SOA and SIA precursor gases react in the atmosphere with other pollutants and with radicals formed by sunlight. One example of this is particulate emissions from agriculture; primary particle emissions are very low because ammonia (NH_3) is emitted as a gas from livestock and fertiliser application. However, after emission ammonia reacts with NOx and SO_2 to form secondary inorganic aerosols, adding to PM concentrations.

The mass enhancement factor M_e is defined as the average ratio of aged particle mass (i.e. primary + secondary PM) to fresh particle mass (primary PM only). In reality, M_e varies depending on the atmospheric conditions including temperature, sunlight and

the mixture of air pollutants. Mass enhancement factors for cooking aerosols can be as high as 10 (Liu et al., 2017), whereas typical factors for diesel engines are 0.7-3.7 (Deng et al., 2017). For wildfire biomass burning, factors have been reported at 1.8-3.0 (Bian et al., 2017) and 1.42 ± 0.36 (Ortega et al., 2013) though there is considerable range.

For domestic burning sources, the PM mass enhancement effect has been measured through smog chamber studies. Here, smoke generated from an appliance is passed into a sealed chamber and exposed to a simulated atmosphere with known levels of artificial sunlight and reactants. The change in the composition of gases and the increase in the mass of PM are monitored throughout the experiment. A visualisation of a smog chamber experiment is given in Figure 10.

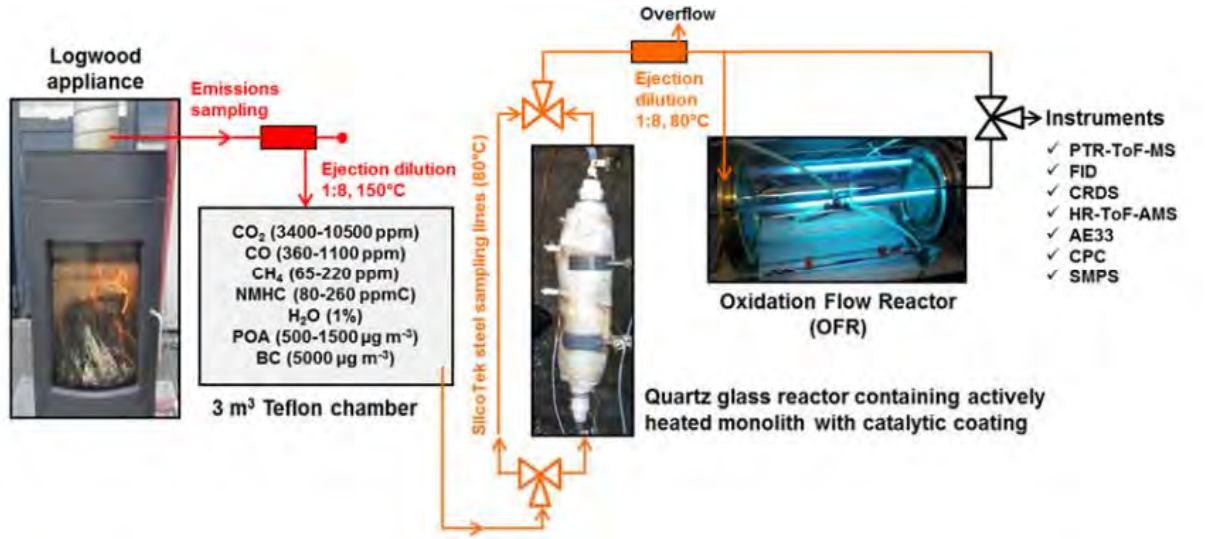


Figure 10. Smog chamber experiment for a domestic wood stove. Source: Pieber et al. (2018).

Bruns et al. (2016) simulated the ageing of smoke from a residential wood burner by injecting it into a smog chamber. The stove used was a 2009 Attika Avant and the fuel was beech logs at 19% moisture content. It found that after ~2 days of ageing, the mass of particulate was 3-7 times higher than directly emitted organic aerosol. Heringa et al. (2011) showed that the mass increase upon ageing is dependent on combustion efficiency, with particulate organic matter increasing on average by a factor of 4.1±1.4 after five hours of aging.

The authors used the same stove as Bruns et al. (2016); the 2009 Attika Avant, but also used a traditional old stove; a 1960 WESO Duplex, and a modern pellet stove; a 2005 Rüegg KEA. The results showed that both total emission factors and SOA formation were dramatically reduced in the more efficient appliances, as shown in Figure 11. It is noteworthy that there was no mass enhancement for the most efficient system, the pellet stove.

SOA formation can substantially increase the total particle emissions from more inefficient domestic burning sources, with ratios of SOA:POA of 1.5-6.0 being reported (Saleh et al., 2013, Grieshop et al., 2009b). It may therefore be concluded that increases in the efficiency of appliances can reduce not only the primary PM emissions but also the secondary PM formation.

It should be noted that some retrofit emissions abatement devices such as electrostatic precipitators or ceramic filters may reduce primary PM but not secondary PM. However, evidence from Pieber et al (2018) showed that a domestic wood burning appliance fitted with a catalytic converter could reduce SOA formation by a factor of three or more, which is achieved by minimising the emission of VOCs and aromatic hydrocarbons. Nevertheless, the most effective way to reduce both primary and secondary emissions is to increase the efficiency of combustion thereby reducing the emissions both soot particles and precursors of SOA.

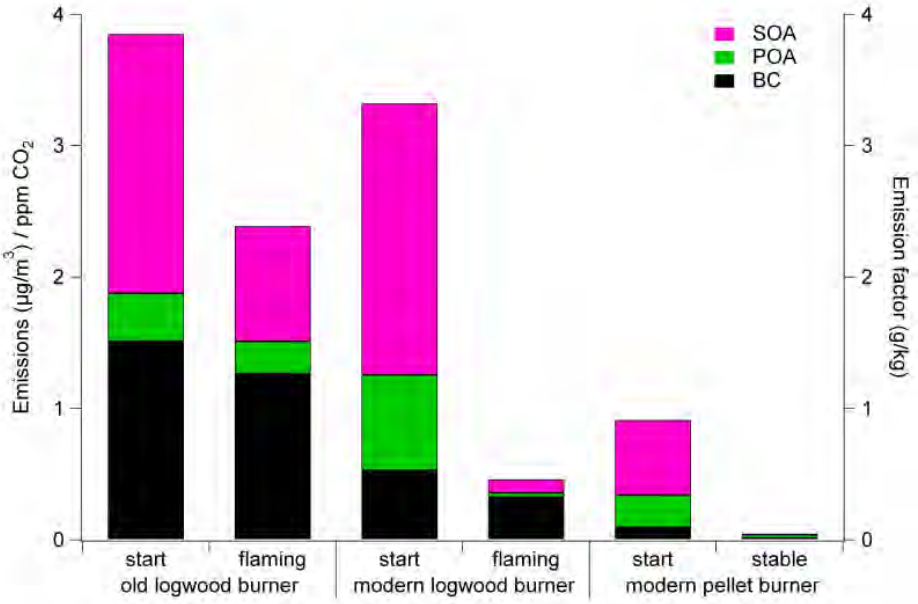


Figure 11. The effect of ageing on emissions from different domestic wood burning technologies. Source: Heringa et al. (2011).

There is evidence showing that domestic burning is an important contributor to ambient OA and SOA concentrations in many areas of the world, but the true contribution in the UK is unclear. Therefore, there is a need to better understand SOA formation from different domestic burning sources and its impacts on air quality (Hallquist et al., 2009).

2.4 Particle toxicology

The toxicity of particles may be measured in a number of ways. In some instances, living animals or human volunteers have been exposed to smoke from different sources and the effects on the body have been analysed *in vivo*. One study found that short-term exposure to residential wood smoke at typical ambient concentrations causes only mild inflammatory responses (Riddervold et al., 2012). However, the overwhelming evidence suggests that air pollution from all sources, including domestic burning, is of major detriment to health (Chafe et al., 2015).

In other cases, individual cells have been exposed to particulate samples and cell damage is measured by assays, such as the comet assay for DNA damage genotoxicity or the MTT assay for cytotoxicity. The toxicity of individual particles is determined by its internal chemical composition and any damaging compounds that may be adsorbed to the surface. Smaller particles such as $PM_{2.5}$ and PM_1 have a greater surface area for adsorption of organics which can act as a carrier deep into the lungs and even into the bloodstream. PAH compounds including benzo[a]pyrene are among the most important adsorbed species, whereas zinc and iron are among the most important internally mixed metals (Dilger et al., 2016).

Some authors have attempted to attribute the toxicity of a mixture of urban air pollution sources, for example Hannigan et al. (2005). The study investigated the source contributions to particle mutagenicity in the Los Angeles area and found that the largest contributors were natural gas combustion and diesel vehicles. Domestic wood burning contributed to on average 14% of the organic aerosol but had a lower contribution to the particle mutagenicity due to the lower relative toxicity (Figure 12).

PM from wood heating appliances has been shown to be less cytotoxic than oil-fired heating systems (Kasurinen et al., 2015), which is thought to have a higher relative toxicity (Samoli et al., 2016).

A limited number of studies indicate that PM from herbaceous biomasses incite a more severe genotoxic response than from wood (Kasurinen et al., 2016), which implies that agricultural burning and garden incinerators may be more harmful to health. Although the open burning of waste in a residential area is prohibited in the UK, the air pollution generated from ‘backyard burning’ can incite a toxic response in those exposed, particularly if the waste contains plastic packaging, PVC or refuse (Hedman et al., 2005). Burning of garden residues such as leaves, wood and trimmings is legal in residential areas, providing that it does not cause a regular nuisance, but PM emission factors can be as high as 32 g/kg for the burning of leaves (Kannan et al., 2005).

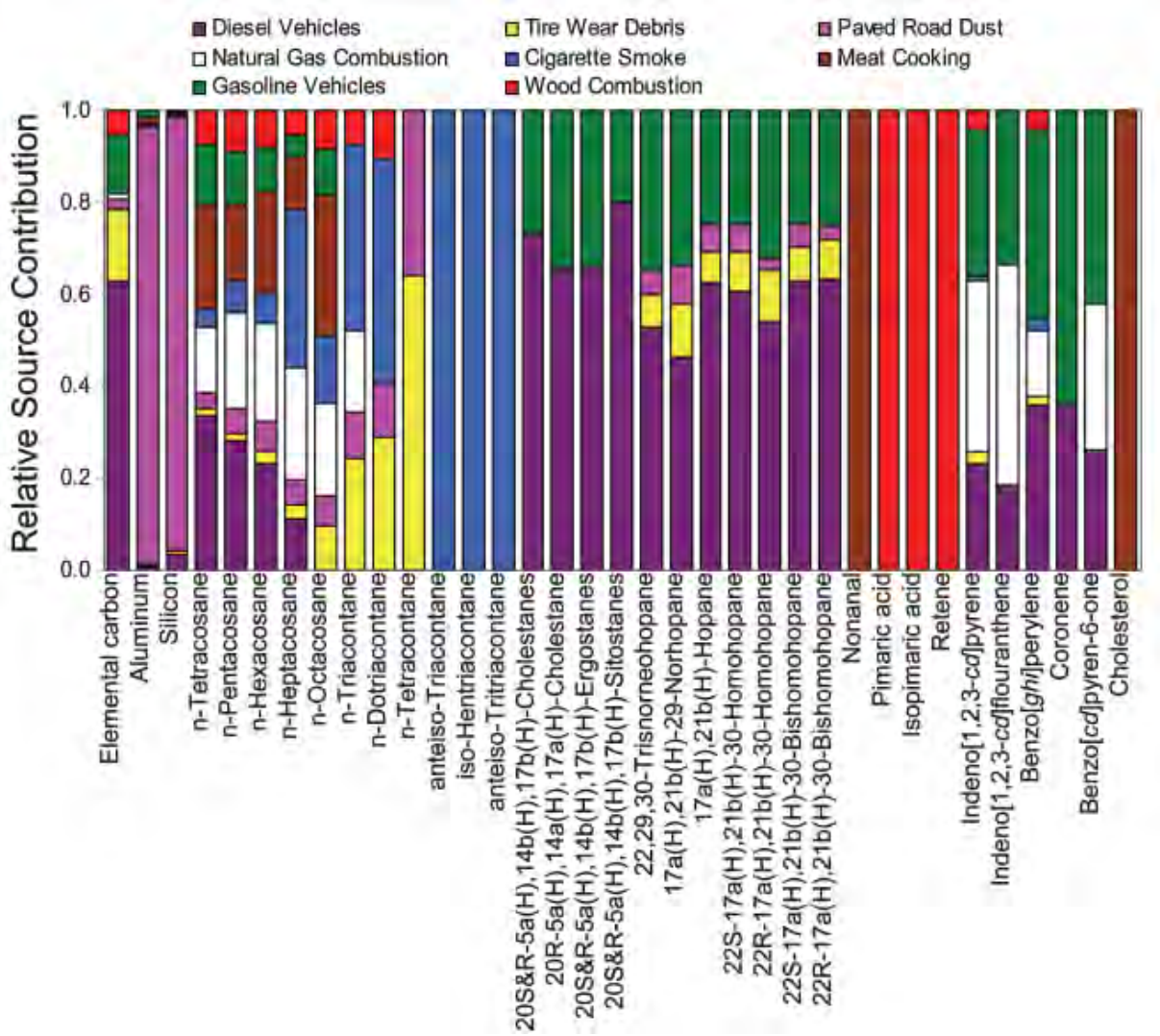


Figure 12. Relative source contributions to particulate mutagenicity in Los Angeles. Source: Hannigan et al. (2005).

Within domestic burning, the type of appliance and efficiency also impacts on the toxicological properties of emitted PM, whereby PM toxicity reduces with increased efficiency (Uski et al., 2014, Longhin et al., 2016). Tapanainen et al. (2011) found that wood PM from a traditional heater induced a 3-fold higher cell death and DNA damage rate than wood PM from an efficient pellet stove. These findings were mirrored in reviews by Naeher et al. (2007) and Bølling et al. (2009). There is no available evidence of the toxicity of PM emitted from Ecodesign ready domestic burning appliances, though the lower PAH emission factors are likely to lead to lower toxicity. PM from larger more efficient modern bioenergy systems can be >95% inorganic material, compared to 30-40% from older traditional log stoves (Vicente and Alves, 2018), and 1-56% for open burning (Hosseini et al., 2013).

A recent study by Gerlofs-Nijland et al. (2019) compared the toxicity profiles of particulate matter from different sources, including a traditional wood stove and a

modern wood stove (Fuga eL), fuelled with beech logs at 15% moisture content. The results suggest that domestic biomass burning PM is not evidently more or less harmful then mixed urban PM (mainly traffic-related). However, fuel combustion efficiency was found to be a more important factor than appliance or technology in terms of particle toxicology.

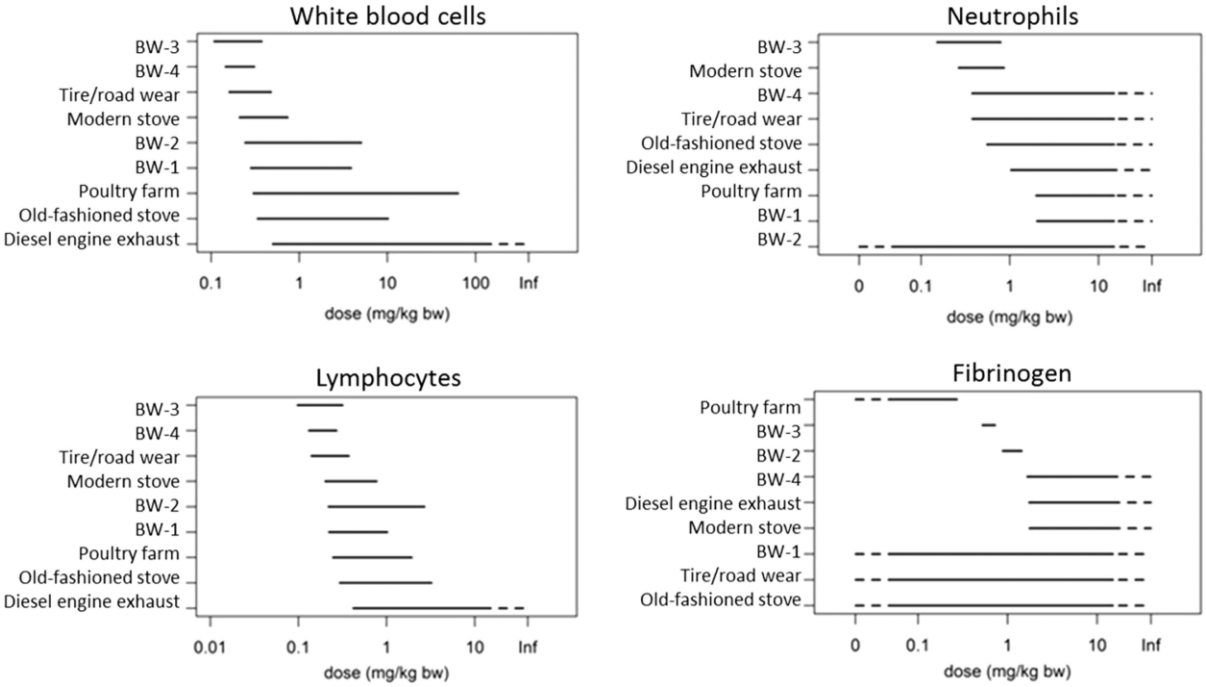


Figure 13. Ranking of PM dose range for haematological parameters resulting in 10% change in response compared to controls. Source: Gerlofs-Nijland et al. (2019).

Van Den Heuvel et al. (2018) investigated the trends between particle toxicity and biomass burning using tracers (levoglucosan, mannosan, galactosan, EC/OC and PAHs). It was found that there was an association between biomass burning tracers and particle mutagenicity, indicating that biomass burning may play a role in adverse health effects observed in areas of high pollution. The authors highlighted the essential need for further research on the impacts of biomass burning on human health in order to better understand the key toxic compounds and mechanisms.

Reis et al. (2009) assessed the intake fraction of wood smoke in urban areas and found that domestic burning may be an important contributor to urban $PM_{2.5}$ in Vancouver, Canada, with a similar intake fraction to vehicle emissions. Surprisingly, residents of more affluent areas had a lower wood smoke intake and the authors highlighted the potential health benefits of reducing wood smoke emissions.

Interventions

A report by the World Health Organisation (WHO) (Chafe et al., 2015) reviewed the evidence for interventions in areas where domestic burning is a major contributor to air pollution. The interventions included:

- ***Fuel switching***
  A coal ban in Ireland saw 116 fewer respiratory deaths and 243 fewer cardiovascular deaths. Ireland is now looking to phase out the burning of peat. Extension of smoke control area limits on coal and smokeless fuel sulphur content, and the restriction of the sale of wet wood.

- ***Improved combustion technology (stove changeout)***
  The most well-documented example of this is in Libby, Montana, USA where 95% of old stoves were replaced with EPA certified stoves. Average wintertime PM2.5 mass reduced by 27% following the change out. The contribution of domestic burning to ambient $PM_{2.5}$ reduced by a factor of 4 following a changeout program in Golden, British Colombia.

- ***In-home filters for indoor air pollution***
  Up to a 55% reduction in indoor PM levels when HEPA filters are operated

- ***Education campaigns targeting burning practices***
  Examples include Burnwise in the USA and Burnright in the UK, but to date there is little quantifiable evidence of the impacts on air quality.

- ***Bylaws/local emission standards***
  Ultra low local emissions limits in Nelson, NZ, helped reduce ambient PM by nearly 60% relative to 2001 levels.

A recent study by Bailey et al. (2019) reviewed the impacts of interventions aimed at reducing emissions from domestic burning around the world, with a view to identifying the most effective of the mitigation strategies listed above for the city of Athens. It found that the most effective interventions were those which replaced older stoves or open fires with modern low-emission equivalents. A summary is shown in Figure 14, but some interventions are missing from the map including those in Ireland, the Po Valley (Italy), Paris (France), Puget Sound (USA), Vancouver (Canada) and Southern Chile.

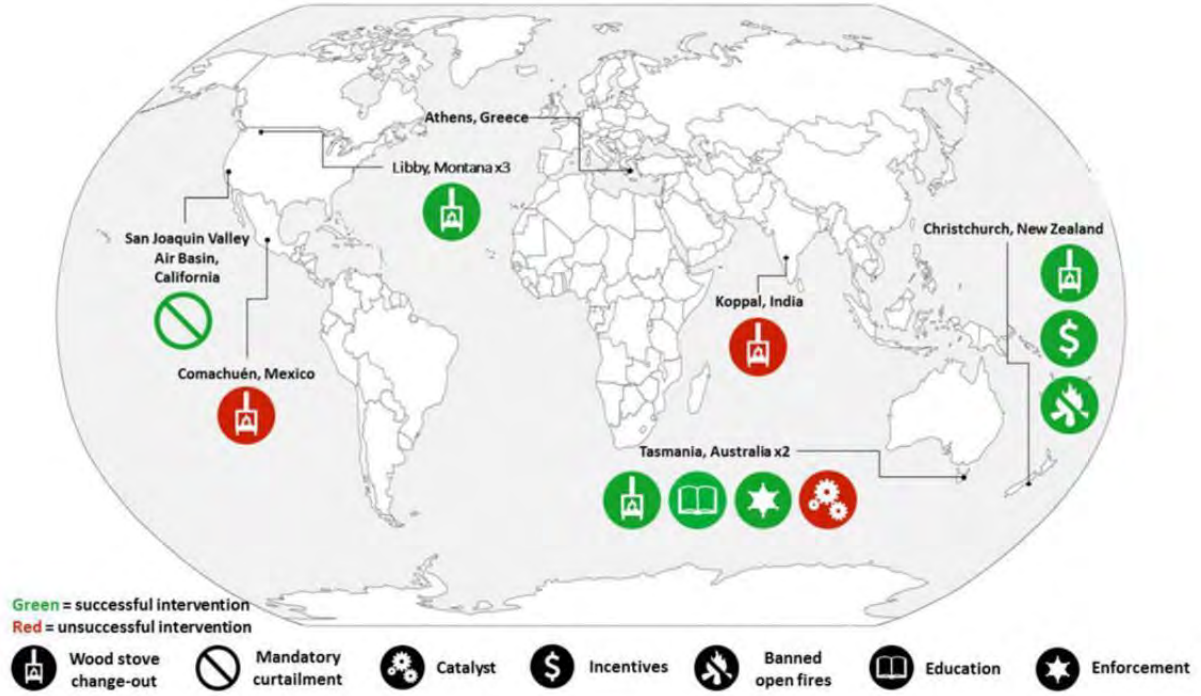


Figure 14. Summary of domestic burning interventions. Source: Bailey et al. (2019)

Among the most successful policy interventions at the European level include awareness campaigns, informal platforms, product declaration, expert advice at the site and fuel switching (Wolters, 2018).

2.5 Section 2 summary

Source apportionment (top-down) methods

- The two most common source apportionment methods for domestic burning are aethalometers and levoglucosan, but these cannot distinguish exclusively between different biomass burning sources.
- Most aethalometers used in the UK network of 14 monitoring sites are two-wavelength instruments. By measuring the difference in the ratio of light absorption at the lower wavelength and the higher wavelength, high black carbon emission sources (e.g. diesel vehicles) can be separated from high brown carbon emission sources (e.g. biomass burning).
- Using a two-wavelength aethalometer for quantitative source apportionment of domestic burning carries a high level of uncertainty because there is considerable variation in the default values of Ångström exponent used by the instrument. It is recommended that a seven wavelength version of the instrument be used in combination with another source apportionment technique which is less prone to interference.

- Levoglucosan is often used as a marker for domestic wood burning, but it is produced by the pyrolysis of cellulose and therefore is a marker for biomass burning in general which can include wildfires, prescribed burning, agricultural burning, barbecues, bonfires, cigarette smoke and some cooking.
- The amount of PM attributable to wood/biomass burning is calculated by multiplying the concentration of levoglucosan by a fixed ratio of 10.7. This is derived from a 2008 study where the ratio of levoglucosan to PM was measured in the emissions from a now 25 year old Austrian tiled stove, which may not be representative of modern UK appliances. Since levoglucosan emission factors may vary by a factor of 5 or more depending on fuel type and appliance type, the applicability of the 10.7 factor may be called into question for UK specific circumstances. We recommend that this factor be determined from a typical UK stove and fuel combination for future use.
- It is therefore recommended that source apportionment of domestic burning sources be carried out by a combination of more accurate techniques including chemical mass balance or positive matrix factorisation based on aerosol mass spectrometer data which includes a much greater range of pollutants and is better able to distinguish between biomass burning sources.

Primary and secondary emissions

- Most pollution sources, including domestic burning, emit both primary PM (that directly emitted from the stove) and precursors of secondary PM (additional particles formed by reactions in the atmosphere).
- Within primary PM, there can be reductions in emissions of 88.6% for an advanced stove/boiler relative to an open fireplace. There is also a reduction in the amount of organic material in the PM and hence a reduction in toxicity. Toxicity can be reduced by 3 times for an advanced stove compared to a traditional stove or open fire.
- Formation of secondary PM can significantly increase the mass of PM emitted by a stove. This is a result of precursor gases (e.g. VOCs and NOX) reacting in the atmosphere and forming additional particles. Evidence is presented that secondary organic aerosol formation is highest from the start-up phase and can be reduced to near zero during stable combustion in an efficient modern appliance.

Particle toxicology

Particle toxicity is highly variable from domestic burning sources depending on fuel type, fuel moisture content and combustion efficiency. Particles emitted from modern high-efficiency appliances have been found to be less damaging to cells than open

fires or older stoves. Although there is little to no evidence for the toxicity of PM emitted from Ecodesign stoves, higher efficiency and more complete combustion increases the proportion of inorganic salts as a proportion of the total $PM_{2.5}$ emissions. Hence switching to more modern appliances has health benefits not only in the reduced mass emissions of gas and particulate pollutants, but also in the relative toxicity of those particles. Evidence of this reduced health burden is presented from a series of domestic burning interventions around the world.

3. Emission inventories for domestic burning in the UK

3.1 Domestic burning activity estimations

Activity data is the amount of fuel used in a given sector in a given timeframe, usually one calendar year. This chapter reviews current estimates for domestic burning activity data in the NAEI and compares this with other evidence.

3.1.1 Activity estimations according to the NAEI

For domestic burning sources within the National Atmospheric Emissions Inventory (NAEI), activity data is presented in units of gross heat energy input. There are 14 fuels listed in this sector, consumption of which is shown in Figure 15. Activity data for solid fuels is presented in mass units (million tonnes) using the calorific values given in the NAEI background information. Calorific values are reviewed for each year of the NAEI and therefore there is some year on year variation, which is taken into account in Figure 15. The calorific value assumed for wood in the NAEI is 16.3 MJ/kg on a gross basis at 20% moisture content (equating to 14.7 MJ/kg on a net basis).

As shown in Figure 15, the most consumed fuel in the domestic sector is natural gas which was 267.3 TWh in 2017, having reduced from a peak of 356.8 TWh in 2004. On an energy basis, the second most consumed fuel is burning oil (kerosene heating oil) at 21.9 TWh followed by wood at 21.5 TWh with consumption of all other fuels being at least one order of magnitude lower.

Non-wood solid fuel consumption in the domestic sector has decreased by more than 84% since 1990; from 5.9 million tonnes in 1990 (49% coal and 18% smokeless fuels) to 0.9 million tonnes in 2017 (35% coal and 25% smokeless fuels). Including wood, total solid fuel consumption reached a low of 2.86 million tonnes in 2005 and since then it has increased by more than 98% to 5.7 million tonnes in 2017. This is due to the NAEI assuming a large increase in the amount of wood consumed in the domestic sector; increasing from 1.8 million tonnes in 2005 to 4.8 million tonnes in 2017. The underlying assumptions, data sources and uncertainties for this are discussed below.

Aside from wood, the solid fuels included in the NAEI are peat, charcoal, solid smokeless fuel (SSF), coke, petroleum coke, coal and anthracite. Wood is taken to include hardwood and softwood logs, briquettes, pellets and wood chips.

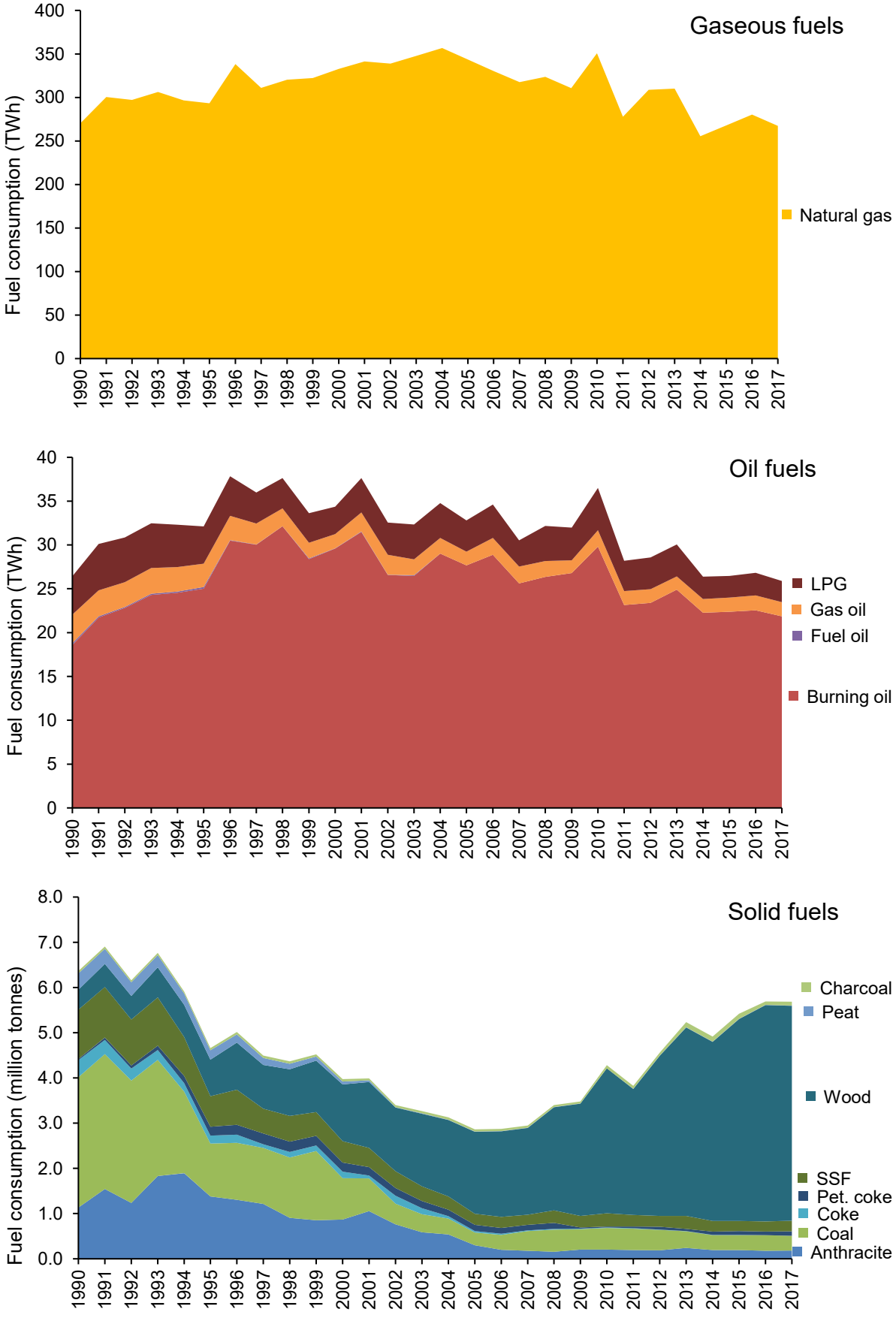


Figure 15. Activity data for domestic burning in the NAEI, 1990-2017.

The amount of wood consumed in the domestic sector was revised in the NAEI following the Government's *Domestic Wood Use Survey* published in March 2016. The survey found that domestic wood fuel use had been previously underestimated by a factor of three and therefore the estimations of activity data and emissions increased significantly in subsequent versions of the NAEI. The accuracy of this survey has been called into question by some, with the argument being made that the amount of wood consumed in the domestic sector is now overestimated in the NAEI. If the activity estimates in the NAEI were found to be overestimated, the total emissions and relative contribution of domestic burning are likely to also be overestimated.

3.1.2 The NAEI methodology for domestic burning

The NAEI is a tool used to produce the annual UK Inventory Report which is submitted under the National Emissions Ceilings (NEC) Directive and the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (CLRTAP) [see section 4.1 for more information on the UK emissions obligations].

The 14th Informative Inventory Report was published in April 2019, presenting a detailed sector-based emissions inventory for NOx, CO, NH_3, SO_2, NMVOCs, PM, PAH, POPs and heavy metals (Richmond et al., 2019). A summary of sector 1A4 (commercial, institutional and households) is given below.

Coal and coal-based fuels

Fuel consumption and activity data for domestic coal burning are derived from the Digest of UK Energy Statistics (DUKES). The proportions of each type of appliance using each fuel are estimated based on a 2007 market report as part of a preparatory study for Ecodesign lot 15, with some more detailed splits based on expert judgement. Given that this report is now 12 years old and assumptions have been held constant over the 1970-2017 timescale of the inventory, the validity and robustness of this data may be called into question.

Emissions factors are taken from the 2016 EMEP/EEA Inventory Guidebook, taking into account the types of appliances used in the UK for specific technologies. In the latest inventory year, a 4% reduction in emissions followed a revision to the DUKES value for the calorific value of coal burned in residential applications. Emission factors for SO_2 are based on UK-specific data on the sulphur content of coals and oils, provided by fuel suppliers.

Wood and biomass fuels

Activity data for residential wood combustion is also taken from DUKES and in the latest inventory year, the estimate of wood burned has been revised upward by 5% in compared to the previous year. DUKES also incorporates the 2014 DECC *Domestic*

Wood Use Survey. This revised upwards the activity data for this sector by a factor of three, which included a back-revision from 2008 and a newly derived time series for 1990-2007 from the BEIS energy statistics team. Despite the best efforts of the statisticians, the report authors note that:

"*activity data for this source category remain highly uncertain; the accurate assessment of wood use in the residential sector is extremely difficult due to the lack of comprehensive fuel sales data for a fuel with a substantial component outside conventional fuel markets*."

The 2014 survey found that around half of wood burned in that year was in open appliances (i.e. open fires), which is a similar finding to the SIA User Survey (SIA, 2019) that found 50.6% of wood is consumed in open fires and stoves older than 10 years. Details on the assumptions for fuel use and appliance population are provided in a 2015 unpublished report from Ricardo Energy & Environment to DEFRA. From this, a key assumption was that the ratio of wood fuel used in open and closed appliances was 3:1 between 1970-1990 and 1:1 between 2014-2017. This ratio can have a significant impact on total sectoral emissions due to the significantly higher emissions of open fires compared to closed stoves.

Although the 2016 update of the EMEP/EEA Guidebook revised upwards PM emission factors from pellet stoves, they remain significantly lower than other appliances and the resulting increase in PM emissions across sector 1A4bi was less than 0.3%.

Model sector uncertainty

The UK Inventory Report demonstrates the high level of uncertainty in sector 1A4 (commercial, institutional and households). Emissions are calculated using literature (the EMEP/EEA Guidebook), Tier 1 emission factors and Tier 2 emission factors for wood.

The Tiers refer to the different modelling approaches which have different levels of uncertainty given in the quality assurance and quality control (QA/QC) procedure which, for this sector, is covered by the general QA/QC of the NAEI. This states that in 2017, the estimated tier 1 uncertainty (95% C.I.) was 45% in PM_{10}, 55% in $PM_{2.5}$ and 390% in benzo(a)pyrene. The uncertainties in tier 2 pollutants were -20% to +30% for CO and benzene and -20% to +50% for black carbon and $PM_{1.0}$. The combined uncertainty for sector 1A is 55% for NH_3, 17% for SOx, 8.1% for NOx, 33% for NMVOCs, 56% for PM_{10} and 53% for $PM_{2.5}$. It should be noted that uncertainty in $PM_{2.5}$ emissions from sector 1A represent 53% as a proportional of total national emissions, which is by far the largest of any sector.

The authors of the Inventory Report state that "*the methodology and assumptions for wood* [domestic burning] *will be kept under review and improved should better data become available*."

Planned sector improvements

- The report authors aim to improve the inventory to reflect the change in emission factors over time as lower-emitting technologies have achieved greater market penetration.
- This is hindered by simplistic methodological approach and a lack of data on
    - The market share of different technologies
    - Limited set of emission factors for different technologies
    - Limited applicability of EMEP/EEA emission factors to the UK
- No current assessment of changes in technology for domestic coal burning (i.e. only wood). Hence improvements in multi-fuel stoves may not be captured
- The highest uncertainty in domestic burning inventory emissions is in those pollutants that are most affected by technology improvements (i.e. PM, CO, NOx, NMVOC and PAH)
- Since the inventory currently shows domestic wood burning to be a major source of PM and PAH emissions, this sector is a key component of overall uncertainty in the UK national inventory as a whole
- Highest priority for improvement is to
    - Gain better information of the market shares of domestic wood burning appliances
    - Improve methodology for coal and smokeless fuels

3.1.3 Other evidence for domestic burning activity

The *Domestic Wood Use Survey* found that there is considerable variation in the hours of operation for domestic wood burning appliances, with the average hours of operation being 22 hours per week in winter and 10 hours per week in summer. This also varied by appliance, with average winter hours of operation being 17 hours/week for an open fire, 27 hours/week for a closed stove and 26 hours/week for 'other' (pellet stove, manual/automatic boiler, range cooker etc.). Based on an efficiency-weighted average fuel use of 17.6 kWh for an open fire and 9.2 kWh for a closed stove, this equates to 8.6 kg of wood per day for an open fire and 7.1 kg/day for a closed stove in the wintertime. This can be compared with daily usage in other countries, as shown in Table 6.

As the table shows, average daily domestic wood consumption may be higher in some countries that have a higher dependence on wood as the primary source of heating, for example in New Zealand. However, most wood burned in stoves and fireplaces in the UK is for supplementary heating, which evidence from the USA shows may consume less than one third of that consumed for primary heating.

Table 6. Comparison of wintertime wood fuel consumption assumptions in the Domestic Wood Use Survey and other countries. Source: Mitchell et al. (2017).

MJ per day	kg per day	Notes
360	20	New Zealand survey data average
277 - 486	15-27	New Zealand real-life
238	13	USA – primary heating
76	4	USA – secondary heating
154	8.6	UK – open fire
128	7.1	UK – heating stove

Within the fuels referred to as “wood”, there are significant variations in the fuel properties and emissions. Higher density fuels such as synthetic logs or hardwoods such as beech or ash may have lower PM emission factors compared to softwoods such as spruce or pine, which contain high levels of resin. If the air supply to the appliance is not adjusted for the fuel type, less dense fuels may burn too quickly resulting in high soot emissions. In addition, the fuel moisture content has a major bearing on emissions with higher moisture fuels emitting more tars and organics than drier fuels. Through the Clean Air Strategy the Government has moved to restrict the sale of ‘wet’ wood with a moisture content of more than 20%, but according to the Domestic Wood Use Survey, 31% of users sourced fuel from the informal “grey” wood market with an unknown moisture content. The survey also found that 22% of users burned waste wood. A growing number of users are looking to burn ‘eco’ fuels derived from waste products or energy crops (e.g. coffee logs and miscanthus briquettes) and the impact of this on total domestic burning emissions is unclear at the present time.

The seasonality of domestic burning emissions must also be taken into account in activity data used in models. The NAEI uses annual emissions data and therefore seasonality is not taken into account. Domestic burning emissions have been shown by many authors to peak in the winter when the space heating demand is greatest, whereas emissions from barbecues and wildfires are greatest in the summer. With this knowledge, a seasonal regression analysis has been applied to the annual data given in the NAEI for domestic solid fuel burning and the results are presented in Figure 16. The seasonal regression for domestic heating has been derived from that presented by Mitchell et al. (2017).

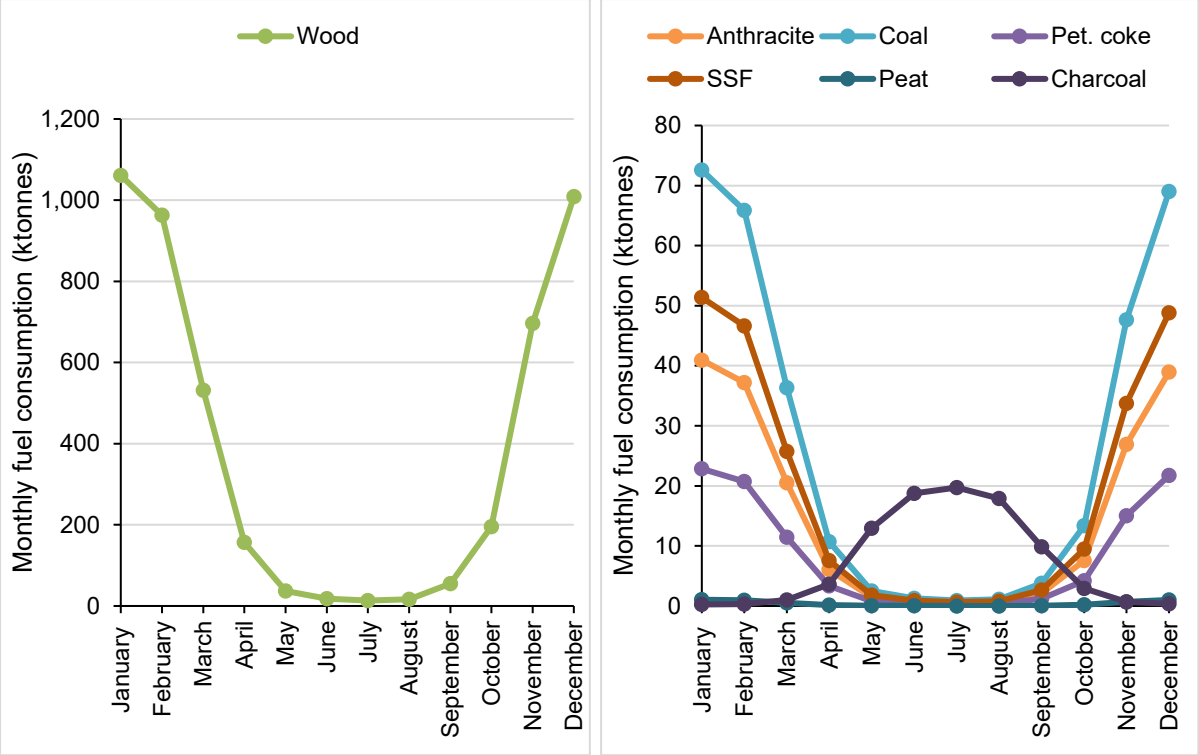


Figure 16. Calculated seasonal variation in activity data based on NAEI annual data.

Based on this analysis, 1.06 million tonnes of wood is burned in the peak month of January which equates to 16.7 kg/day in each of the 2.05 million households using wood fuel in the UK. Although consumption may be this high in some properties with a high wood fuel demand, it is very unlikely that the average across all properties will be this high without further supporting evidence. It is therefore recommended that a comprehensive review of domestic burning activity data be carried out, taking into account seasonal variations in more detail than the *Domestic Wood Use Survey*.

Of the total amount of fuel wood imported into the UK, it is estimated that 93 thousand tonnes were imported for domestic applications (HMRC, 2019). This fuel wood includes logs, billets, twigs, faggots and agglomerated sawdust and wood scrap in similar forms. Wood pellets and chips are excluded in this as they are predominately used in power stations and commercial boilers. The figure includes only imported wood fuel and it is not known what the split is between indigenously grown wood logs and imported wood logs used in homes. However, it is known that 73 thousand tonnes of wood fuel meeting the above criteria was exported in 2018, with a peak of 726 thousand tonnes exported in 2012.

In 2019, the Stove Industry Alliance (SIA) published the results of an industry-led survey into the habits of wood fuel users in the UK (SIA, 2019). A comparison of some of the key results of the industry survey with the previous BEIS Domestic Wood Use Survey is presented in Table 7.

Table 7. Comparison of the 2016 domestic wood use survey and the 2019 industry survey.

	Industry Survey	BEIS Domestic Wood Use Survey
Number respondents using wood fuel	10,620	1,206
Average weekly hours of use in winter	35.8	22.3
Average number of months in use	4.7	4.8
Total annual wood use	1.87m tonnes	65,077,549 GJ (4.43m tonnes)

The results of the survey suggest that domestic wood consumption may have been overestimated in the BEIS survey by as much as a factor of 2.4. This is likely to have a significant effect on total emissions in the NAEI and on the relatively source contributions.

3.2 Annual and diurnal variations in domestic burning

The trends in annual and diurnal atmospheric PM levels have been used to support source appointment studies. Figure 17 reports the monthly mean $PM_{2.5}$ concentrations measured across the UK monitoring network in 2018 (Defra, 2018). The levels seen at both types of monitoring sites show the same trends, with peaks occurring during the spring and winter months. In urban areas the greatest mean concentration was seen in May (13.2 µg/m^3) followed by a peak in November. Defra specifically stated that "burning of wood and coal by households in stoves and open fires is a large contributor to emissions of particulate matter both in the UK and across Europe and is more common in winter months". They then stated that "there are a large number of emission sources for particulate matter and there may be other sources that contribute to this pattern". In the report, no reasoning or source appointment was mentioned in regard to the highest concentration's levels seen in May.

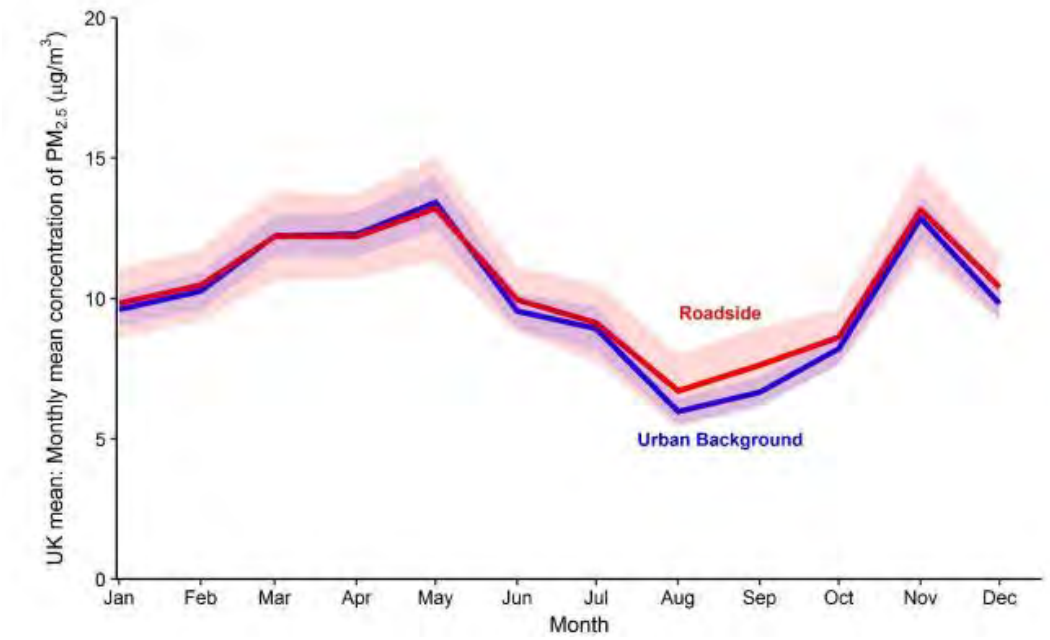


Figure 17. Monthly mean $PM_{2.5}$ concentration at roadside and urban background sites in the UK in 2018 (shading represent 95% confidence level) (Defra, 2018)

Font and Fuller (2017) used the DEFRA Black Carbon Network to estimate the contribution of wood smoke mass concentrations to PM levels at 21 urban background, suburban and rural sites from January 2009 to February 2016. They reported that "as expected" the pollution from wood burning was greatest in the winter and almost absent in the summer. The authors reported that the mean wintertime contribution varied between cities, ranging between 0.2 and 2.7 µg/m^3, and on an annual basis $PM_{2.5}$ ranged between 4 to 6% across rural areas and 6 to 9% in urban areas. When considering secondary pollution formation, the authors estimated that wood burning was responsible for between 23 and 31% of total urban derived $PM_{2.5}$ in London and Birmingham.

Font and Fuller (2017) investigated trends at three urban locations, Glasgow Centre, Manchester Piccadilly, and London North Kensington, and a rural site, Harwell, shown in Figure 18. The four sites show the highest levels occurred in winter with concentrations decreasing through spring and low levels reported in summer. The May peak in $PM_{2.5}$ reported by DEFRA (2018) is not seen at any of the sites.

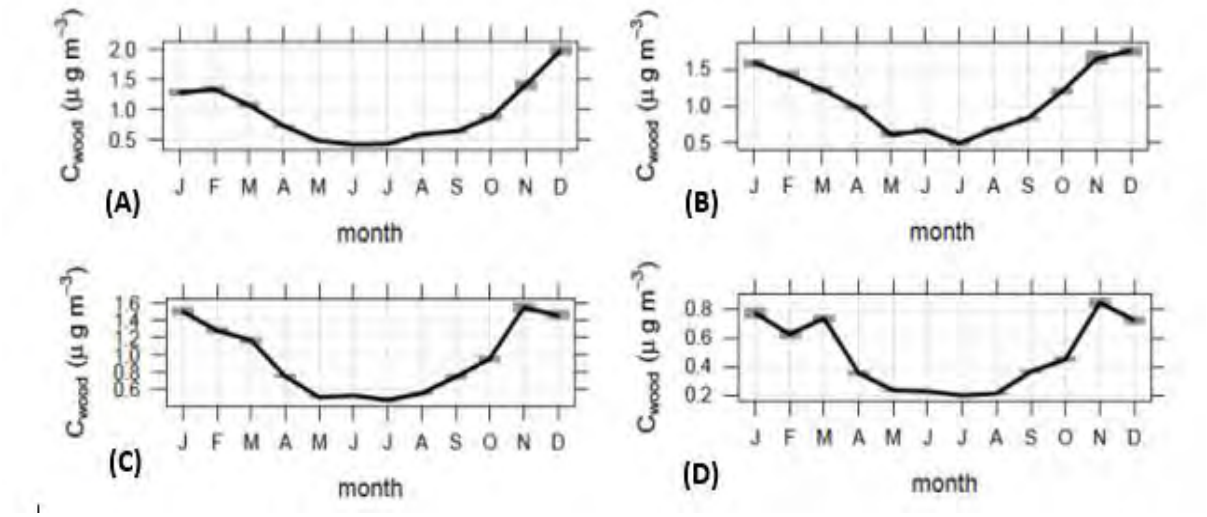


Figure 18. Mean monthly wood smoke levels, (A) Glasgow Central, (B) Manchester Piccadilly, (C) North Kensington and (D) Harwell (Font and Fuller 2017)

Diurnal PM trends have also been used to attribute emissions to solid fuel combustion in homes. Figure 19 highlights the $PM_{2.5}$ concentrations at roadside and urban background sites in 2018. The highest levels at both types of monitoring sites peak between 8pm and 9pm and DEFRA again suggested that this is the result of "*households burning, wood coal and other solid fuels in stoves and open fires for heating*".

Font and Fuller (2017) also reported the diurnal trends for the four locations shown in Figure 18. As is shown in Figure 20, at the Glasgow Central site the lowest levels are seen at 5-6am, after which they increase and stabilise by *ca*. 10am before declining after *ca*.10pm during the week. At weekends the trends continue to increase after 6am peaking at *ca.* 11pm. At the Manchester site the same early morning and midday trends are seen with concentrations slightly enhancing between 8pm and midnight.

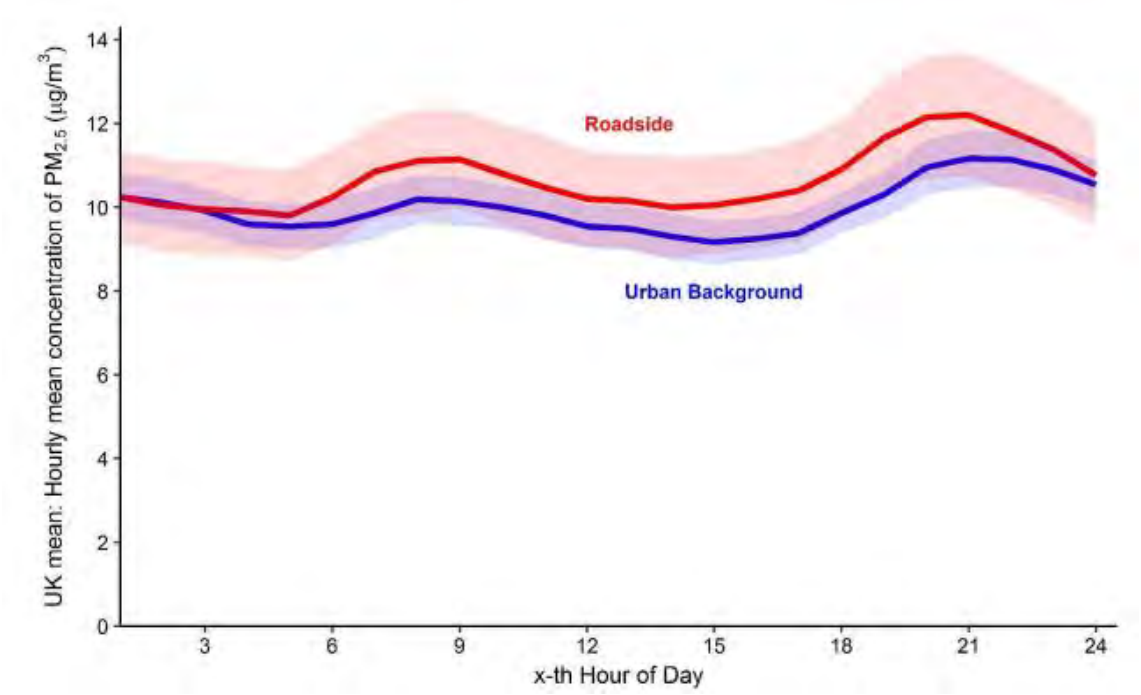


Figure 19. Hourly mean $PM_{2.5}$ concentration at roadside and urban background sites in the UK in 2018 (shading represent 95% confidence level) (Defra, 2018)

The North Kensington site observed declining concentrations in the early hours of the morning followed by a peak between 6 and 7am during the week days. Concentrations reduce through midday then increase reaching a peak at *ca*. 8pm. The Harwell site showed the same trends as the North Kensington site, although with a lower total concentration.

The increase seen at the North Kensington and Harwell sites are also seen in the DEFRA $PM_{2.5}$ figure but not to the same extent. The DEFRA data shows that the peak at 6am is almost as high as the evening peak seen at between 8 and 9pm while the evening peak reported by Font and Fuller is significantly higher.

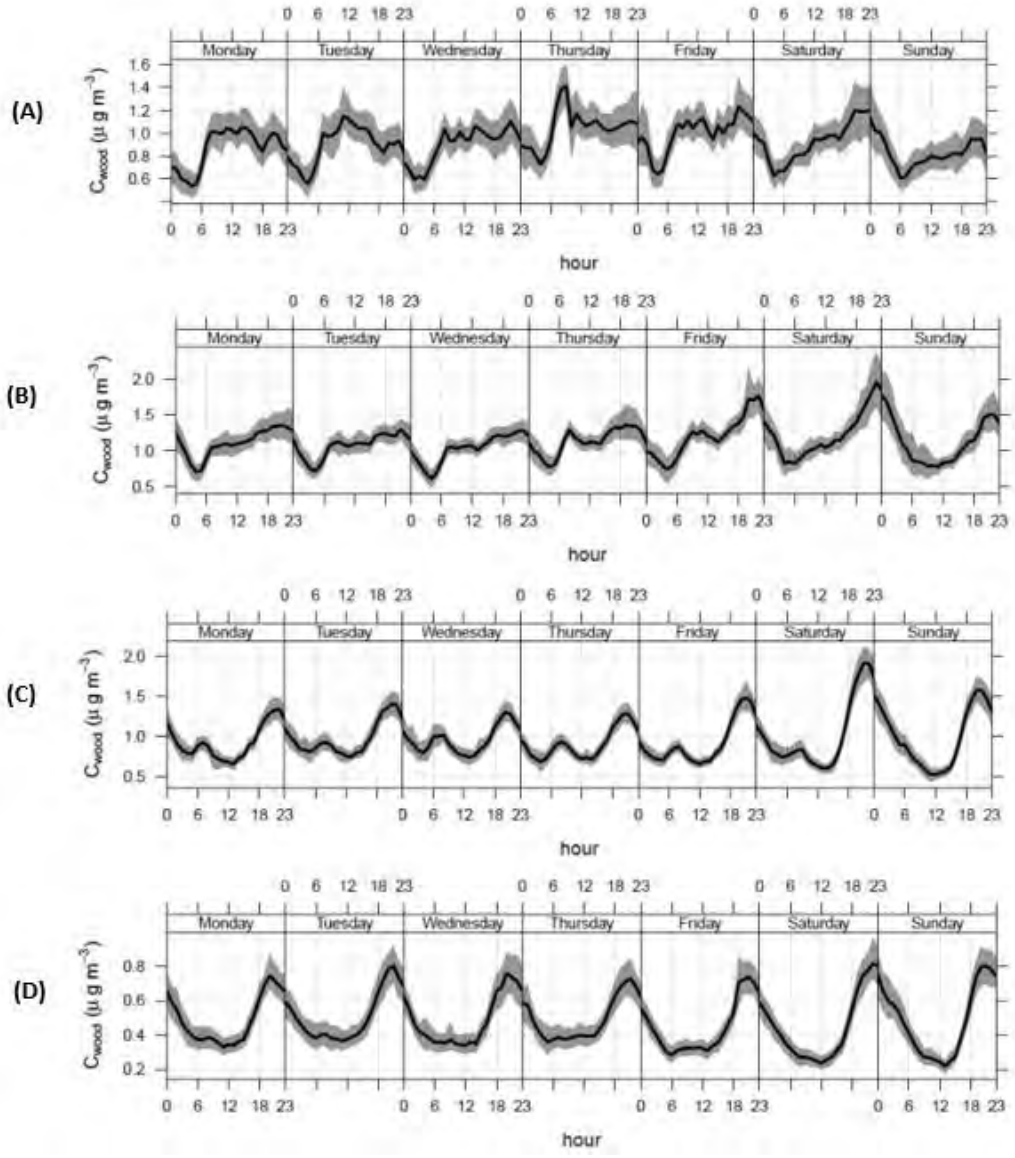


Figure 20. Mean hourly variation per day of the week of wood smoke for (A) Glasgow Central, (B) Manchester Piccadilly, (C) North Kensington and (D) Harwell (Font and Fuller 2017).

3.3 The contribution of domestic burning to total UK emissions

3.3.1 Emissions contributions according to the NAEI

This section summarises the total emissions for the domestic burning sector (category 1.A.4.b.i) according to the NAEI as of June 2019. The latest inventory year available is for 2017. The results shown here are reproduced from the NAEI in order to provide clarification of the different fuel contributions to this category. As such, these values have not been amended or altered in anyway and are reproduced here for the purposes of discussion.

As shown in Figure 22, natural gas combustion is the largest contributor to greenhouse gas emissions in the domestic sector, followed by burning oil. GHG emissions from the combustion of solid fuels in the domestic sector have reduced by 81.3% since 1990 to 3.4 million tonnes of CO_2e in 2017. This is largely due to reductions in the amount of coal and peat used for space and water heating. Although GHG emissions from solid fuel have not reduced significantly since 2005, they were 57% of the GHG emissions of burning oil and 6% of the emissions of natural gas.

Wood is a renewable biomass resource that is supported by the Renewable Heat Incentive (RHI) and has significantly lower lifecycle GHG emissions than natural gas, heating oil, coal and smokeless fuel. A comparison of GHG emissions factors for different heating fuels is shown in Figure 21.

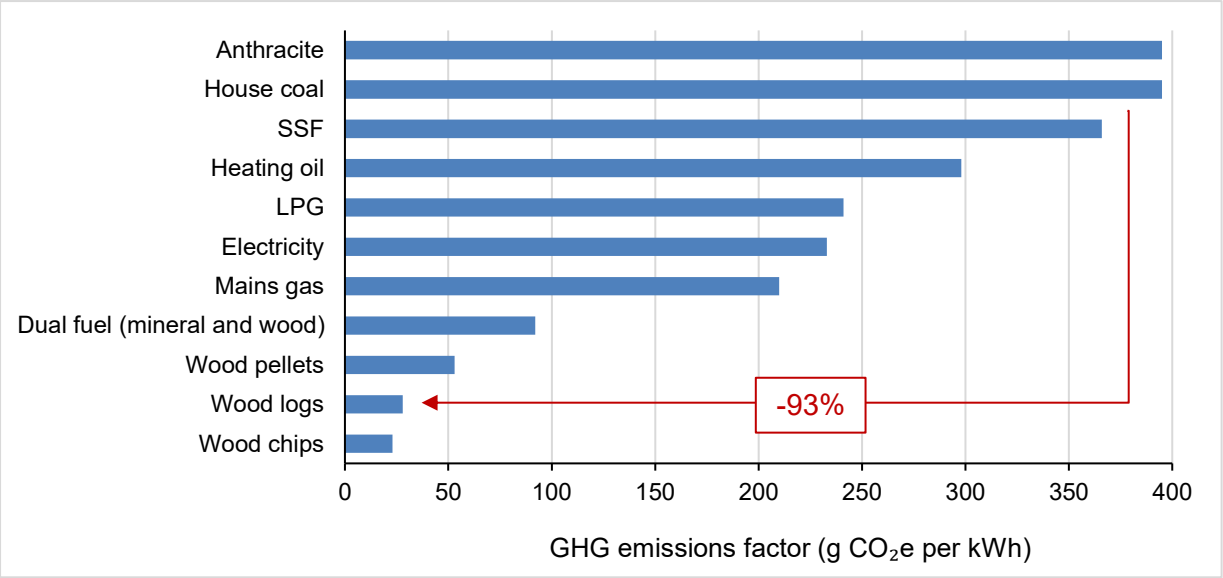


Figure 21. Greenhouse gas emission factors for different heating fuels. Source: SAP 10.0[2].

As shown in Figure 21, efficient modern bioenergy heating systems have the potential to significantly reduce greenhouse gas emissions from domestic heating. However the more inefficient the appliance, the greater the wasted heat and the greater the emissions of black carbon, which has a global warming potential of 900 (Bond et al., 2013). Particulate emissions from domestic burning are therefore of crucial importance, not only to air quality and health but also to climate change.

PM_{10} emissions from solid fuels (excluding wood) have reduced considerably since 1990, in line with the reductions in activity data (Figure 23). Total PM_{10} emissions from domestic burning were 45.1 kilotonnes in 2017, of which 43.6 kilotonnes were from solid fuels and 38.8 kilotonnes were from wood burning.

[2] Available at https://www.bregroup.com/sap/sap10/

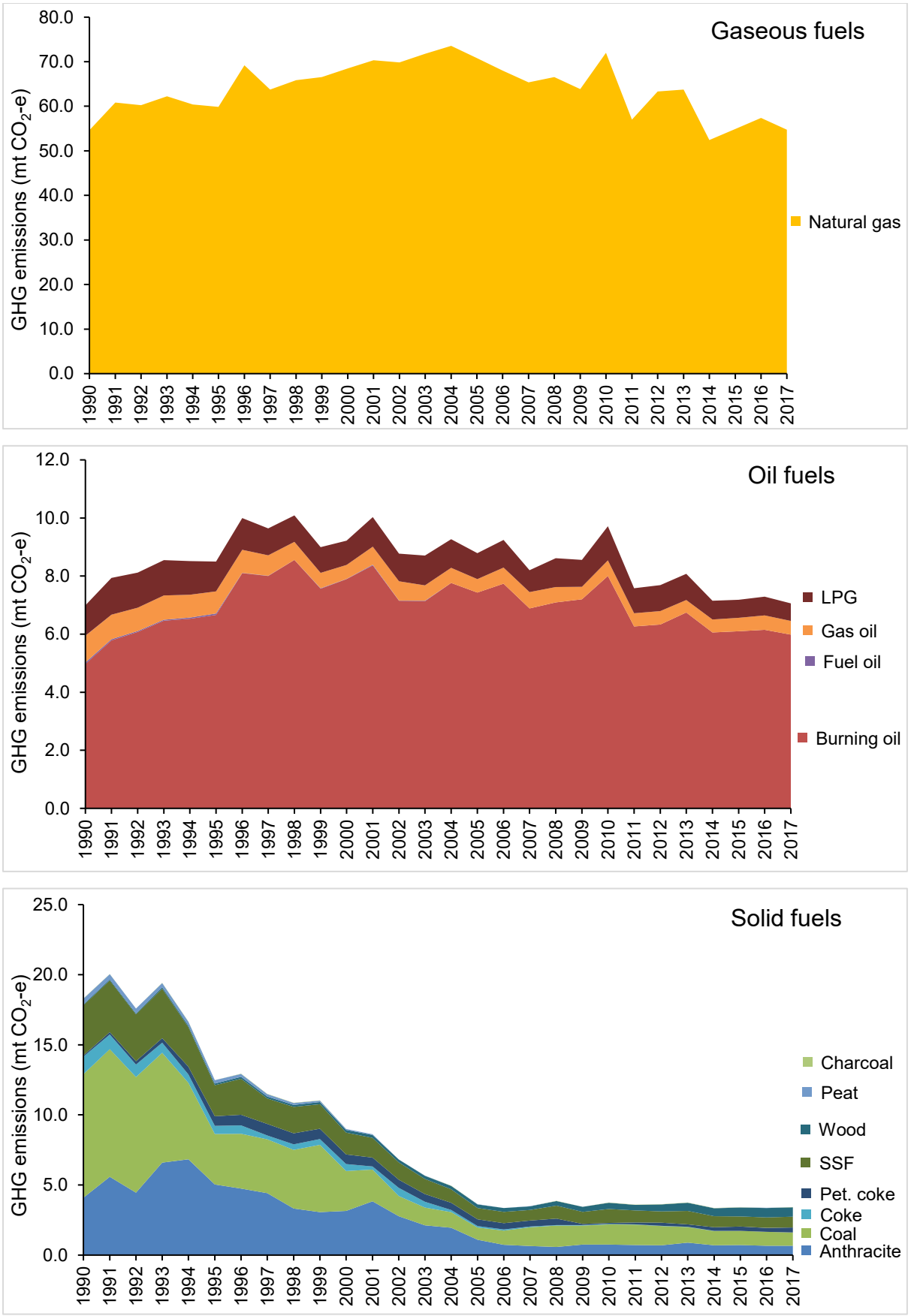


Figure 22. Greenhouse gas emissions from domestic burning, 1990-2017.

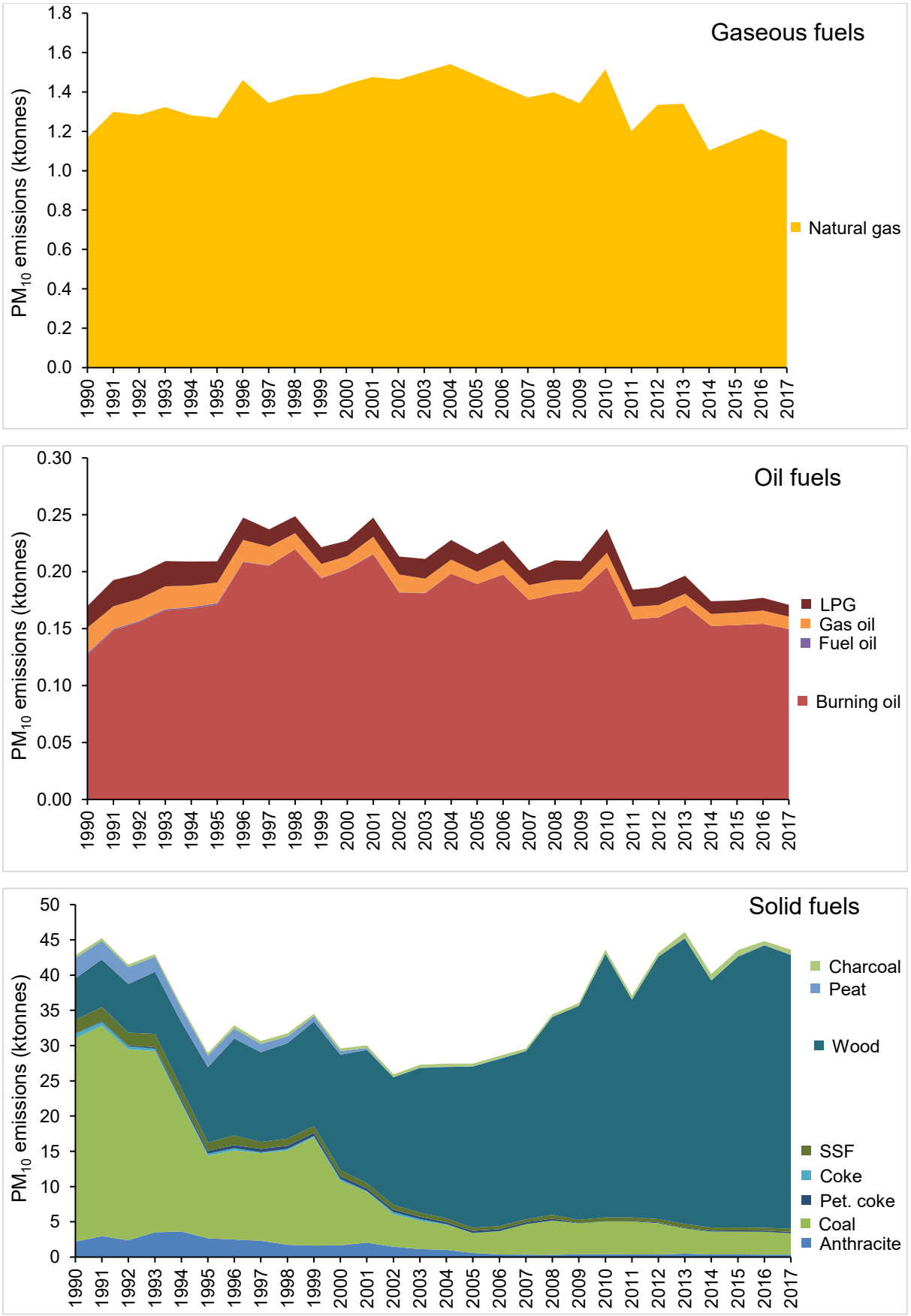


Figure 23. PM_{10} from domestic burning, 1990-2017.

The contributions of the domestic burning sector and domestic wood burning have been calculated for the year 2017 from the NAEI accessed in June 2019, as presented in Figure 24.

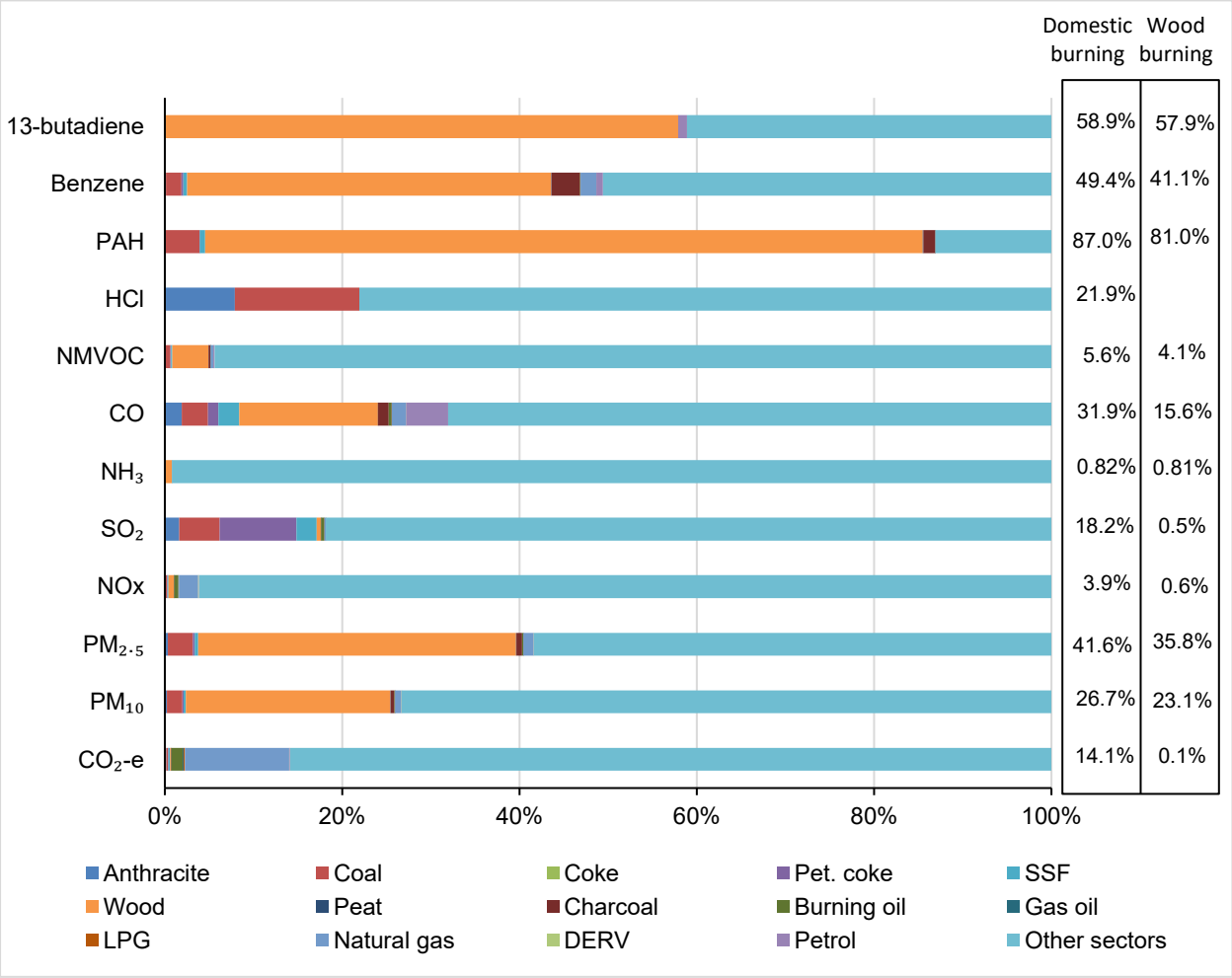


Figure 24. Contributions of domestic burning to total UK emissions of air pollutants in 2017 according to the NAEI in June 2019.

The data shows that according to the NAEI, domestic burning is a major contributor to national PM_{10}, $PM_{2.5}$, CO, benzene and 1,3-butadiene emissions. This includes all solid, liquid and gaseous fuels burned in category 1A4bi (residential stationary combustion) and 1A4bii (residential mobile combustion; lawnmowers, strimmers etc.). The largest contribution is to polycyclic aromatic hydrocarbons (PAH) at 87%, followed by 1,3-butadiene at 58.9%, benzene at 49.4% and $PM_{2.5}$ at 41.6%. Wood burning is the single largest contributing fuel to domestic burning emissions, with the exception of SO_2, CO_2-e and HCl. Wood burning contributes to 23.1% of national PM_{10} emissions and 35.8% of national $PM_{2.5}$ emissions, which is likely to be due to differences in the size fractions of particles emitted from other sectors. More than 90% of the particles emitted from wood burning are believed to be below 2.5 microns in diameter.

3.3.2 Revised estimates of wood burning emissions

Given the findings of section 3.1.3, activity data for domestic wood burning is likely to have been overestimated. Total emissions in the NAEI are the product of activity data and emissions factors and therefore if the activity data is reduced, the total emissions described above will also be reduced.

Assuming that the true activity data is a factor of 2.4 lower than previous estimates, the total PM_{10} emissions from domestic wood burning will reduce from 38.8 kilotonnes to 16.2 kilotonnes. This would reduce the contribution of domestic wood burning from 23.1% of PM_{10} to 9.6% of PM_{10} and from 35.8% of $PM_{2.5}$ to 14.9% of $PM_{2.5}$ on an annual basis.

3.4 Emissions factors for domestic burning sources of air pollution

3.4.1 Regulatory emissions limits

There are a number of regulatory emissions limits which apply to domestic burning appliances, including the Clean Air Act, the Renewable Heat Incentive, technology- and nominal power-based British standards, and most recently the European Ecodesign Directive.

The overarching emission limits for domestic burning appliances is the Clean Air Act of 1956, revised in 1993. The Act established smoke control areas in which the emission of visible black smoke is prohibited. Exemptions are available if the emitter has been found to be using an appliance or fuel that has been approved. A fuel is exempt if it has a sulphur content of less than 2% and the smoke emission rate is less than 5 g/hour when tested to BS 3841 (total gravimetric PM). An appliance ≤44 kW is exempt if the smoke emission is 5 g/hour + 0.1 g per 0.3 kW when tested to BS PD 6434:1969.

Some biomass boilers and pellet stoves may also be eligible for payments under the domestic Renewable Heat Incentive (RHI). The emissions limits under the RHI are 30 $g.GJ^{-1}$ for PM and 150 $g.GJ^{-1}$ for NOx, and any domestic biomass boiler or a non-domestic biomass boiler less than 45 kW_{th} must be certified through the Microgeneration Certification Scheme (MCS). The majority of wood burning stoves, fireplaces, chimineas and firepits are therefore not eligible. The MCS includes limits for solid fuel room heaters under BS EN 13240:2001 and for pellet-fired room heaters up to 50 kW under BS EN 14785:2006, as summarised in Table 8. Solid biomass boilers <45kW must not exceed class 5 of BS EN 303-5:2012, as summarised in

Table 9.

Table 8. Emissions limits for pellet heaters up to 50 kW output according to BS EN 14785:2006.

	CO	Efficiency
Unit	g.GJ^{-1}	%
Nominal output	242	75
Reduced output	362	70

Table 9. Emissions limits for biomass boilers under BS EN 303-5:2012.

	Power (kW)	CO			OGC			PM		
		g.GJ^{-1}								
		Class 3	Class 4	Class 5	Class 3	Class 4	Class 5	Class 3	Class 4	Class 5
Manual	≤ 50	2415	580	338	72	24	14	72	36	29
	> 50 ≤ 150	1208			48			72		
	> 150 ≤ 500	580			48			72		
Automatic	≤ 50	1449	483	242	48	14	10	72	29	19
	> 50 ≤ 150	1208			39			72		
	> 150 ≤ 500	580			39			72		

A list of suitable fuels is available, and the size, shape and layout of the fuel bed is specified in some standards. The fuel moisture content is typically 10-12% for pellets, 15-25% for logs and up to 35% for chip.

Additional emissions limits are placed on appliances at the European level, most notably through Ecodesign Directive 2009/125/EC. The two key regulations for domestic burning appliances under Ecodesign are Regulation 2015/1185 (local space heaters <50 kW) and Regulation 2015/1189 (solid fuel boilers <500 kW). The regulations will apply to all new appliances from 1 January 2022, although many are currently “Ecodesign ready” and are labelled accordingly. Ecodesign regulations include efficiency limits and emission limits for PM, CO, NOx and OGC, which are summarised in Table 10.

Under Ecodesign, there are three standard European test methods that can be followed by test laboratories in different member states; principally NS 3058/ NS 3059 in Norway, DIN-plus in Germany and BS PD 6434 in the UK. Each test method uses different test conditions and sampling equipment, which are discussed and compared in Mitchell et al. (2017) and in the Ricardo report to DEFRA (Stewart, 2017). Ecodesign emission limits are expressed as a concentration at a given oxygen concentration, which is converted to a grams per unit fuel energy input assuming a given standard dry flue gas volume (SDFGV). SDFGV is calculated from fuel composition and varies

for different fuel types, but standard values are used in emissions test laboratories which are available from Ricardo.

Table 10. Emissions limits for biomass heating systems under the Ecodesign regulations.

		CO	Efficiency	PM		OGC	NOx
Unit		**g.GJ^{-1}**	**%**	**g.GJ^{-1}**	**g.kg^{-1}**	**g.GJ^{-1}**	**g.GJ^{-1}**
Local space heaters <50kW	Open fronted	1328	30		6	80	133
	Closed fronted (non-pellet)	996	65		2.4-5.0*	80	133
	Closed fronted (pellet)	199	79		1.2-2.5*	40	133
Solid fuel boilers <500kW	Automatic feed	242	75 if <20kW, 77 if >20kW			10	97
	Manual feed	338				14	97

*Ranges are due to the ability to test to three separate European standards

3.4.2 Emission factors used in models

Emissions factors are reviewed and updated for each inventory year in the NAEI and are expressed as the mass of pollutant emitted per unit fuel input. This is usually kilotonnes per megatonne for solid and liquid fuels, and kilotonnes per megatherm for gaseous fuels (1 megatherm = 105.5 TJ = 29.3 GWh). In order to facilitate comparison, emission factors have been converted to g/GJ in Table 11 and g/kg in Table 12.

Table 11. NAEI emission factors for domestic heating fuels in grams per GJ of fuel input (gross basis) in 2017.

	Anthracite	Charcoal	Coal	Peat	Pet. Coke	SSF	Wood	Burning oil	Gas oil	LPG	Natural gas
13-Butadiene	-	-	-	-	-	-	12.5	-	-	-	-
NH_3	0.80	-	0.80	-	-	-	26.4	-	-	-	-
Benzene	2.2	148.2	21.6	23.6	6.1	6.6	57.7	0.055	0.053	0.34	0.20
CO	4,616	7,119	4,649	3,050	4,616	4,616	2,856	54	54	24	23
NOx	127	102	118	61	127	127	63	48	48	48	18
NMVOC	49.8	600.0	488.4	457.5	137.0	149.7	391.3	1.1	1.1	3.8	2.2
SO_2	425.0	11.0	810.2	8.4	3,977	489.0	10.0	7.4	7.8	0.28	0.27
PM_{10}	55.6	267.8	307.0	617.2	53.1	55.0	455.2	1.8	1.8	1.14	1.04
$PM_{2.5}$	52.7	261.0	303.5	601.6	53.1	55.0	442.9	1.8	1.8	1.14	1.04

Table 12. NAEI emission factors for domestic heating fuels in grams per kilogram of fuel (as received basis) in 2017.

	Anthracite	Charcoal	Coal	Peat	Pet. Coke	SSF	Wood	Burning oil	Gas oil	LPG	Natural gas
13-Butadiene	-	-	-	-	-	-	0.20	-	-	-	-
NH_3	0.027	-	0.023	-	-	-	0.43	-	-	-	-
Benzene	0.075	4.4	0.62	0.30	0.22	0.22	0.94	0.0025	0.0024	-	-
CO	157.6	210.0	133.3	39.0	165.1	151.0	46.4	2.5	2.4	-	-
NOx	4.3	3.0	3.4	0.78	4.5	4.2	1.0	2.2	2.2	-	-
NMVOC	1.7	17.7	14.0	5.9	4.9	4.9	6.4	0.051	0.048	-	-
SO_2	14.5	0.32	23.2	0.11	142.2	16.0	0.16	0.34	0.35	-	-
PM_{10}	1.9	7.9	8.8	7.9	1.9	1.8	7.4	0.083	0.081	-	-
$PM_{2.5}$	1.8	7.7	8.7	7.7	1.9	1.8	7.2	0.083	0.081	-	-

As shown, the $PM_{2.5}$ emission factors for domestic wood and coal burning are 7.2 g/kg and 8.7 g/kg respectively. This equates to 443 g/GJ and 304 g/GJ, where the gross calorific values are 28.7 GJ per tonne of coal and 16.3 GJ per tonne of wood (assuming a 20% moisture content).

At the current time it is unknown what the assumptions and data sources behind these values are, but the data shows that $PM_{2.5}$ emissions factors have reduced since 1990, as shown in Figure 25. This is perhaps due to efficiency improvements in domestic burning appliances leading to lower emissions factors, but as shown the NAEI assumes emissions factors to be significantly higher than the limit values for Ecodesign and for the RHI. In order to understand what the most appropriate emission factors are to be used in the NAEI, it is crucial to have an understanding of the type and age of appliances installed and the variability in emissions. The *Domestic Wood Use Survey* found that nearly 70% of open fires were installed more than 15 years ago whereas there is a greater spread in ages for closed stoves; 53% of which were 2-15 years old.

Most countries in Europe derive emissions factors for use in models and inventories from the EMEP/EEA air pollutant emission inventory guidebook (EMEP/EEA, 2016). The guidebook is designed to facilitate reporting of emission inventories by countries to the UNECE Convention on Long-range Transboundary Air Pollution and the EU National Emission Ceilings Directive, through which a number of categories are assigned including category 1.A.4.b residential combustion.

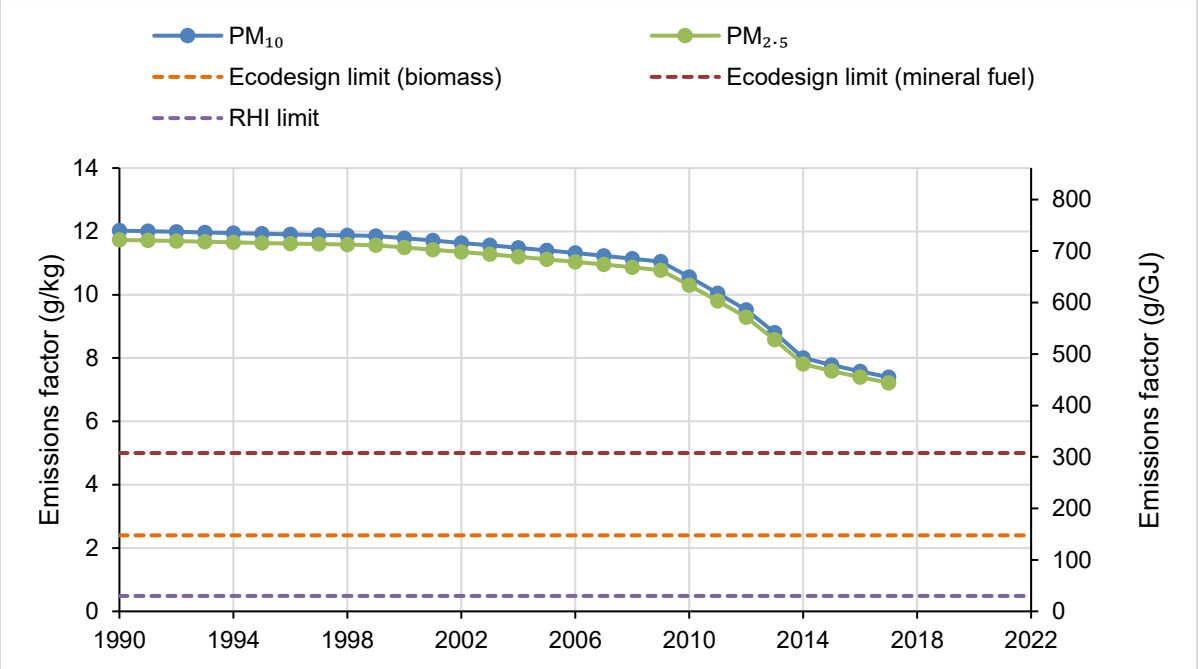


Figure 25. Comparison of $PM_{2.5}$ emission factors for domestic wood burning in the NAEI over time.

The default PM and $PM_{2.5}$ emission factors for domestic burning of wood in category 1.A.4.b.i are 800 g/GJ and 740 g/GJ respectively. The 95% confidence interval is 400-1,600 g/GJ and 370-1,480 g/GJ which reflects the variability and uncertainty in this area. It should be noted that these emission factors are stated on a net calorific value and a dry fuel basis, rather than on a gross basis at 20% moisture content as is used in the NAEI. The variability in particulate matter emissions factors between different technologies is illustrated in Figure 7. Further details on the $PM_{2.5}$ emission factors and 95% confidence interval for wood burning technologies with sector 1A4bi are given in Table 13, together with a reference to the original source of information.

As shown in Table 13, there is considerable variation and uncertainty in emission factors for domestic burning technologies with the 95% confidence interval being plus or minus a factor of 2 from the average value. It also shows that high efficiency modern appliances typically have emission factors less than 100 g/GJ so significant emissions savings could be achieved by replacing open fires and older stoves with modern Eco-design ready appliances. The values used are also derived from just one or two references in most cases.

Table 13. Comparison of $PM_{2.5}$ emission factors and uncertainties for domestic wood burning technologies in the EMEP/EEA inventory guidebook.

Fuel	Technology	$PM_{2.5}$ (solid particles only)		$PM_{2.5}$ (Total PM)		Reference
		EF	95% CI	EF	95% CI	
Wood	Open fireplace	240	120-480	880	440-1760	Alves et al. (2011)
Wood & similar wood waste	Conventional stove	140	70-280	740	370-1480	Alves et al. (2011) and Glasius et al. (2005)
Wood	High-efficiency stoves	140	70-280	370	285-740	Glasius et al. (2005)
Wood	Advanced/ ecolabelled stoves & boilers <50 kW_{th}	47	10-110	93	19-233	Johansson et al. (2003), Goncalves et al. (2010), Schmidl et al. (2011)
Wood & similar wood waste	Conventional boilers < 50 kW_{th}	140	70-280	470	235-940	Winther (2008) and Johansson et al. (2003)
Wood pellets	Pellet stoves and boilers <50 kW_{th}	30	15-60	60	30-120	Denier van der Gon et al. (2015)

The Guidebook also stresses that such country specific differences are not accounted for: "*The split between conventional, high-efficiency and ecolabelled stoves should be made based on country specific information. The Guidebook does not contain specific information on a country basis for this split.*" P.83

In January 2017, Ricardo published a report '*Assessment of particulate emissions from wood log and wood pellet heating appliances*' (Stewart, 2017). The authors recommended that the emissions factors in the EMEP/EEA Guidebook be reviewed and that the NAEI should consider the implications of emissions variability with regard to uncertainty in sector 1A4bi. In addition, there is a need for better standardisation of emissions testing of domestic burning technologies and the real-world applicability of laboratory-derived emissions factors should be examined. Results showed average total PM emissions at nominal output to range from 65 g/GJ to 192 g/GJ for stoves tested with an electrostatic precipitator (UK method BS 3841 / BS PD 6434). For other methods tested, the range was 36 to 108 g/GJ with a dilution tunnel (Norwegian method NS3058) and 3.3 to 16.2 with a heated filter (German/Austrian DINplus method). These values are significantly lower than the average used in the NAEI.

There is a strong body of evidence which demonstrates lower emissions factors for modern high-efficiency domestic burning appliances. This is usually where a traditional open fire or manual stove is compared to an automated pellet stove or chip boiler. To date there is limited evidence of the emissions of Ecodesign-ready appliances. However, Tschamber et al. (2016) tested two high-end wood burning stoves which

meet the criteria for Ecodesign and the French Flamme Verte ecolabel. PM emissions factors were reported as 0.02 g/kg (1.1 g/GJ) for the WABI stove and 0.23 g/kg (12.8 g/GJ) for the XP54-IN stove. Ozgen et al. (2014) reported PM emissions factors of 512 g/GJ (434-611 g/GJ) for an open fireplace, 143 g/GJ (120-176 g/GJ) for an advanced stove and 109 g/GJ (75-139 g/GJ) for a pellet stove.

3.4.3 The ability of Ecodesign to reduce emissions and recommendations to achieve further reductions

Regulation 2015/1185 to the Ecodesign Directive setting requirements for solid fuel local space heaters is expected to result in emissions savings and efficiency improvements through the use of modern appliances. The projected impacts of this across the EU are shown in Table 14 relative to 2010.

Table 14. Projected annual emissions savings across the EU through the implementation of Regulation 2015/1185 to the Ecodesign Directive.

Parameter	Annual saving by 2030
Energy saving	41 PJ (0.9 Mtoe)
CO_2 saving	400 kilotonnes
Particulate matter (PM)	27 kilotonnes
Organic gaseous compounds (OGCs)	5 kilotonnes
Carbon monoxide (CO)	399 kilotonnes

Similar proportionate benefits are likely to be achieved in the UK. From the above it is clear that appliances which meet the Ecodesign limits can achieve large reductions in emissions in comparison with open fireplaces and older more traditional stoves. Independent laboratory testing by Kiwa Gastec demonstrates that on average, Ecodesign-ready appliances emit 90% less particulate matter than an open fire and 80% less PM than a traditional 10 year old stove[3]. This is due to major improvements in the design and technology of Ecodesign-ready appliances, which increase combustion efficiency through careful control of the time-temperature history of gases as the fuel decomposes. As described elsewhere in this report, improvements in combustion efficiency not only reduce the mass of particles emitted but also the relative toxicity of those particles. Co-benefits also include a reduction in the products of incomplete combustion such as CO, NMVOCs, PAH, benzene, levoglucosan, organic carbon and tars. This has a profound effect on the chemical signature of emissions, as well as the relative proportion of tracers which are used in source apportionment. Other co-benefits of efficiency improvements include a reduction in the

[3] Laboratory test results available to view at http://www.stoveindustryalliance.com/ecodesign-ready-stoves-and-air-quality/

amount of fuel required to meet the heat demand of properties, thereby reducing total activity data.

The recent User Survey (SIA, 2019) found that 27.4% of appliances are open fires or stoves older than 10 years. Replacing these with Ecodesign-ready stoves could achieve PM emissions reductions of 45% for the domestic burning sector.

At the time of writing, Ecodesign will be implemented in just over 2 years' time. Appliance manufacturers have already made significant progress in reducing emissions and now market a large number of appliances under the 'Ecodesign-ready' label. However, several manufacturers now offer appliances which can achieve emissions significantly lower than the Ecodesign requirements (see for example Tschamber et al., 2016) but at present, Ecodesign remains the lowest standard available. Going forward beyond Ecodesign to 2030, both the Government and industry may wish to consider how best to encourage manufacturers to produce cleaner and more efficient appliances. Advice may be gleaned from the transport sector, whereby ultra-low emission vehicles (ULEVs) are encouraged and support. A similar scheme incentivising ultra-low emissions stoves (ULESs) may be appropriate post-Ecodesign, encouraging manufacturers to continue to innovate and reduce emissions through design and abatement technologies.

3.5 Section 3 summary

Activity data

- Within the National Atmospheric Emissions Inventory (NAEI), domestic burning is dominated by natural gas, heating oil and wood fuels. The amount of wood burned is said to have increased from 1.8 million tonnes in 2005 to 4.8 million tonnes in 2017.
- This number is derived from the Government's *Domestic Wood Use Survey* which found that previous figures for the amount of wood consumed was underestimated by a factor of three based on a sample of 1,206 wood fuel users. Applying a seasonal regression analysis to this data would mean that 1.06 million tonnes of wood is burned in the peak heating months, equating to 16.7 kg per household per day. This is significantly higher than use in other countries.
- Data from HMRC shows that 93 thousand tonnes of wood fuel were imported and 73 thousand tonnes were exported in 2018 though this does not include UK-grown and used wood.
- Data from an industry survey of 10,620 wood fuel users revealed that wood fuel is used for heating for 4.7 months of the year, with an estimated total of 1.87

million tonnes of wood burned per year. This is up to a factor of 2.4 (61%) lower than NAEI estimates.

Emissions contributions

- According to the NAEI, total PM_{10} emissions from domestic burning were 45.1 kilotonnes in 2017, of which 43.6 kilotonnes were from solid fuels and 38.8 kilotonnes were from wood burning.
- NAEI data shows that domestic wood burning contributed to 23.1% of national PM_{10} emissions and 35.8% of national $PM_{2.5}$ emissions in 2017 but given the reduction in activity data above this is expected to reduce to 9.6% of PM_{10} and 14.9% of $PM_{2.5}$ respectively.

Emissions factors

- Emissions factors of $PM_{2.5}$, NOx and CO for domestic wood burning in the NAEI are 443 g/GJ, 63 g/GJ and 2,856 g/GJ respectively. This $PM_{2.5}$ emissions factor is 3 times higher than the limit under the Ecodesign regulations. Replacing open fires and older stoves could therefore have a significant impact.
- Evidence is presented from the literature demonstrating that appliances can and do meet the emissions limits for Ecodesign and therefore it is possible that emissions factors are overestimated in the NAEI.
- Further evidence is required for the appropriate average emissions factors to be used in the NAEI which represents the current mix of appliance types and ages

4. UK domestic burning emissions in the international context

4.1 UK emissions obligations under European legislation

Both the United Kingdom and European Union are required to report total annual emissions of key air pollutants under the Gothenburg Protocol to the Long-range Transboundary Air Pollution (LRTAP) Convention of the United Nations Economic Commission for Europe (UNECE), as well as the National Emissions Ceilings (NEC) Directive (2016/2284/EU).

The revised Gothenburg Protocol was ratified by the European Union in August 2017 and places an obligation on member states to reduce national emissions by a specified amount by 2020 relative to a 2005 baseline, as shown in Table 15 (UNECE, 2012). Additional targets for 2030 apply under the revised NEC Directive (EU) 2016/2284 which came into force in December 2016.

Table 15. UK and EU air pollutant emission reduction targets under the Gothenburg Protocol and National Emissions Ceilings Directive 2016/2284/EU.

Pollutant	Reduction on 2005 levels			
	EU-28		UK	
	2020-2029	From 2030	2020-2029	From 2030
SO_2	-59%	-79%	-59%	-88%
NOx	-42%	-63%	-55%	-73%
NH_3	-6%	-19%	-8%	-16%
NMVOC	-28%	-40%	-32%	-39%
$PM_{2.5}$	-22%	-49%	-30%	-46%

Regarding sector 1A4bi (residential stationary combustion), the following is an excerpt from the 2012 amendment to the Gothenburg Protocol Annex X, section A, paragraph 16 concerning particulate matter emission limit values (UNECE, 2012):

"*This paragraph is recommendatory in character and describes the measures that can be taken insofar as a Party considers them to be technically and economically feasible for the control of particulate matter*:

a) Residential combustion installations with a rated thermal input < 500 kWth:

i. Emissions from new residential combustion stoves and boilers with a rated thermal input < 500 kWth can be reduced by the application of:

A. Product standards as described in CEN standards (e.g., EN303–5) and equivalent product standards in the United States and Canada.

Countries applying such product standards may define additional national requirements taking into account, in particular, the contribution of emissions of condensable organic compounds to the formation of ambient PM; or

B. Ecolabels specifying performance criteria that are typically stricter than the minimum efficiency requirements of the EN product standards or national regulations.

Table 16. Recommended limit values for dust emissions released from new solid fuel combustion installations with a rated thermal input < 500 kWth to be used with product standards:

	Dust (mg/m^3)
Open/closed fireplaces and stoves using wood	75
Log wood boilers (with heat storage tank)	40
Pellet stoves and boilers	50
Stoves and boilers using other solid fuels than wood	50
Automatic combustion installations	50

Note: O_2 reference content: 13%

ii. Emissions from existing residential combustion stoves and boilers can be reduced by the following primary measures:

A. Public information and awareness-raising programmes regarding:
- *The proper operation of stoves and boilers;*
- *The use of untreated wood only;*
- *The correct seasoning of wood for moisture content.*

B. Establishing a programme to promote the replacement of the oldest existing boilers and stoves by modern appliances; or

C. Establishing an obligation to exchange or retrofit old appliances."

Note that the recommended emission limit values are less stringent that the limits under Ecodesign, which are 50 mg/m^3 for open fireplaces, 40 mg/m^3 for log/multifuel stoves and 20 mg/m^3 for pellet stoves at 13% O_2. The UK Government has taken the steps recommended in part iiA above under the Clean Air Act, but has not implemented parts B or C on the replacement of older appliances with modern low-emission equivalents (aside from adopting Ecodesign limits for new appliances from 2022). Literature from the European Commission (Wolters, 2018) shows that the EU is committed to reducing emissions from domestic burning through the implementation of Ecodesign and Energy Labelling regulations. The role of national authorities in market surveillance and enforcement is stressed, with the help of the European Eco-

design Compliance Project (ECOPLIANT) for inspection of labelling, product testing and cross-border cooperation. The Commission gives the following advice for the mitigation of emissions from residential wood combustion:

1) Reduce the use of non-regulated stoves
2) Reduce combustion under non-optimal conditions
3) Good maintenance of old and new stoves
4) The use of standardised biomass

4.2 Domestic burning source contributions in the European Union

Data from the most recent EU Inventory Report (EEA, 2019a) shows that in 2017, the UK had reduced its $PM_{2.5}$ emissions by 15.2% relative to 2005 while the EU had achieved a reduction of 21.8%. Conversely, the EU inventory report shows UK $PM_{2.5}$ emissions from sector 1A4bi (residential stationary combustion) to have increased by 54% relative to 2005. Since the UK is unlikely to meet its 2020 target for $PM_{2.5}$, the Government should consider appropriate actions to mitigate emissions from domestic burning. However, if as evidence suggests the true activity data is a factor of 2.4 lower than that stated in the current inventory, the UK may in fact be closer to achieving its 2020 target.

Across the whole of the EU-28, the Inventory Report (EEA, 2019a) found that $PM_{2.5}$ emissions from most sectors have reduced considerably since 2000; by 69% for public electricity and heat production (1A1a) and by 57% for passenger cars. Category 1A4bi (residential stationary) was found to be the largest contributor to $PM_{2.5}$ emissions across Europe at 51% of the total and total emissions from this category were reported to have changed little since 2005. The contribution of domestic burning to PM_{10} was found to be much lower than for $PM_{2.5}$ at 39% of the total, owing to the difference in size fractions between particles emitted from different sources. The report also found that category 1A4b contributes to 48% of European CO emissions, 26% of PCDD/F emissions and 36% of PAH emissions. The highest emissions in this sector were reported in Italy, Romania, France and Poland, whereas the greatest emissions reductions since 2000 have been achieved in Germany owing to a move away from solid fuels such as lignite and towards gaseous fuels.

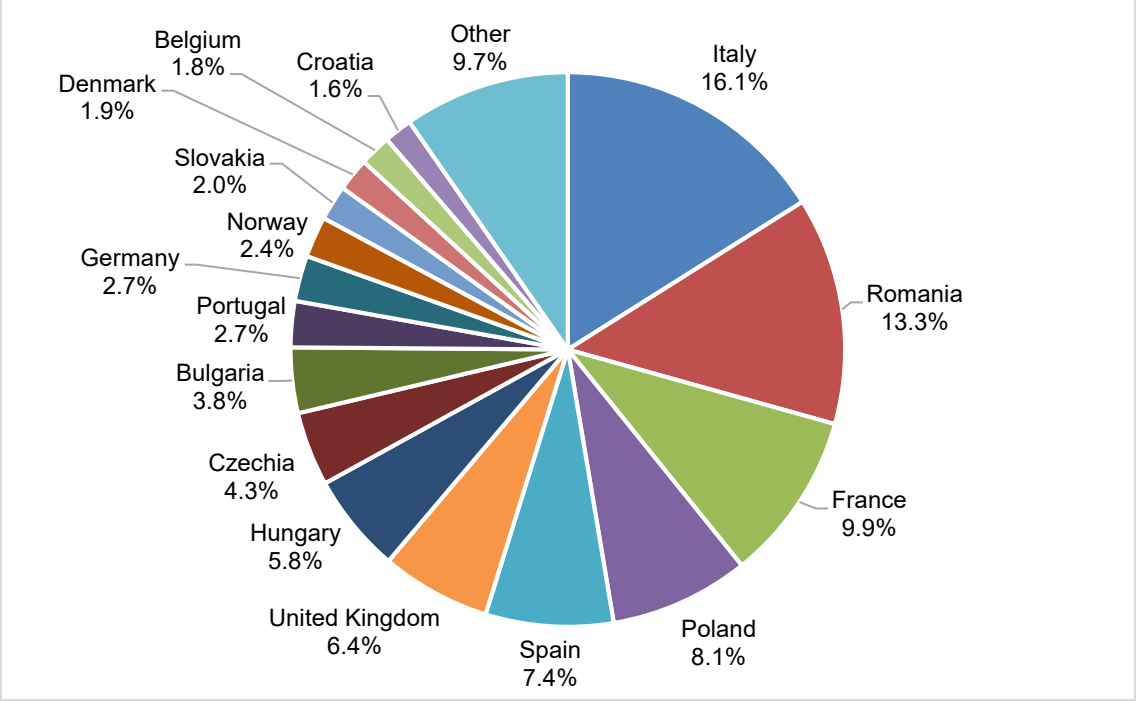


Figure 26. National contributions to total European $PM_{2.5}$ emissions from sector 1A4bi (residential stationary combustion) in 2017. Data source: EEA (2019a).

UK domestic burning contributes 6.4% of total European domestic burning $PM_{2.5}$ emissions (Figure 26). In comparison to many European nations, the contribution of domestic burning to total national $PM_{2.5}$ is relatively low, as shown in Figure 27.

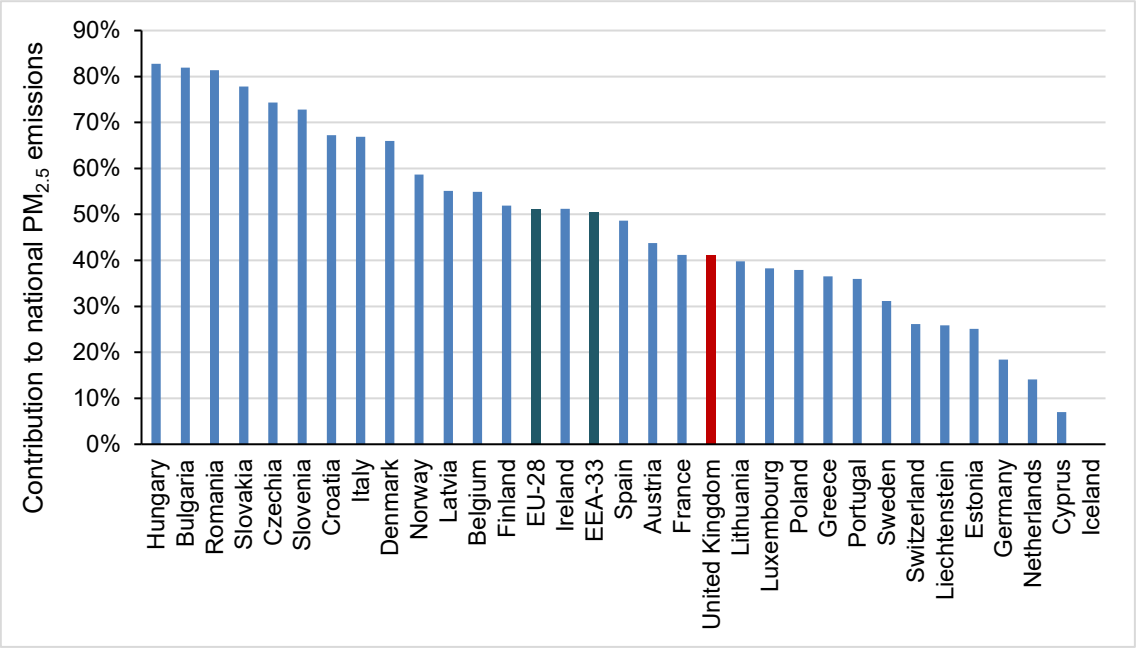


Figure 27. Contribution of domestic burning (1A4bi) to national $PM_{2.5}$ emissions in 2017 for European nations. Data source: EEA (2019a).

4.3 Progress towards emissions reduction obligations

The latest data from the European Environment Agency (EEA, 2019b) shows that across Europe, member states have already met 2020 emissions reduction commitments for SO_2 and NMVOCs. However in order to meet 2020 targets for other pollutants, emissions reductions of 3.2% in NOx, 2.3% in NH_3 and 1.3% in $PM_{2.5}$ must be achieved across Europe compared with 2017. In order to meet the more stringent 2030 targets, reductions of 33% in SO_2, 36% in $PM_{2.5}$ and 40% in NOx are required based on 2017 emissions.

When a Member State is not on track to meet its emission reduction commitments, it is required to report projections complemented 'with additional measures' (WaM) — scenarios that reflect adopted and planned measures contributing to a further reduction of emissions. Sixteen Member States reported WaM projections for the five main pollutants for both 2020 and 2030, but the UK did not, even though projections based on current measures indicate that **compliance will not be attained for all pollutants**. The UK progress towards attaining 2020 and 2030 $PM_{2.5}$ levels is summarised in Table 17.

Table 17. $PM_{2.5}$ emissions projections for the UK to 2030 and the contribution of the commercial, institutional and households sector 1A4 to total emissions. Data source: EEA (2019a,b)

	Base year	Latest Inventory year	Projections (WM)**		
	2005	2017	2020	2025	2030
Total UK $PM_{2.5}$ emissions (kilotonnes)	126.02	106.81	98.21	94.58	93.20
Percent reduction on base year	0%	-15.2%	-22.1%	-24.9%	-26.0%
Total UK $PM_{2.5}$ emissions from Commercial, institutional and households sector (1A4)* (kilotonnes)	35.46	46.15	44.35	43.60	43.43
Percent contribution of sector 1A4 to national $PM_{2.5}$ emissions	28.1%	43.2%	45.2%	46.1%	46.6%

*Note that projections refer to aggregate sector 1A4 which includes domestic burning (1A4bi) as well as commercial and institutional sources.
** The 'with measures' (WM) projection is calculated by Member States on the basis of adopted policies and measures currently in place, which are used to assess attainment of 2020 and 2030 reduction commitments.

Projections currently show that the UK is not on course to meet its legally binding target for a 46% reduction in primary $PM_{2.5}$ emissions from 2030 under the NEC Directive, with current estimates at just 26%. Total $PM_{2.5}$ emissions from the

commercial, institutional and households sector (1A4) are projected to reduce by less than 6% by 2030 relative to 2017. Due to emissions reductions in other sectors, the contribution of sector 1A4 to total UK $PM_{2.5}$ emissions is projected to increase from 43.2% in 2017 to 46.6% in 2030.

4.4 UK domestic burning & environmental policy post-Brexit

Environmental policy in the UK is underpinned by a range of European regulations and directives. Since domestic burning is seen to be a major contributor to air pollution across Europe, policies put in place to improve air quality in member states have a direct impact on UK domestic burning and it is imperative to understand if and how this may change when the UK leaves the European Union.

The Government stated in its 25 Year Environment Plan that the commitment to achieving the legally binding targets to cut emissions under the LRTAP and NEC Directive and will not be affected by the UK's departure from the European Union. The Government also reaffirmed its ambition to work with local authorities and others to address emissions from domestic burning through campaigns such as Ready to Burn, as well as to implement the Clean Air Strategy.

DEFRA published its Draft Environment (Principles and Governance) Bill in December 2018, with a view to introducing a full Environment Bill in 2019. The purpose of the Bill is to ensure environmental principles and governance are protected when the UK leaves the EU, including specific targets for improving air quality. This will be overseen by the newly established Office for Environmental Protection (OEP). In July 2019, the Government issued a summer policy statement on the Bill and reaffirmed its commitment to improving air quality by legislating on key measures outlined in the Clean Air Strategy, including enabling local authorities to take a greater role in tackling emissions from domestic burning. However the details of Government strategy in this area are unclear at the present time.

Under the NEC Directive (which has already been transposed into UK law), member states are required to submit National Air Pollution Control Programmes (NAPCP) which set out measures and technical analysis to demonstrate how the 2020 and 2030 emission reduction commitments for NOx, SO_2, NH_3, NMVOCs and $PM_{2.5}$ can be met. The UK submitted its NAPCP in March 2019 (DEFRA, 2019e). The programme points to existing air quality policy and legislation, such as the Clean Air Act, and the efforts of the devolved administrations to produce their own strategies to reduce air pollution. The UK NAPCP refers directly to domestic combustion, reiterating the 38% contribution and stating that "*proposals for tackling emissions from this source in England are outlined in the Clean Air Strategy*". It goes on to describe the efforts of the Welsh Government in this area; developing an evidence base on domestic burning with relevant stakeholders and the potential impact of introducing future product standards, as well as launching a public engagement campaign to encourage behavioural changes through communications.

4.5 Achieving WHO guideline concentrations of $PM_{2.5}$

Alongside the summer policy statement, DEFRA released a report on achieving World Health Organisation (WHO) guideline levels of $PM_{2.5}$ (DEFRA, 2019d).

The report, produced by Imperial College London, modelled a range of scenarios to explore the potential routes to reducing $PM_{2.5}$ concentrations in line with the WHO guideline annual mean concentration of 10 μg/m^3. A previous report by the same authors identified the growth in wood burning to be a major contributor to the UK's $PM_{2.5}$ emissions, particularly from wet and improperly cured wood. Following this previous report, domestic wood and coal burning emissions were remodelled to include buoyancy plume rise effects from chimneys on dispersion, as opposed to gas and oil emissions which are treated as a volume source from building vents. This is a significant change in methodology, since plumes are now assumed to disperse downwind of the chimney and are clear of urban drag effects of local buildings. It should be noted that the most polluting domestic burning sources such as chimeneas, firepits, barbecues and garden incinerators do not benefit from the chimney buoyancy effects and therefore emissions are less dispersed than appliances fitted with flues.

The impact of the change in modelling methodology was a reduction in the local $PM_{2.5}$ concentrations from domestic burning by a factor of 2, which were found to be more consistent with observations from King's College researchers using aethalometers and levoglucosan (Font and Fuller, 2017). The authors found that the domestic burning contribution to winter $PM_{2.5}$ concentrations was 0.2-2.7 μg/m^3 and almost entirely absent in summer. Removing domestic wood burning from the model reduces the population exposed to concentrations above the WHO recommended 10 μg/m^3 from 15 million to 9.2 million people.

Based on these findings and assuming a linear reduction in concentrations in line with the reduction in activity data in section 3.1.2, domestic burning is estimated to contribute 0.08-1.1 μg/m^3 towards total $PM_{2.5}$ concentrations. As a result, the population exposed to $PM_{2.5}$ >10 μg/m^3 is likely to reduce to 11.5 million people. Revisions to domestic burning activity data and a support programme for the replacement of older stoves and open fires with modern Ecodesign stoves is likely to help the Government to meet the WHO Guideline for $PM_{2.5}$. DEFRA stated in its summer policy announcement that "*whilst challenging, it would be technically feasible to meet the World Health Organization guideline level for $PM_{2.5}$ – the most harmful air pollutant to public health - across the UK*".

4.6 Section 4 summary

- The UK and other EU member states are required to reduce emissions of SO_2, NOx, NH_3, NMVOC and $PM_{2.5}$ by 2020 and 2030 under the Gothenburg Protocol and the NEC Directive.

- Across the whole of the EU, 2020 targets have already been met for SO_2 and NMVOCs but not for other pollutants.
- Major efforts are required for all member states in order to meet the more stringent 2030 targets (Reductions of 33% for SO_2, 36% in $PM_{2.5}$ and 40% in NOx compared to 2017 emissions).
- For the UK, projections based on current measures indicate that **compliance will not be attained for all pollutants**.
- The contribution of sector 1A4 (commercial, institutional and households) to national $PM_{2.5}$ emissions is projected to increase from 43.2% in 2017 to 46.6% in 2030 based on existing inventory data. Hence this sector could be crucial in achieving future $PM_{2.5}$ targets, which could be done cost-effectively in comparison to other sectors since any changes are likely to have a large impact on national emissions.
- Revisions to UK domestic burning activity data in line with the industry survey could mean that the UK is more likely to meet its $PM_{2.5}$ reduction targets under the revised Gothenburg Protocol and the NEC Directive.
- The UK National Air Pollution Control Programme (NAPCP) outlines how the Government plans to implement air pollution reduction policies in line with its obligations. For domestic burning, the NAPCP refers to the Clean Air Strategy but does not present any further strategies specific to this important sector.
- Despite this, the Government stated that it would be technically feasible for the UK to meet WHO Guideline limit concentrations for $PM_{2.5}$ (10 $\mu g/m^3$), based on modelling work from Imperial College London.
- Achieving the WHO Guideline is likely to be challenging, requiring emissions reductions across multiple sectors including domestic burning. However, given the uncertainties in activity data and in source apportionment, it is likely that the contribution of domestic stoves and fireplaces to $PM_{2.5}$ concentrations is lower than current estimates.

5. Other solid fuel combustion sources

Particulate emissions are known to have a multitude of sources, including a significant amount from biomass combustion processes both within the domestic sector and outside, as shown in Table 18. This section will address these alternate PM sources and their scale to better understand how they may contribute to overall emissions and how their levels of uncertainty can conflict in the source apportionment of domestic burning sources.

Table 18. Comparison of NAEI PM_{10} and $PM_{2.5}$ emission factors for various biomass combustion emission sources

Source	NRF Code	Fuel	PM_{10} EF (kt)	$PM_{2.5}$ EF (kt)	Activity Units
Domestic combustion	Residential: stationary	Wood	7.4	7.2	Mt fuel consumed
Other industrial combustion	Stationary combustion in manufacturing industries and construction	Wood	2.7	2.7	Mt fuel consumed
Small-scale waste burning	Open burning of waste	Waste	0.014	0.013	Kilotonne
Bonfire night	Other waste	Mass burnt	12	11	Megatonne
Accidental fires – forests	Forest fires	Area burnt	0.00032	0.00027	hectares
Accidental fires – vegetation	Other natural emissions	Area burnt	0.000022	0.000018	hectares
Accidental fires – straw	Other natural emissions	Mass burnt	0.011	0.0090	Kilotonne

5.1 Other emission sources within the domestic burning sector

There are many other sources of air pollution within the domestic burning sector, particularly those which burn wood or biomass. Many of these appliances are unregulated and unabated, whereas wood burning stoves must meet Smoke Control Area limits and Ecodesign limits from 2022. The type of appliance in which wood or biomass is burned has a major effect on the emissions produced per kilogram of fuel burned and assuming just one average emission factor for the appliance population may result in significant errors.

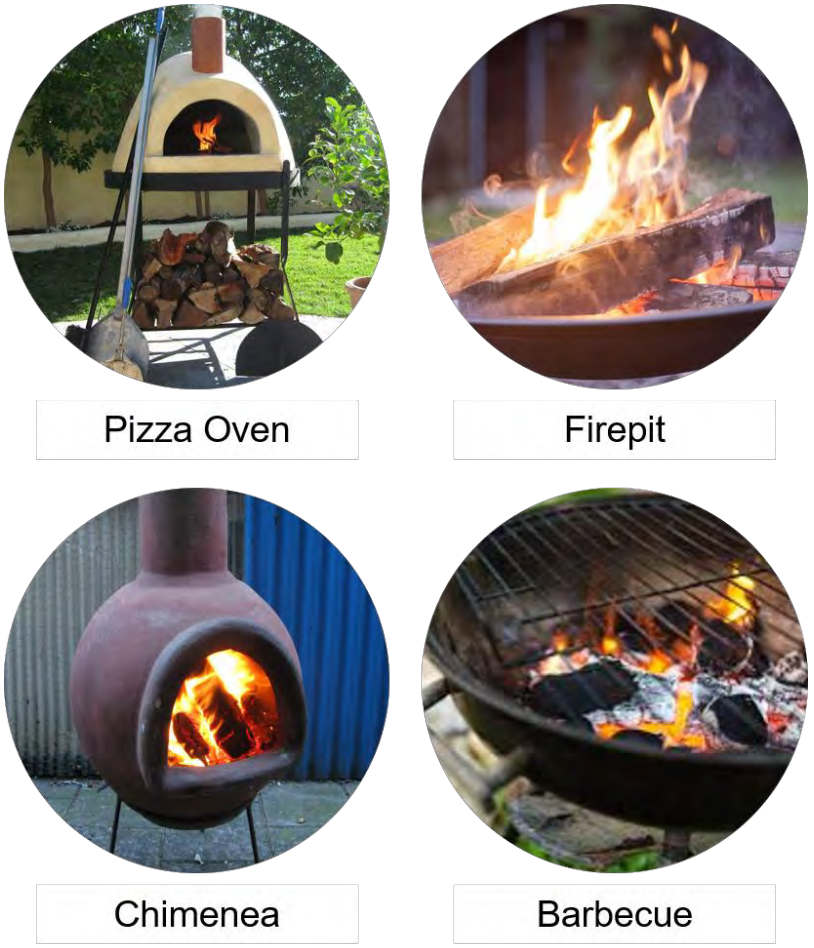


Figure 28. Examples of non-stove solid fuel domestic burning appliances

5.1.1 Domestic barbecues

One key area of air quality and human health research relates to cooking fumes, which are known to contain substances such as $PM_{2.5}$, black carbon, heterocyclic amines and polycyclic aromatic hydrocarbons (PAHs). The majority of existing, historic studies have focused on indoor cooking, however, the risks associated with outdoor cooking requires greater attention. Eating outdoors and the embrace of street food within western society is becoming an increasingly prevalent and important part of our diets. As suggested by Wu et al. (2015), the fumes associated with both domestic and commercial barbecuing may have become a significant, but neglected, contributor to poor air quality.

In addition to the establishment of commercial street food enterprises, domestic barbecuing is becoming an increasingly popular pastime in the UK, especially from early spring onwards as the weather improves. Indeed, as shown in Figure 29, during the last 5 years the net imports of charcoal to the UK have continued to increase, reaching a total of ~86,000 tonnes in 2018 (HMRC, 2019). Furthermore, in the first quarter of 2019, net imports of charcoal have continued at the same rate as the previous year, indicating that this trend looks set to continue. The majority of the

charcoal imported into the UK is sourced from Namibia and Paraguay, produced from savannah shrubs and coppiced hardwoods, respectively.

Although outdoor cooking can be weather dependent, increased occurrences of warmer weather in the UK may explain the growing demand for charcoal as a domestic cooking fuel. This is further supported by the imports of barbecues; since 2015 there has been a net import total of 9.3 million units into the UK, with the majority of these sourced from China (HMRC, 2019). The increasing numbers of barbecues – in addition to the growing use of imported charcoal within the UK – demonstrates the importance of properly understanding and quantifying their impact on air quality.

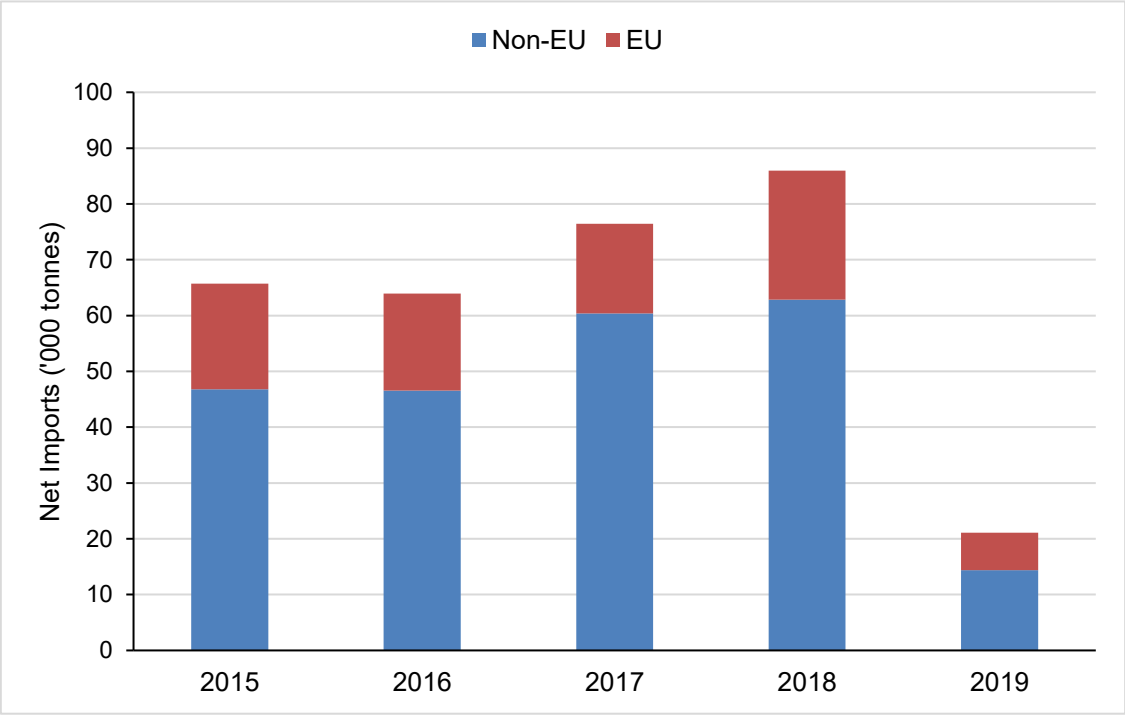


Figure 29. Net imports of charcoal to the UK (HMRC, 2019)

Indeed, this is a growing area of interest, with an increasing number of researchers concerned over the emissions barbecues produce. Wu et al. (2015) demonstrated that the PM_{10} and $PM_{2.5}$ emissions of a charcoal burning barbecue measured at 900-5,300 and 700-4,100 $\mu g.m^{-3}$, respectively, within a 2 metre radius.

At greater distances, the measurements show a decrease in PM_{10} and $PM_{2.5}$ concentrations, although these are still noticeable higher than expected ambient conditions. These were measured at 37-150 and 20-75 $\mu g\ m^{-3}$, respectively. The increased concentrations of PM, in relation to ambient urban air, is supported further by Song et al (2018), which reported $PM_{2.5}$ mass concentrations of 250-1083 $\mu g\ m^{-3}$ within 1m of the barbecue. However, the study also suggested that in addition to the high concentrations of particulate matter within the immediate proximity of the barbecue grill, the concentrations can peak at a distance between 10-15m from the

cooking unit. This is potentially due to a smoke transport mechanism, involving an initial lift from the grill followed by the dispersion of the PM, before dropping back towards the ground. Vicente et al, (2018) conducted a study, aimed at experimentally quantifying and characterising the particulate and gaseous emissions from charcoal combustion when using a barbecue grill. As a result, the study reported a $PM_{2.5}$ emission factor of 7380 ±353 mg kg^{-1}, however it also highlighted the significant discrepancies that currently exist of the emissions reported within the existing literature.

In addition to barbecues, which are used for cooking, there are a wide range of alternative steel- and clay-based appliances that utilise simple open combustion systems for both heating and cooking. These include chimeneas and firepits which are marketed as multi-fuel patio heaters, predominantly fuelled by wood. Unlike existing appliances that are designed specifically for heating, chimeneas and firepits tend to focus on the aesthetics of wood burning, often resulting in poor combustion conditions and lots of smoke. There is very little published literature on the particulate matter emissions of solid fuel patio heaters, however when considering their basic open combustion chamber design, they are comparable to bonfires and other open burning systems (see Section 4.1.3). In recognition of this, there are now several 'smokeless' firepits on the market, containing simple secondary combustion systems, however these only reduce the amounts of visible smoke and have not undergone any formal testing against regulations such as those required for European Ecodesign stoves.

5.1.2 Commercial cooking

The relationships between particulate air pollution and commercial cooking activities has continued to grow in interest, specifically the impact on the associated microenvironments. Of these, restaurants that utilise wood burning grills and ovens, such as traditional pizzerias, are of particular importance when considering particulate emissions within populated areas.

Cooking with a wood-fired pizza oven is a time-consuming process as once the fire is established, it ranges between 1-2 hours before the correct cooking temperature is reached. Consequently, in commercial pizzerias, once the oven is lit the fire will be maintained continually throughout the restaurants opening hours. The wood consumption of wood-fired pizza ovens will depend on its size and its operating times. For example, an oven with a 105 cm diameter, capable of cooking 3-5 pizzas at once, will consume ~40 kg of hardwood during a single firing. Indeed, an accepted industrial benchmark for wood consumption is at a rate of ~7 kg/hr (Forno Bravo, 2019).

The study by Buonanno et al. (2010) investigated indoor particulate matter concentrations within 15 traditional Italian pizzerias that utilised wood-fired pizza ovens. The PM_{10} and $PM_{2.5}$ concentrations understandably differed between restaurants due to different sized ovens and different extraction equipment, with measured concentrations ranging from 15-482 $\mu g.m^{-3}$ and 12-368 $\mu g.m^{-3}$ respectively. As a result, PM_{10} had a calculated mean concentration of 123 $\mu g.m^{-3}$, while $PM_{2.5}$ was

slightly smaller, calculated at 95 μg.m^{-3}. This clearly demonstrates that commercial food cooking units produce large quantities of particulate matter. The PM measurements stated within this study were concerned with indoor emissions, with the actual amounts produced and extracted outside most likely to be much greater.

In the UK, restaurants that are located within smoke control areas, which utilise ovens and grills burning unauthorised fuels such as wood, must use appliances that are exempt and feature on DEFRA's published exemption list. For an appliance, such as a wood-fired oven, to be considered for exemption it must first demonstrate that its emissions fall within the acceptable limits, utilising approved testing methods by an accredited test centre. For appliances with an output of ≤44kW, the emission limit is calculated as 5 g/h + 0.1 g/0.3 kW output of the appliance. For example, a wood-fired pizza oven with an output of 10 kW has a permitted smoke emission limit of 8.3 g/h to qualify for exemption (HETAS, 2018). The ability to successfully build, light and maintain a wood fire over a prolonged period is dependent on a number of important factors, particularly those relating to the individual user. Although an appliance meets certain criteria within a laboratory test environment, the devices use under real world conditions adds an increased level of variability which is difficult to effectively quantify. However, it is clear that even under ideal conditions, these types of cooking unit produce significant emissions which should be individually accounted for.

5.1.3 Open burning of waste/bonfires

A simple and effective way of disposing of waste plant residues is via combustion, with the open burning of biomass often practiced in domestic settings. There are no specific by-laws prohibiting garden bonfires, however there are existing environmental laws that relate to the health issues associated with smoke, or that affect people's enjoyment of their property (UKELA, 2017). The legality of allotment bonfires usually depends on their location and the tenancy agreement with the landowner; different councils will have different by-laws that should be adhered to. Although there are good practice guides on bonfires giving advice on weather conditions and what should and shouldn't be burned – such as those published by the National Allotment Society – there is no active enforcement of this, meaning that the quality of the burn is directly dependent upon the user (NSALG, 2019). At their peak, allotment plots within the UK totalled approximately 1.5 million however current estimates indicate there are ~330,000 plots in use.

Open-air burning of wastes – usually following horticultural activities such as pruning and scarification – is a common practice, predominantly as a quick and inexpensive option for managing vegetative debris and residues. The poor combustion conditions and the quality of the feedstocks can however have detrimental impacts on local air quality. This has been evidenced in Alves et al. (2019) which detailed the emission factors of different biomass residue feedstocks from open burning. Pruned woody branches have calculated particulate matter EFs of 8.76 g/kg to 16.9 g/kg, dependent upon the species. Holder et al. (2017) analysed the emissions produced from the open

burning of grass, stating particulate matter emissions of 4.2 g/kg to 11 g/kg depending on the combustion conditions. Gonçalves et al. (2011) determined that the open combustion of weeds and other 'green' vegetative offcuts produced between 0.9-1.34 g/kg of PM_{10}. The results of a study investigating the burning of leaves and leaf litter within an open chamber combustion system measured $PM_{2.5}$ emissions of 3.33-4.38 g/kg. In addition to the emission factors, the results suggest that larger quantities of feedstock and poor air flows, such as those found within large piles of raked leaves for example, can result in greater emissions of particulate matter (Chantara et al., 2019).

With more than 300,000 actively used allotment plots, in addition to an estimated 400,000 ha of private gardens contained within Great Britain (WLGF, 2014), the annual production and consequent disposal of vegetative residues is potentially substantial. For example, Wiedinmyer *et al.* (2014) estimated that within the UK, 398,131 tonnes of waste is burned by residents annually. This is calculated to lead to 4.74 kt of PM_{10} a year, equivalent to 4.7% of annual anthropogenic PM_{10} emissions.

Open burning is particularly prominent during the end of October and beginning of November as there are often a large number of public and private bonfire events related to bonfire night. Singh *et al.* (2015) reported decreased visibility on bonfire night compared to previous days, whilst Pope *et al.* (2016) found large peaks occur in monitored PM emissions around bonfire night, with these capable of exceeding the WHO 24h mean safe exposure limits. According to the NAEI estimates, 1.4 kt of PM_{10} and 1.2 kt of $PM_{2.5}$ are associated with bonfire night. These are calculated using emission factors of 12 kt/Mt mass burnt and 11 kt/Mt mass burnt respectively, as shown in Table 18.

5.2 Other non-domestic emission sources

5.2.1 Wildfires

Wildfires are a potentially significant source of biomass burning in the UK. As defined by the Forestry Commission England (2019), and The Scottish Government (2013) Fire and Rescue Service, a wildfire incident is "any uncontrolled vegetation fire which requires a decision, or action, regarding suppression." This includes both woodland (forest) fires, and non-woodland (i.e. arable land, grassland, moorlands etc.) fires. Their distinction from prescribed or agricultural burning covered in the subsequent sections is therefore their uncontrolled manner.

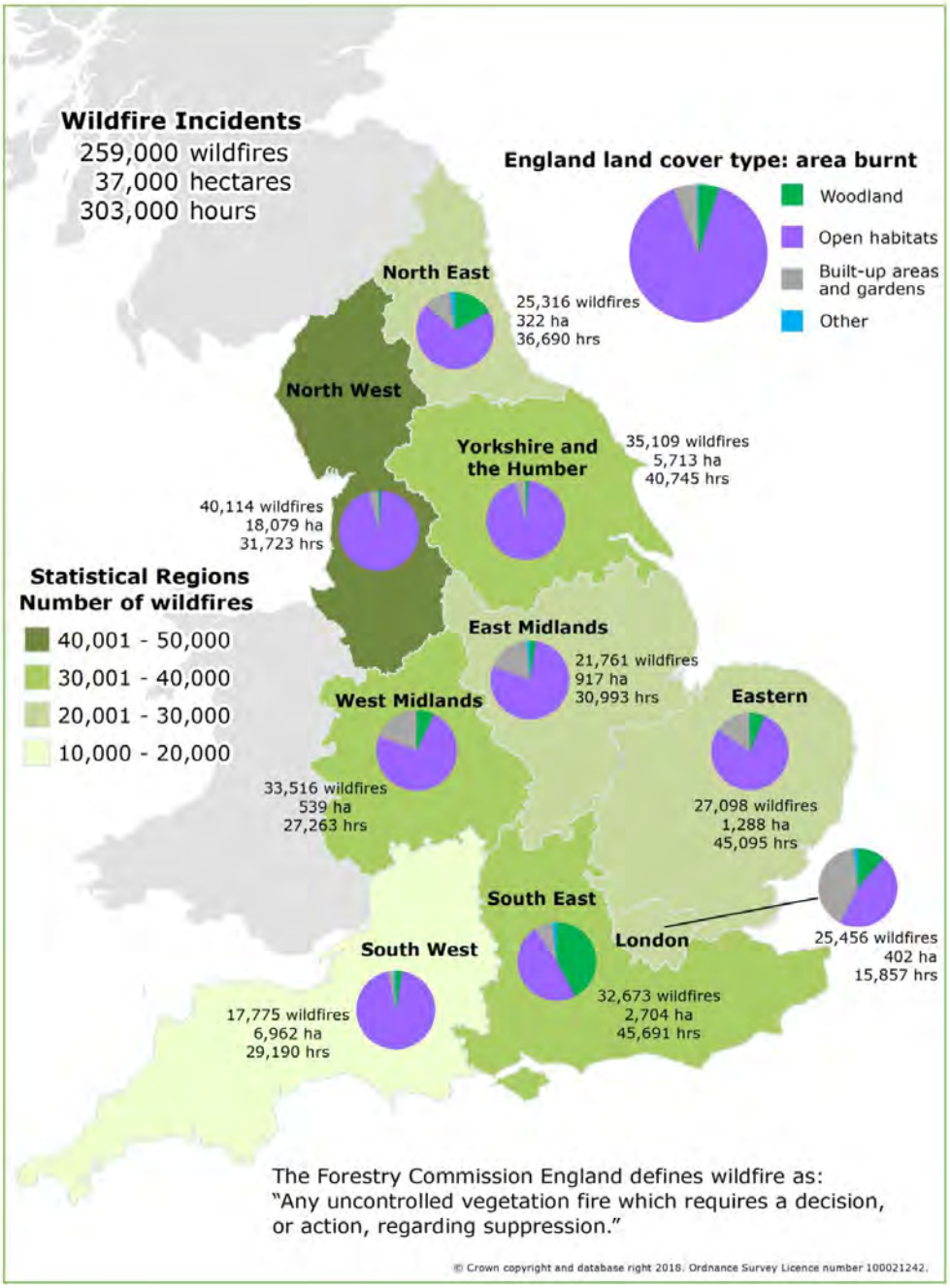


Figure 30. Wildfire incidents from 2009-10 to 2016-17 in England (Forestry Commission England, 2019).

Despite the UK not usually being associated with wildfires on the scale seen in other regions such as North America or Australia, they are nevertheless relatively common in the UK. For example, the Forestry Commission England, 2019 reports around 259,000 wildfire incidents over an 8 year period in England alone, burning 37,000 hectares in total, equivalent to around 4,600 hectares a year (Figure 30). Of this, the majority of burnt area was non-woodland land use, however, woodland fires with high fuel loading accounted for on average 215 hectares per year (~5%). Likewise, Scotland and Wales also reported a high number of wildfire incidents, with woodland

fires alone burning on average 1,500 ha and 381 ha a year respectively (Forestry Commission, 2018). As these typically account for a small proportion total wildfire incidents, particulrly in comparison to grassland wildfires, the total burnt area is actually significantly higher (Figure 31).

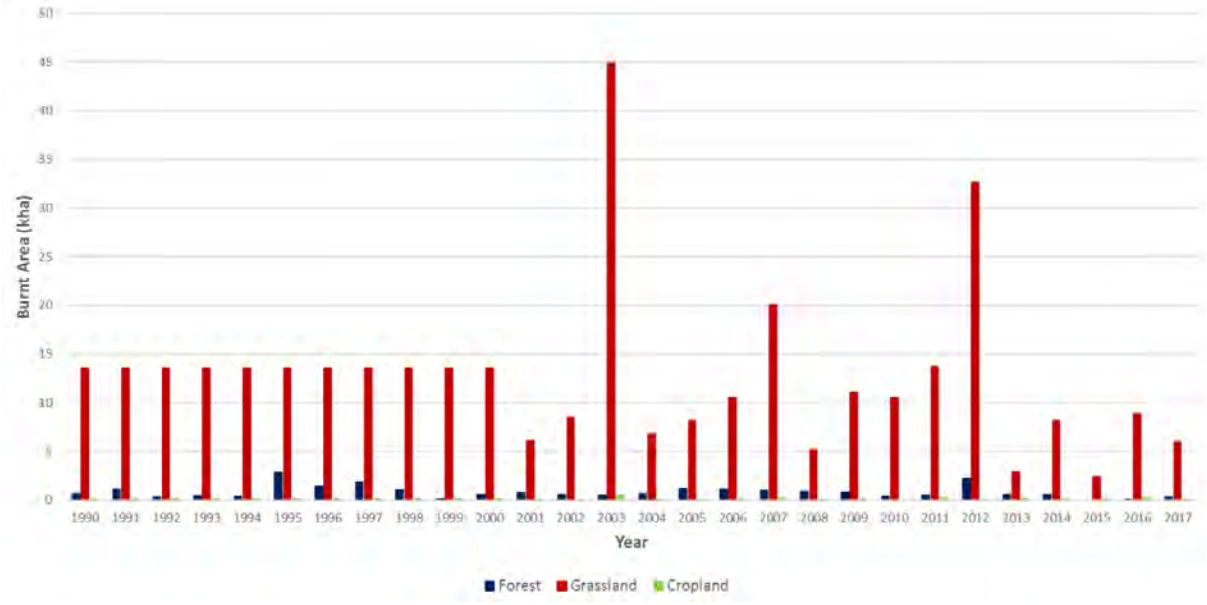


Figure 31. Time series of wildfire burnt area in the UK for 1990 to 2017 *(Brown* et al*., 2019).*

Research by Jollands *et al.* (2011) found the majority of wildfires in South Wales were believed to have been started deliberately. This gave temporal patterns in the incidents whereby weekends showed higher occurrences of wildfires, particularly in the evening hours with 60% occurring between 1600 and midnight. Additionally, seasonal patterns were also noticed in regards to wildfires, with a peak occurring in spring between the months of March and May. This is supported by the work of Davies and Legg (2016) with the seasonal pattern attributed to the abundance of dead herbaceous and aerial shrub fuels. Interestingly, these temporal patterns coincide somewhat with the diurnal and seasonal variations of the PM measurements as reported in section 3.2, therefore alluding to their possible contribution.

In relation to air quality, wildfires are thought to be an important source of particulate matter due to the large quantities of vegetation capable of being burned. For example, Reff *et al.* (2009) found in the United States that the wildfire and agricultural burning were the third and fourth highest emission sources of $PM_{2.5}$ after unpaved road dust and agricultural soil. Considering only biomass combustion sources, these therefore are the highest contributors, followed by residential wood burning and prescribed burning.

The importance of wildfires however is not only limited to North America, European wildfires also show large contributions to air quality. For example, Barbosa *et al.* (2009)

concluded that between 2000 and 2005, there were 600,000 ha of forest land burnt every year across 23 European Countries. These wildfires released on average 52 and 44 kt/year of PM_{10} and $PM_{2.5}$ respectively. Unfortunately, the UK was omitted from this study due to lack of data at the time, however recent estimates from the NAEI for 2017 put accidental fires PM_{10} emissions at 0.48 kt and $PM_{2.5}$ emissions at 0.39 kt, calculated based on the emission factors shown in Table 13 and applying fuel loadings relative to the type of burnt area. As reported by the EMEP/EEA these fuel loadings are 2 kg/m^2 for grassland, 7.5 kg/m^2 for scrubland and 35 kg/m^2 for temperate forest, with burning efficiencies ranging between 20-50%. Likewise, total suspended particle emission factors as high as 17 g/kg wood burned were also reported, with a large potential range of between 2 - 80 g/kg (San-Miguel-Ayanz, 2016). With wildfires being largely dependent on dry events, these UK accidental fire PM emissions naturally show large variations by year, some of which reach 1.5 kt, as was the case for PM_{10} in 2003.

Critically, when source apportioning emissions, consideration needs to be given to the potential for emissions having undergone long-range transportation. For wildfire and other large-scale burning events this is particularly important as significant peaks can occur. For example, Witham and Manning (2007) performed forward and backward dispersion modelling for two UK episodes of unusually high PM_{10}, and attributed these to the long-range transport of smoke from agricultural burning and forest fires in western Russia.

5.2.2 Prescribed burning

Prescribed burning differs from the aforementioned wildfires as it is generally performed in a controlled manner as a land management technique. The UK has a long history of prescribed burning. Currently it is being performed almost entirely in upland areas, in a rotational manner of between 8 to 25 years, with the intention of increasing the productivity of livestock-grazing pasture and red grouse populations (Harper, 2018).

Yallop *et al.* (2006) estimates that for the English uplands alone, approximately 114 km^2 (11,400 ha) of moorland is burnt annually. However, for the UK as a whole, the value presently is likely to be significantly higher due to the Scotland showing the largest amount of prescribed burning, and due an increasing rate in annual number of burns from 2001 to 2011 (Douglas, 2015).

In contrast to wildfires which can span thousands of hectares, prescribed burns are on a much smaller scale per burn, typically between 0.12 ha and 0.55 ha (Yallop, 2005). Considering the previously mentioned total annual burn size is in to the thousands of hectares, this would therefore suggest many thousands of prescribed burns occur each year. Defra (2007) state all of these prescribed burns must be within the burning season from the 1st October until 15th April. This winter burning therefore coincides with the peak usage of domestic stoves for heating.

The emissions associated with prescribed burning in the UK are currently uncertain, however, the primary burn materials of dwarf shrub heath and bracken fall within the accidental fires vegetation category of the NAEI source sectors (Tsagatakis, 2018), giving emission factors of 0.000022 kt/ha and 0.000018 kt/ha for PM_{10} and $PM_{2.5}$ respectively. This is somewhat lower than those provided by the EMEP/EEA if burning of shrubland is assumed, which if after converting to the same units gives 0.00026 kt/ha for PM_{10} and 0.00022 kt/ha for $PM_{2.5}$ (San-Miguel-Ayanz, 2016). Tian *et al.* (2007) further notes that the frequency of the prescribed burns also impacts the level of emissions, with a 5-year fire return interval giving 72% greater emissions per fire than a 2-year interval, but causing 32% less emissions overall.

5.2.3 Agricultural burning

Agricultural burning of waste, also commonly referred to as crop residue burning or stubble burning, is the practise of intentionally burning the remains of agricultural crops after harvest. This is performed with the intention of cheaply and quickly clearing the fields for subsequent planting of crops, whilst killing pests and weeds.

Historically, agricultural burning is used a globally widespread technique, with Yevich and Logan (2003) estimating that as of 1985, 400,000 kt of crop residues were burnt in fields globally, accounting for around one third of all biofuel use, and 1,890 kt of total particulate matter. This problem has persisted in many areas into modern times, with regular air quality impacts reported in China (Zhuang *et al.*, 2018), India (Jain *et al.*, 2014) and the USA (McCarty *et al., 2008*).

Although agricultural burning within the UK used to be common practice, the crop residues (burning) regulations introduced in 1993 prohibited this technique, along with many other European nations. Despite this, it remains a controversial topic, with many farmers calling for it to be reintroduced. Furthermore, agricultural burning is still thought to be conducted in several other European countries regardless of any legislative restrictions. As discussed previously, this can cause negative implications for the UK's air quality due to long range transportation, with Whitham and Manning (2007) finding episodes of high PM in the UK could be traced back to agricultural burning and wildfires in Russia.

The EEA/EMEP 2016 guidebook estimates that as of 2005, agricultural burning accounted for 0.19% of total PM_{10} emissions in Europe, equivalent of 3.7 kt. Within this, the emission factors, given according to work by Jenkins et. al. (1996), are 5.8 $g.kg^{-1}$ dry matter for TSP, 5.7 $g.kg^{-1}$ dry matter for PM_{10}, and 5.4 $g.kg^{-1}$ dry matter for $PM_{2.5}$. In the UK, the NAEI assumes agricultural burning does not occur due to the regulations banning its use, however, accidental fires burning straw is still accounted for. This has emission factors of 11$g.kg^{-1}$ for PM_{10} and 9.0 $g.kg^{-1}$ for $PM_{2.5}$, leading to emissions for 2017 being estimated at 0.22 kt for PM_{10} and 0.18 kt for $PM_{2.5}$.

5.3 Section 5 summary

Other domestic burning emission sources

- Barbeques can have significant emissions of PM and are an increasingly popular method to cook food with 9.5 million units imported since 2015, and a rise in charcoal imports totalling around 86,000 tonnes per year in 2018.
- Commercial cooking may be an important source of PM emissions, with wood-fired pizza ovens alone consuming around 7 kg/hr of wood.
- Open burning of waste is a significant PM emission source, with estimates that this produces 4.7 kt of PM_{10} a year in the UK, equivalent to 4.7% of anthropogenic PM emissions.
- PM emission factors from open burning of garden waste vary drastically depending on the exact feedstock and combustion characteristics, with values as high as 16.9 g/kg reported.
- Bonfire night is known to cause peaks in PM emissions capable of exceeding WHO 24h exposure limits. The NAEI states that in 2017, 1.4 kt of PM_{10} and 1.2 kt of $PM_{2.5}$ derived from bonfire night.

Other non-domestic burning emission sources

- Wildfires are common in the UK, with on average 4,600 hectares burnt every year in England alone. This number is much greater over the UK as a whole, with some years having as much as 45,000 hectares burnt. These wildfires are often started deliberately, with peaks commonly occurring in spring and during evening hours.
- Wildfires are also widespread throughout Europe, with an estimated 600,000 hectares of forest land burnt each year, releasing 52 kt/year of PM_{10}. These emissions have the potential to travel great distances and impact UK's air quality.
- Emissions from wildfires are significant, with PM_{10} emission factors of 320 kg/ha reported for forest fires and 22 kg/ha for other vegetation.
- Prescribed burning of upland areas is used extensively in the UK, with 11,400 hectares burnt in England each year alone. This is restricted to between October and April when the vegetation is wet, therefore likely overlapping with peak stove usage.
- Agricultural (stubble) burning is banned in the UK, however, it is still common in some European countries with estimated emission of 3.7 kt of PM_{10} as of 2005. Agricultural burning in other countries such as Russia is believed to cause episodes of high PM in the UK.

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7. Appendices

7.1 Appendix 1 – Results of source appointment studies for RSF burning in literature[4]

Country	City	Species	Season	Class	RSF contr. (%)	Method	Reference
Australia	Huon Valley, Tasmania	$PM_{2.5}$	Winter	Urban/rural	77	PMF	Reisen et al. (2013)
	Launceston	PM_{10}	Winter	Urban	95	^{14}C and levoglucosan	Jordan et al. (2006)
	Brisbane	PM_{10}	Annual	Urban	7	CMB, TTFA and MLR	Chan et al. (1999a,b)
	Griffith University, Brisbane	$PM_{2.5}$	Annual	Suburban	15	CMB, TTFA and MLR	Chan et al. (1999a,b)
Austria	Vienna	OC	Winter	Mixed urban	31-40	Anhydrosugars	Caseiro et al. (2009)
	Vienna	OC	Summer	Mixed urban	5-6	Anhydrosugars	Caseiro et al. (2009)
	Graz	OC	Winter	Mixed urban	38-59	Anhydrosugars	Caseiro et al. (2009)
	Graz	OC	Summer	Mixed urban	8-15	Anhydrosugars	Caseiro et al. (2009)
	Salzburg	OC	Winter	Mixed urban	34-70	Anhydrosugars	Caseiro et al. (2009)
	Salzburg	OC	Summer	Mixed urban	7-10	Anhydrosugars	Caseiro et al. (2009)
	Vienna	PM_{10}	Winter	Mixed urban	7-10	Anhydrosugars	Caseiro et al. (2009)
	Vienna	PM_{10}	Summer	Mixed urban	1-2	Anhydrosugars	Caseiro et al. (2009)
	Graz	PM_{10}	Winter	Mixed urban	12-19	Anhydrosugars	Caseiro et al. (2009)
	Graz	PM_{10}	Summer	Mixed urban	4-6	Anhydrosugars	Caseiro et al. (2009)
	Salzburg	PM_{10}	Winter	Mixed urban	10-20	Anhydrosugars	Caseiro et al. (2009)
	Salzburg	PM_{10}	Summer	Mixed urban	2-4	Anhydrosugars	Caseiro et al. (2009)
	Sonnblick	OC	Winter	Rural	5.8-11	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	Sonnblick	EC	Winter	Rural	0.5-4.8	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	Sonnblick	OM	Winter	Rural	23	Levoglucosan	Puxbaum et al. (2007)
Belgium	Borgerhout	PM_{10}	Winter	Urban	8.6 ± 4.3	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Ghent	PM_{10}	Winter	Urban	9.2 ± 4.9	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Mechelen	PM_{10}	Winter	Suburban	11.3 ± 5.4	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Hamme	PM_{10}	Winter	Rural	21.9 ± 15.8	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Lier	PM_{10}	Winter	Rural	10.6 ± 6.3	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Retie	PM_{10}	Winter	Rural	9.9 ± 5.8	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Houtem	PM_{10}	Winter	Rural	9.3 ± 12.0	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Borgerhout	OC	Winter	Urban	36 ± 11	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Ghent	OC	Winter	Urban	40 ± 13	Monosaccharide anhydrides	Maenhaut et al. (2012)

NOTE: EC-Elemental Carbon, OC- Organic Carbon, OA-Organic Aerosol

[4] Adapted from Mitchell (2017)

Table A1 Continued

Country	City	Species	Season	Class	RSF contr. (%)	Method	Reference
Belgium	Mechelen	OC	Winter	Suburban	43 ± 12	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Hamme	OC	Winter	Rural	60 ± 22	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Lier	OC	Winter	Rural	43 ± 12	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Retie	OC	Winter	Rural	43 ± 12	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Houtem	OC	Winter	Rural	40 ± 53	Monosaccharide anhydrides	Maenhaut et al. (2012)
	Antwerp	PM_{10}	Winter	Urban	5.83 ± 1.84	Levoglucosan	Cordell et al. (2016)
	Ghent	OC	Winter	Urban	35	Monosaccharide anhydrides	Zdráhal et al. (2002)
Canada	Golden, BC	$PM_{2.5}$	Winter	Rural valley	31	PMF	Jeong et al. (2008)
	Edmonton, Alberta	PM_{1}	Winter	Suburban	17.1	PMF	Bari et al. (2015)
Chile	Temuco	PM_{10}	Winter	Urban	87		Sanhueza et al. (2012)
	Santiago	$PM_{2.5}$	Winter	Urban	10-40	CO tracer and chemical model	Saide et al. (2016)
	Valdivia	PAH	Winter	Urban	90	PAH by GC-MS	Bravo-Linares et al. (2016)
	Southern cities in Chile	$PM_{2.5}$	Winter	Urban	90	CO tracer and chemical model	Saide et al. (2016)
Czech Republic	Mladá Boleslav	PM_{1}	Winter	Urban	49	PMF	Hovorka et al. (2015)
	Prague	$PM_{2.5}$	Winter	Urban	39	Monosaccharide anhydrides	Saarikoski et al. (2008)
	Prague	OC	Winter	Urban	79	Monosaccharide anhydrides	Saarikoski et al. (2008)
	Ostrava	$PM_{2.5}$	Winter	Urban	42	PMF	Vossler et al. (2016)
	Ústí nad Labem	OC	Winter	Urban	5	CMB	Schladitz et al. (2015)
	Ústí nad Labem	OC (coal)	Winter	Urban	43	CMB	Schladitz et al. (2015)
	Ústí nad Labem	EC	Winter	Urban	57	PMF	Schladitz et al. (2015)
	Ústí nad Labem	$PM_{2.5}$	Winter	Urban	31	PMF	Schladitz et al. (2015)
	Brno	OC	Winter	Urban	24.1-34.2	Monosaccharide anhydrides and monocarboxylic acids	Křůmal et al. (2015)
	Brno	EC	Winter	Urban	16.6-17.7	Monosaccharide anhydrides and monocarboxylic acids	Křůmal et al. (2015)
	Šlapanice	OC	Winter	Rural	20.2-51.7	Monosaccharide anhydrides and monocarboxylic acids	Křůmal et al. (2015)
	Šlapanice	EC	Winter	Rural	14.7-45.8	Monosaccharide anhydrides and monocarboxylic acids	Křůmal et al. (2015)
Denmark	Vindinge	$PM_{2.5}$	Winter	Rural	10	Gaussian plume dispersion model	Glasius et al (2008)
	Copenhagen	PM10	Winter	Urban	4.0	Anhydrosugars	Caseiro and Oliveira (2012)
	Copenhagen	PM10	Winter	Rural	3.4	Anhydrosugars	Caseiro and Oliveira (2012)
Estonia	Tartu	PM2.5	Annual	Urban	40	Receptor model / CMB	Orru et al. (2010)

Table A1 Continued

Country	City	Species	Season	Class	RSF contr. (%)	Method	Reference
Finland	Kuopio	$PM_{2.5}$	Winter	Suburban	16	PMF	Yli-Tuomi et al. (2015)
	Kurkimäki	VOCs and benzene	Winter	Rural	26-48 and 35-62	CMB with VOC and PAH	Hellén et al. (2008)
	Helsinki	$PM_{2.5}$	Autumn	Urban	25	Monosaccharide anhydrides	Saarikoski et al. (2008)
	Helsinki	OC	Autumn	Urban	58	Monosaccharide anhydrides	Saarikoski et al. (2008)
	Helsinki	$PM_{2.5}$	Winter	Urban	18-29 and	Monosaccharide anhydrides	Saarnio et al. (2012)
	Helsinki	$PM_{2.5}$	Winter	Suburban	27-66	Monosaccharide anhydrides	Saarnio et al. (2012)
	Helsinki	OA	Winter	Suburban	50	Multilinear engine algorithm	Aurela et al. (2015)
France	Paris	$PM_{2.5}$	Winter	Urban BG	10-30	Aethalometer	Favez et al (2009)
	Lens	PM_{10}	Winter	Urban BG	25	PMF	Waked et al. (2014)
	Puy de Dôme	OC	Winter	Rural	7.1-14	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	Puy de Dôme	EC	Winter	Rural	0.6-6.5	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	Lille	PM_{10}	Winter	Urban	11.57 ± 3.38	Levoglucosan	Cordell et al. (2016)
	Puy de Dôme	OM	Winter	Rural	21	Levoglucosan	Puxbaum et al. (2007)
	Marseille	OA	Winter	Urban	48	AMS and PMF of offline filter extracts	Bozzetti et al. (2017)
	Lille	PM_{10}	Winter	Urban	7.8-15.4	Levoglucosan	Cordell et al. (2016)
Germany	Dettenhausen	PM_{10}	Winter	Rural	59	PMF	Bari et al. (2010)
	Augsburg	PM_{10}	Winter	Urban	25	CMB	Gu et al. (2013)
	Seiffen	OA	Winter	Rural	20	PMF	Poulain et al. (2011)
	Seiffen	PAH	Winter	Rural	62	PMF	Poulain et al. (2011)
	Schauinsland	OC	Winter	Rural	12-23	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	Schauinsland	EC	Winter	Rural	1-10	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	Schauinsland	OM	Winter	Rural	21	Levoglucosan	Puxbaum et al. (2007)
	Duisburg	$PM_{2.5}$	Autumn	Urban	13	Monosaccharide anhydrides	Saarikoski et al. (2008)
	Annaberg-Buchholz	$PM_{2.5}$	Winter	Rural town	30	PMF	Schladitz et al. (2015)
		EC	Winter	Rural town	55	PMF	Schladitz et al. (2015)
		OC	Winter	Rural town	22	CMB	Schladitz et al. (2015)
		OC (coal)	Winter	Rural town	33	CMB	Schladitz et al. (2015)
Greece	Thessaloniki	PM_{10}	Winter	Urban	8-12	CMB	Argyropoulos et al (2012)
	Thessaloniki	OM	Winter	Urban BG	39	CMB	Manoli et al. (2015)
	Athens	OA	Winter	Urban	43	PMF	Florou et al. (2017)
	Patras	OA	Winter	Urban	60	PMF	Florou et al. (2017)
Hungary	K-Puszta	OC	Winter	Rural	33-56	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	K-Puszta		Winter	Rural	2.7-13	14C, levoglucosan and cellulose	Gelencsér et al. (2007)

Table A1 Continued

Country	City	Species	Season	Class	RSF contr. (%)	Method	Reference
Hungary	K-Puszta	OM	Winter	Rural	47	Levoglucosan	Puxbaum et al. (2007)
Ireland	Tivoli Docks, Cork	EC	Summer	Urban BG	20	PMF	Healy et al. (2010)
		OC	Summer	Urban BG	21	PMF	Healy et al. (2010)
		$PM_{2.5}$	Summer	Urban BG	5	PMF	Healy et al. (2010)
		OC	Summer	Urban BG	28	PCA–MLR	Kourtchev et al. (2011)
		OC	Winter	Urban BG	80	PCA–MLR	Kourtchev et al. (2011)
		$PM_{2.5}$	Summer	Urban BG	6	PCA–MLR	Kourtchev et al. (2011)
		$PM_{2.5}$	Winter	Urban BG	28	PCA–MLR	Kourtchev et al. (2011)
		$PM_{2.5}$	Winter	Urban BG	46-50	PMF	Dall'Osto et al. (2014)
Italy	Lombardy	OC	Annual	Mixed	20-50	PMF and anhydrosugars	Piazzalunga et al. (2011)
	Lombardy	PM_{10}	Annual	Mixed	5-25	PMF and anhydrosugars	Piazzalunga et al. (2011)
	Po Valley	PAH	Winter	Rural	77% of BaP	CMB	Pietrogrande et al. (2015)
	Po Valley	OC	Winter	Rural	35	CMB	Pietrogrande et al. (2015)
	Milan	PM_{10}	Winter	Urban BG	14	PMF	Bernardonia et al. (2011)
	Oasi Le Bine	OC	Summer	Rural	10	CMB	Perrone et al. (2012)
	Oasi Le Bine	OC	Winter	Rural	85-96	CMB	Perrone et al. (2012)
	Oasi Le Bine	$PM_{2.5}$	Summer	Rural	3	CMB	Perrone et al. (2012)
	Oasi Le Bine	$PM_{2.5}$	Winter	Rural	27-31	CMB	Perrone et al. (2012)
	Propata	EC	Winter	Rural	53±9	Optical measurement	Massabò et al. (2015)
	Propata	OC	Winter	Rural	61±5	Optical measurement	Massabò et al. (2015)
	Genoa	EC	Winter	Urban BG	16±7	Optical measurement	Massabò et al. (2015)
	Genoa	OC	Winter	Urban BG	15±5	Optical measurement	Massabò et al. (2015)
Japan	Tokyo	PM_{10}	Winter	Suburban	24-28	14C	Uchida et al. (2010)
	Tokyo	PM_{10}	Summer	Suburban	39-42	14C	Uchida et al. (2010)
	Tokyo	PM_{10}	Winter	Urban	12.7	CMB	Okamoto et al. (1990)
Luxembourg	PM_{10} is dominated by the fine fraction (<1µm) in the winter months (Sep-Feb) due to domestic heating						Buchholz et al. (2014)
Netherlands	Amsterdam	$PM_{2.5}$	Winter	Urban	11	Monosaccharide anhydrides	Saarikoski et al. (2008)
	Wijk aan Zee	PM_{10}	Winter	Urban	1.3-4.1	Levoglucosan	Cordell et al. (2016)
	Cabauw	OA	Annual	Rural	0-23	PMF	Schlag et al. (2016)
	Wijk aan Zee	PM_{10}	Winter	Urban	2.74 ± 1.45	Levoglucosan	Cordell et al. (2016)
	Amsterdam	PM_{10}	Winter	Urban	4.78 ± 1.90	Levoglucosan	Cordell et al. (2016)

Table A1 Continued

Country	City	Species	Season	Class	RSF contr. (%)	Method	Reference
New Zealand	Auckland (KLD)	$PM_{2.5}$	Winter	Suburban	69	Ion beam analysis and PMF	Trompetter et al. (2010)
	Hastings	$PM_{2.5}$	Winter	Urban	91	Ion beam analysis and PMF	Trompetter et al. (2010)
	Masterton	$PM_{2.5}$	Winter	Rural	80	Ion beam analysis and PMF	Trompetter et al. (2010)
	Upper Hutt	$PM_{2.5}$	Winter	Urban	63	Ion beam analysis and PMF	Trompetter et al. (2010)
	Christchurch	$PM_{2.5}$	Winter	Urban	79	Ion beam analysis and PMF	Trompetter et al. (2010)
	Auckland (KLD)	$PM_{2.5}$	Summer	Suburban	13	Ion beam analysis and PMF	Trompetter et al. (2010)
	Hastings	$PM_{2.5}$	Summer	Urban	9	Ion beam analysis and PMF	Trompetter et al. (2010)
	Masterton	$PM_{2.5}$	Summer	Rural	45	Ion beam analysis and PMF	Trompetter et al. (2010)
	Upper Hutt	$PM_{2.5}$	Summer	Urban	25	Ion beam analysis and PMF	Trompetter et al. (2010)
	Christchurch	$PM_{2.5}$	Summer	Urban	26	Ion beam analysis and PMF	Trompetter et al. (2010)
	Nelson	$PM_{2.5}$	Annual	Urban	77	Ion beam analysis and PMF	Ancelet et al. (2015)
	Nelson	PM_{10}	Annual	Urban	48	Ion beam analysis and PMF	Ancelet et al. (2015)
	Wainuiomata	$PM_{2.5}$	Winter	Suburban	48	PMF	Davy et al. (2012)
	Alexandra	PM_{10}	Winter	Urban	86-91	PMF	Ancelet et al. (2014)
	Masterton	PM_{10}	Winter	Rural	89-90	PMF	Ancelet et al. (2012)
Norway	Hurdal	EC	Winter	Rural	7-12	^{14}C & monosaccharide anhydrides	Yttri et al. (2011)
	Oslo	EC	Winter	Urban BG	6-11	^{14}C & monosaccharide anhydrides	Yttri et al. (2011)
	Hurdal	OC	Winter	Rural	27-41	^{14}C & monosaccharide anhydrides	Yttri et al. (2011)
	Oslo	OC	Winter	Urban BG	25-39	^{14}C & monosaccharide anhydrides	Yttri et al. (2011)
	Oslo	$PM_{2.5}$	Winter	Urban	27	PMF	Laupsa et al. (2009)
Poland	National average	PM_{10}	Annual	Nationwide	50		Juda-Rezler et al. (2011)
	Nowa Huta, Krakow	PM_{10} (coal)	Winter	Urban	70	AMS	Mira-Salama et al. (2008)
	Krakow	PM_{10} (coal)	Winter	Urban	50	CMB and C-PMF	Junninen et al. (2009)
	Krakow	B(a)P (coal)	Winter	Urban	90	CMB and C-PMF	Junninen et al. (2009)
Portugal	Aveiro	OC	Winter	Urban	52-69	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	Aveiro	EC	Winter	Urban	4-12	14C, levoglucosan and cellulose	Gelencsér et al. (2007)
	Nationwide average	PM_{10}	Annual		18	MM5/CHIMERE air quality model	Borrego et al. (2010)
	Foros de Arrão	$PM_{2.5}$	Winter	Rural	17	PMF	Canha et al. (2014)
	Porto	PM_{10}	Winter	Rural	3	Anhydrosugars	Caseiro and Oliveira (2012)
	Porto	PM_{10}	Winter	Urban	3	Anhydrosugars	Caseiro and Oliveira (2012)
	Aveiro	OM	Winter	Urban	68	Levoglucosan	Puxbaum et al. (2007)
	Azores	OM	Winter	Rural	18	Levoglucosan	Puxbaum et al. (2007)
Slovakia		High contribution to PM_{10} from local residential heating across the country					Krajčovičová et al. (2014)

Table A1 Continued

Country	City	Species	Season	Class	RSF contr. (%)	Method	Reference
Slovenia	Maribor	Substantial winter emissions from residential wood burning					Kitanovski et al. (2012)
South Korea	Seoul	$PM_{2.5}$	Summer	Urban	12.1	PMF	Heo et al. (2009)
	Jeju Island	$PM_{2.5}$	Spring	Rural	25	PMF	Han et al. (2006)
	Incheon	$PM_{2.5}$	Annual	Urban	6.1	PMF	Choi et al. (2013)
Spain	Grenada	TC	Winter	Suburban	41-47	Aethalometer and levoglucosan	Titos et al. (2017)
	Huelva, Seville	OA	Winter	Urban	15	PMF	Diesch et al. (2012)
Sweden	Tanumshede	$PM_{2.5}$	Winter	Rural	25	PMF	Molnar and Sallsten (2013)
	Hagfors		Winter	Rural			Gustafson et al. (2007)
	Lycksele	$PM_{2.5}$	Winter	Rural town	70	PMF and levoglucosan	Hedberg et al (2006)
	Lycksele	PM_{10}	Winter	Rural town	36-82	PMF	Krecl et al. (2008)
	Vavihill	OC % of TC	Winter	Rural	32	Levoglucosan and 14C	Genberg et al. (2011)
	Vavihill	EC % of TC	Winter	Rural	7	Levoglucosan and 14C	Genberg et al. (2011)
Switzerland	Zurich	OM	Summer	Urban BG	10	AMS & PMF	Lanz et al. (2007)
	Zurich	OM	Winter	Urban BG	35-40	AMS & CMB/PMF	Lanz et al. (2008)
	Roveredo	OM	Winter	Rural valley	94	^{14}C	Alfarra et al (2007)
	Zurich	OC	Winter		41	^{14}C	Szidat et al. (2006)
	Zurich	EC	Winter		20-30	^{14}C	Szidat et al. (2006)
	Zurich	OC	Summer		10	^{14}C	Szidat et al. (2006)
	Zurich	EC	Summer	Urban	4-8	^{14}C	Szidat et al. (2006)
	Zurich	BC	Winter		24±11	Aethalometer	Herich et al. (2011)
	Payerne	BC	Winter	Rural	33±12	Aethalometer	Herich et al. (2011)
	Magadino-Cadenazzo	BC	Winter	Rural	30±11	Aethalometer	Herich et al. (2011)
	Alpine regions	EC	Rural	Winter	42-49	14C, levoglucosan and water soluble ionic species	Zotter et al. (2014) Gianini et al. (2013)
Turkey	Zonguldak	$PM_{2.5}$ (coal)	Winter	Urban	81	PAH ratios	Akyüz et al. (2008)
	Bogazici Univ,	PAH	Winter	Rural	19	CMB	Hanedar et al. (2011)
	Bursa	PAH	Winter	Urban	22.8	PAH ratios	Esen et al. (2008)
	Bursa	PAH (coal)	Winter	Urban	5.7	PAH ratios	Esen et al. (2008)
UK	LDN and BMX	Concentrations generally very low compared to the rest of Europe and with other studies					Harrison et al. (2012)
	London	OA	Winter	Urban BG	38	AMS and PMF	Young et al (2015)
	London	OA	Winter	Urban BG	11	AMS and PMF	Young et al (2015)
	London	OA	Annual	Urban BG	34	AMS and PMF	Young et al (2015)
	London	POA	Annual	Urban BG	43	AMS and PMF	Young et al (2015)
	London	PM_{10}	Annual 3 year	Urban BG	7-10	Aethalometer & levoglucosan	Fuller et al. (2014)

Table A1 Continued

Country	City	Species	Season	Class	RSF contr. (%)	Method	Reference
UK	London	PM_{10}	Annual 3 year	Urban BG	7-10	Aethalometer & levoglucosan	Fuller et al. (2014)
	London	PM_{10}	Winter	Urban BG	10	Aethalometer	Fuller et al. (2014)
	London	PM_{10}	Summer	Urban BG	2	Aethalometer	Fuller et al. (2014)
	London	BC	Winter	Urban BG	23	Aethalometer	Fuller et al. (2014)
	London	BC	Summer	Urban BG	11	Aethalometer	Fuller et al. (2014)
	North Kensington	OC	Winter	Urban BG	15	CMB	Yin et al. (2015)
	Harwell	OC	Winter	Rural	28	CMB	Yin et al. (2015)
	North Kensington	$PM_{2.5}$	Winter	Urban BG	4	CMB	Yin et al. (2015)
	Harwell	$PM_{2.5}$	Winter	Rural	7	CMB	Yin et al. (2015)
	North Kensington, London	BC	Winter	Urban	26±13	PMF, SP2, aethalometer	Liu et al. (2014)
	Holme Moss, West Yorkshire	BC	Winter	Rural	45	PMF	Liu et al. (2011)
	Leicester	PM_{10}	Winter	Urban	3.21 ± 2.36	Levoglucosan	Cordell et al. (2016)
USA	Truckee Meadows, Nevada	$PM_{2.5}$	Winter	Urban valley	11-51	CMB	Chen et al. (2012)
	Fresno, CA	OA	Wnter	Urban valley	33	AMS and PMF	Young et al (2015)
	Fresno, CA	$PM_{2.5}$ and OC	Winter	Urban valley	18% and 41%	Anhydrosugars	Gorin et al. (2006)
	Fresno, CA	$PM_{2.5}$	Winter	Urban valley	29-31	CMB	Chow et al. (2007)
	Underhill, Vermont	$PM_{2.5}$	Winter	Rural	12.5-24.3	PMF	Polissar et al. (2001)
	Underhill, Vermont	$PM_{2.5}$	Summer		6.1-13.7	PMF	Polissar et al. (2001)
	Pasadena	$PM_{2.0}$	Annual	Urban	9.6	CMB	Schauer et al. (1996)
	Pasadena	OA	Annual	Urban	19.3	CMB	Schauer et al. (1996)
	Downtown LA	$PM_{2.0}$	Annual	Urban	5.7	CMB	Schauer et al. (1996) Schauer et al. (1996)
	Downtown LA	OA	Annual	Urban	12.3	CMB	
	West LA	$PM_{2.0}$	Annual	Urban	10.8	CMB	Schauer et al. (1996)
	West LA	OA	Annual	Urban	22.0	CMB	Schauer et al. (1996)
	Rubidoux	$PM_{2.0}$	Annual	Urban	1.3	CMB	Schauer et al. (1996)
	Rubidoux	OA	Annual	Urban	5.0	CMB	Schauer et al. (1996)
	Libby, Montana	$PM_{2.5}$	Winter	Rural	81	CMB	Ward et al. (2010)
	Seattle (multiple)	$PM_{2.5}$	Annual	Suburban	7-31	PMF	Kim and Hopke (2008a)
	Olympic N.P, WA	$PM_{2.5}$	Annual	Rural	13	PMF	Kim and Hopke (2008b)
	Portland, OR	$PM_{2.5}$	Annual	Urban	27	PMF	Kim and Hopke (2008b)
	Fairbanks, Alaska	$PM_{2.5}$	Winter	Urban	62.7-81.2	CMB	Ward et al. (2012)
	Central LA & Riverside	OC	Annual	Urban	9-10	PMF	Heo et al. (2009)
	Waterbury, VM	$PM_{2.5}$	Annual	Rural	28-46	CMB and others	Sexton et al. (1985)

Table A1 Continued

Country	City	Species	Season	Class	RSF contr. (%)	Method	Reference
USA	Boise, Idaho	$PM_{2.5}$	Winter	Urban	62-94	14C and DMP Isomers	Benner et al. (1995)
	Fairbanks, Alaska	$PM_{2.5}$	Annual	Urban	31-66	14C	Busby et al. (2016)
	Fairbanks, Alaska	$PM_{2.5}$	Annual	Urban	20-61	Levoglucosan	Busby et al. (2016)
	Fairbanks, Alaska	$PM_{2.5}$	Annual	Urban	65-68	CMB	Busby et al. (2016)
	Rochester, NY	$PM_{2.5}$	Winter	Urban	17.30%	Aethalometer, levoglucosan & K	Wang et al. (2011)
	BH, Seattle	$PM_{2.5}$	Annual	Urban	24-31%	CMB model	Wu et al. (2007)
	Montana	$PM_{2.5}$	Winter	Rural	55.5-77.0%	CMB and ^{14}C	Ward and Lange (2010)