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The Air Quality and Climate Impacts of Biomass Use as an Energy Source: A Review

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ABSTRACT

Commitments made to meet the goals of the Paris Agreement, and the need to develop low carbon sources of power and heat, are likely to lead to the continued or even increased use of biomass as an energy source. Biomass is already replacing coal within large scale electricity generation systems. At smaller scales it is utilised within combined heat and power systems, as well domestically for the provision of heat. Unabated, biomass combustion leads to a range of pollutant emissions, including CO, CO_2, methane, black and organic carbon, which are of relevance from a climate perspective, as well as particulate matter, nitrogen oxides, volatile organic compounds, dioxins, furans and precursors of ozone and secondary organic aerosols, which impact on air quality (AQ) and human health. Epidemiological data suggests that biomass smoke contributes to at least 40 000 premature deaths per year in Europe, as well as negatively affecting respiratory and cardiovascular health. Hence, if biomass is to be used as a future energy source, its AQ impacts need to be addressed. Here, a short review is provided of the potential climate and AQ impacts of the direct use of solid biomass as a fuel for the provision of power and heat. The review provides a brief summary of the chemical and physical characteristics of emissions from biomass utilisation for energy provision. The impacts of biomass burning on ambient AQ and health (in particular for fine particulates) are addressed in both outdoor and indoor environments, and for industrialised and developing countries. Issues of appliance scale, and how these influence available mitigation options, emissions factors and regulatory limits for biomass combustors, are discussed. Final perspectives are offered, from the point of view of the need to develop "win-win" strategies for the future utilisation of biomass which minimise both climate AND air quality impacts.

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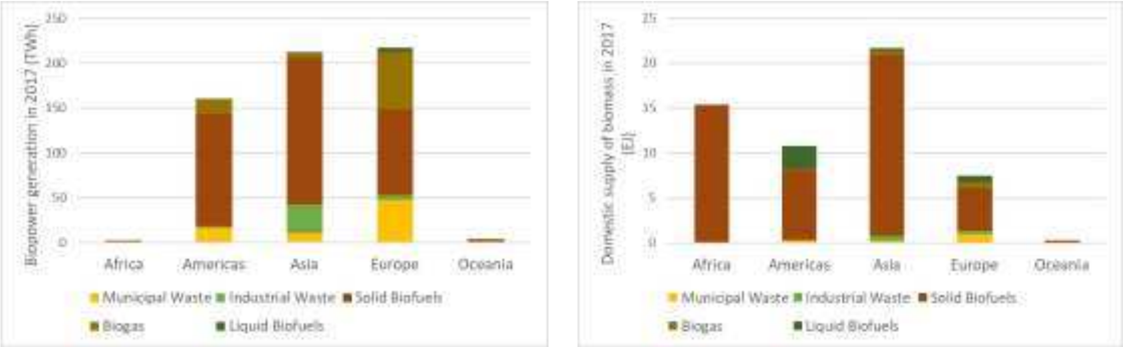
1. INTRODUCTION

The commitment of countries around the world to meeting the goals of the Paris Agreement[1] necessitates the rapid uptake of low carbon energy systems for the provision of electricity, heat and mobility. The large-scale deployment of low carbon technologies such as solar, wind and hydro power has led to reductions in the carbon intensity of the electricity sector in many countries, with the greenhouse gas (GHG) emission intensity of electricity generation within the EU for example, falling from over 500 g CO_2eq/kWh in 1990 to around 275 CO_2eq/kWh in 2019.[2] However, in sectors that are harder to decarbonise, such as heat and transportation, progress has been slower. The transport sector unfortunately provides some alarm bells in terms of strategies that were previously chosen with the aim to reduce GHG emissions, that have impacted on other sustainability goals including air quality (AQ). Within the EU, for a period of time, diesel vehicles were favoured over their gasoline counterparts due to lower CO_2 emissions per km. However, the increased uptake of diesel vehicles has perpetuated high concentrations of nitrogen oxides (NOx) and airborne particulate matter (PM) in European cities, with known consequences on human health. It is important to acknowledge therefore, that climate change is not the only environmental challenge that results from emissions to the atmosphere. The World Health Organisation (WHO) estimates that around 7 million deaths occur globally each year as a result of exposure to air pollution, including both outdoor and indoor exposure.[3] Although it is somewhat difficult to separate the effects of individual pollutants on morbidity and mortality from epidemiological data,[4] it is known that a number of different pollutants are involved, including nitrogen dioxide (NO_2), fine particulates ($PM_{2.5}$), sulphates, ozone and dioxins. There are some complex issues at play, since technologies proposed to reduce CO_2 emissions do not necessarily provide the best way of tackling local air pollution, or even of reducing other compounds of relevance to radiative forcing such as nitrous oxide, methane and atmospheric aerosol. It is therefore important to ask whether proposed strategies for reducing CO_2 have the desired effect on atmospheric greenhouse gases (GHGs) and aerosols and consequently the total radiative forcing of the atmosphere, as well as what the implications might be for local AQ. Low carbon options should only be chosen as part of future energy strategies if they do not lead to a further deterioration of AQ, and better still if they lead to AQ improvements.

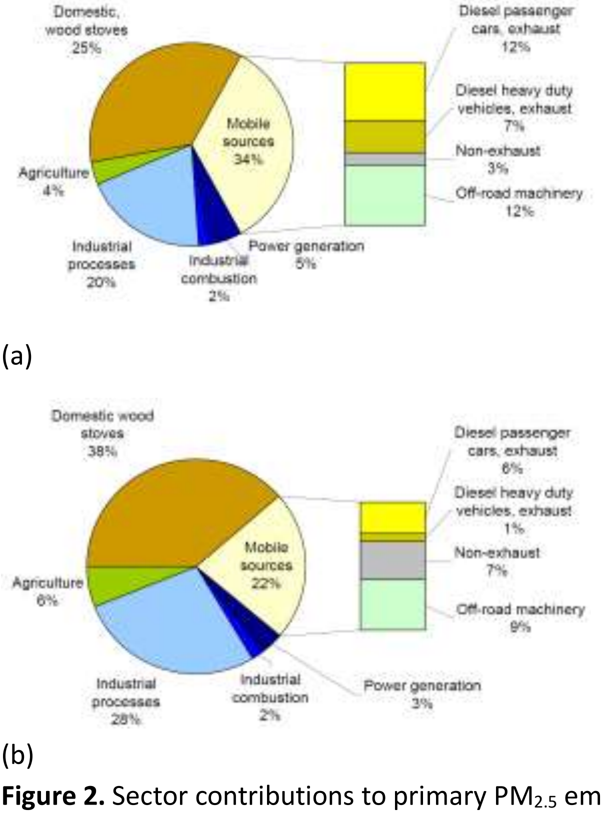
Passenger transport is likely to move away from internal combustion engines, with the electrification of passenger cars rising in many European countries, led by Norway which surpassed 50% of new car sales being electric in 2020.[5] At least part of the impact of diesel exhaust emissions on air urban quality is likely to decline as a result. Globally, however, despite the increasing penetration of renewables and hydro, electricity production is still dominated by the combustion of fossil fuels, with the International Energy Agency (IEA) estimating that 38% of that production came from coal combustion in 2018. As countries seek to address commitments made through the Paris agreement, a move away from coal for electricity production will be necessary, with the replacement of coal by biomass fuels being one potential option within mixed energy strategies.[6] In 2017, renewables made up around 14 % of global energy supply, with biomass forming 70 % of the renewables total.[7] Globally, biomass supplied close to 600 TWh of electricity in 2019, more than double the amount supplied in 2009.[8] Usage of biomass for power generation, is dominated by Europe and Asia as illustrated in Figure 1a. A report from the International Institute for Applied Systems Analysis estimates that, between 2005 and 2030, the use of biomass for energy production in the EU is likely to double.[9] Globally, as part of the Sustainable Development Scenario, 2000-2030, the use of biomass for power is predicted to almost double from current rates by 2030.[8]



(a) (b)

Figure 1. Global biomass usage figures for 2017 within different continents (a) power generation from biomass (b) domestic supply of biomass.[7]

There are also significant challenges in providing low carbon heat, which is required in both industrial and domestic sectors. Industrially, natural gas is currently used for heat provision in many different processes including steel and glass production, chemical and mineral industries. Whilst liquid biofuels are being explored as a potential part of the future energy mix for such industries, solid biomass is unlikely to form a major component. The same may not be true however, for the provision of domestic heat and for combined heat and power (CHP). Across Europe for example, biomass is used in small scale residential (<50 kilowatt thermal (kWth)) heating appliances and in medium scale (<20 MWth) district heating/industrial systems as well as in large centralised CHP plants.[10] The current number of residential biomass combustion systems within the EU is estimated to be ~65 million direct heating appliances (fireplaces, stoves and cookers) and ~8 million indirect heating appliances (boilers), firing mainly using wood logs, but also wood chips and fuel pellets.[11] Figure 2 shows that such devices, particularly domestic stoves, have been estimated to make a substantial contribution to primary fine particulate emissions within the EU, and were predicted to have grown in terms of their relative importance compared to other sources such as traffic, from 25% of total primary sources in 2000, to 38% in 2020.[12]



(a)

(b)

Figure 2. Sector contributions to primary $PM_{2.5}$ emissions in the EU15, predicted for (a) 2000 and (b) 2020. Reproduced from Amann, M.; Bertok, I.; Cofala, J.; Gyarfas F.; Heyes, C.; Klimont, Z.; Schöpp, W.; Winiwarter, W., M. A Baseline Scenarios for the Clean Air for Europe (CAFE) Programme, 2005, Submitted to the European Commission Directorate General for Environment, Directorate C – Environment and Health. Copyright [2005] with permission from IIASA.[12]

CHP offers efficiency benefits over conventional power generation since the process captures and utilises heat that is a by-product of the electricity generation process. By generating heat and power simultaneously, CHP can reduce carbon emissions by up to 30% compared to the separate means of conventional generation via a boiler and power station, with an overall efficiency of > 80%. CHP is therefore likely to form part of strategies to decarbonise both electricity and heat sectors with low carbon fuels such as biomass likely to form part of the mix, potentially replacing natural gas. Williams et al., based on a study of several pathways to meet carbon reduction targets in the UK Climate Change Act, suggest that to a certain extent, particulate emissions reductions resulting from the electrification of transport could be balanced by significant increases in biomass burning in pathway scenarios involving CHP and domestic biomass use.[13] These issues therefore pose the question as to whether the use of biomass as an energy source has the potential to become the "new diesel" in terms of being promoted as a lower carbon source of power and heat compared to fossil fuels, but with potentially unwanted side effects in terms of increased air pollution exposure, especially to fine particulates and NO_2.

Globally, biomass is extensively used for both cooking and heating with domestic supplies of biomass being significant across all continents apart from Oceania as shown in Figure 1b. The regional profile for domestic usage of biomass is unsurprisingly quite different from that for power generation, with the Asian and African continents dominating domestic supply, due to usage of firewood and charcoal fuels for cooking and heating. Within the developing world the current energy landscape and future perspectives are quite different from those in industrialised countries. It was estimated in 2000 that approximately half of the global population relied on coal and biomass for domestic heating and cooking, with significant impacts on AQ and health.[14] Here then, improving combustion technologies for the utilisation of biomass fuels in order to lower emissions, has the potential to provide significant benefits to both health-related air pollution problems as well as climate change.

The review will focus on the AQ, and to a lesser extent, the health and climate impacts of the utilisation of solid biomass fuels for the provision of heat and electricity both in indoor and outdoor settings. Solid biomass dominates the global energy use of biofuels, with 86% of use in the form of primary solid biofuels in 2017.[7] Whilst solid biomass can be processed to form a wide range of products including gas and liquid fuels, the intention here is to focus on the impact of its use in solid form i.e. where the fuel fed directly to the combustion device is a solid, although this may include processed solid fuels and internal gasification strategies. The nature of the short review necessitates a selective rather than a comprehensive use of references. Examples of both policy and technically driven mitigation strategies will be given. For policy driven measures in developed countries, the focus will be more on the European region, since not all countries can be covered in such a short review and the challenges faced by Europe are representative of other industrialised nations. Final discussions will offer future directions for the effective use of solid biomass resources within the context of reducing AQ and GHG impacts in both industrialised and developing regions.

2. POTENTIAL CLIMATE IMPACTS FROM BIOMASS BURNING

The combustion of biomass can result in emissions of relevance to both air quality and climate, and it is important to consider both impacts in an integrated way when designing strategies for the utilisation of biomass. Hence in this section, the potential climate impacts of biomass utilisation as an energy source are covered from the perspective of both emissions to atmosphere, and assumptions about the carbon neutrality of biomass resources.

2.1 Climate Impacts of Black Carbon and Organic Carbon

Black carbon (BC) plays an important role in the Earth's climate system because it absorbs solar radiation, influences cloud processes, and through deposition can alter the melting of snow and ice cover. Bond et al. provide an estimate for the direct radiative forcing of atmospheric BC in the industrial-era (1750 to 2005) due to absorption of +0.71 Wm^{-2} with 90% uncertainty bounds of (+0.08, +1.27) Wm^{-2}.[15] In addition to direct forcing effects, BC aerosol can influence cloud characteristics and can also influence the surface albedo through deposition onto snow and ice. When cloud and cryosphere adjustments are taken into account this increases radiative forcing effects to +1.1 Wm^{-2} with 90% uncertainty bounds of +0.17 to +2.1 Wm^{-2}. This makes BC the second most important contributer to changes in radiative forcing since 1750, with only CO_2 being higher at 1.68 Wm^{-2} (1750 to 2011).[16] Since BC has a short atmospheric lifetime, several authors have suggested that reducing emissions of BC would provide a rapid payback in terms of reducing global warming.[17,18] Hence reducing BC emissions from combustion provides a win-win scenario of leading to improvements both in climate forcing and in human health outcomes by improving AQ. Organic carbon (OC) on the other hand is thought be a climate cooler with estimates of forcing in the industrial era of -0.29 (-0.47 - -0.08) Wm^{-2}.[16] BC and OC are both major pollutants from biomass burning and their relative emissions will influence subsequent climate impacts. It is therefore vital that emissions profiles of both BC and OC from biomass combustors are well understood and this point will be returned to in section 7.

2.2 Gaseous Emissions of Relevance to Climate Impact

In addition to aerosols, biomass burning may contribute to emissions of gaseous pollutants such as methane, NOx, N_2O and SO_2 that can impact on radiative forcing. Methane and N_2O both have positive global warming potentials (GWP) which, on a 100 year time horizon, are significantly higher on a per molecule basis than CO_2 (GWP of 1) at 28 and 265 respectively.[16] The situation for emissions of NOx and SO_2 is different. Nitrate and sulphate aerosols that form within the atmosphere as a result of their emissions, contribute negatively to radiative forcing since such aerosols can scatter incoming radiation. NOx also contributes to the atmospheric formation of the OH radical which can help to consume methane, again leading to a negative radiative forcing effect. NOx emissions however, can also contribute to the formation of ozone in the troposphere which acts as a GHG and a positive radiative forcer. There is a high degree of uncertainty within the radiative forcing estimates for NOx but the most recent report from the IPCC suggests that NOx emissions have had a slightly negative overall effect on changes to radiative forcing since 1750 of -0.15 W/m^{-2} although with a large uncertainty range of -0.34 - +0.03 W/m^{-2}.[16] The overall impact of biomass utilisation as an energy source on global NOx will to a large extent depend on the energy/fuel sources that are being replaced and the relative emissions factors for the different fuels as discussed further in section 8.

Globally, N_2O emissions are dominated by agricultural sources rather than combustion ones, although this is of relevance to the use of biomass as an energy source where nitrogen based fertilisers are used to enhance yields. Full life cycle analyses (LCAs) are required to assess the overall GHG emissions of biomass fuel utilisation from production, to transportation and final end use within different energy sectors. The review of such lifecycle studies is outside the scope of this work, and although there are a growing number of reviews of LCAs for liquid biofuel use within the transport sector (so called Well-to-Wheels analyses),[19,20] there are fewer addressing the use of solid biomass fuels in the electricity and heat sectors. A recent example however, compares the life cycle impacts of bioenergy power plants fuelled by three types of residual biomass (forest residues, cereal straws and cattle slurry) with fossil based systems using the European power generation mix as well as coal and natural gas power plants.[21] Systematic reviews of bioenergy LCAs were also presented by Muench and Guenther[22] and Varun et al.[23] focussing on the electricity and heat sectors. A specific study for the heat sector was carried by Giuntoli et al. who estimated that emissions of N_2O are responsible for 27%, 13%, 6% of the total GHG impact of the use of logs within a domestic stove, wood chips used within a district heating plant, and pellets used within a domestic stove respectively.[24] Emissions of methane from both biogenic and fossil sources had a more minor influence: 9%, 8% and 7% for the log, chip and pellet pathways respectively. GHG emissions from biomass utilisation cannot therefore be ignored. Such LCAs are critical to the assessment of policies being developed to mitigate climate change, but of course the robustness of the conclusions drawn will depend heavily on the accuracy of the inputs to the models, including emissions factors for each part of the pathway such as crop fertilisation, fuel transportation, fuel processing, combustion, etc. The methodologies used for estimating GHG emissions factors for a range of solid and gaseous bioenergy pathways is discussed in Giuntoli et al. within the context of assessing compliance with the EU's Renewable Energy Directive (RED) (2009/28/EC) and the Fuel Quality Directive (FQD) (2009/30/EC).[25] The report presents emissions factors for N_2O, methane and overall $CO_{2\,eq}$ for a range of production and utilisation pathways including those for pellet boilers, pellet and wood chip based CHP plant, and straw based boilers, and for different biomass sources, fuel transportation, processing and fertilisation strategies.

2.3 Negative Emissions Technologies (NETs)

Biomass is a very heterogeneous material and the impacts of its utilisation on both climate and AQ depend very much on how it is sourced and treated prior to its combustion.[26] In this section, factors potentially affecting the carbon neutrality of selected biomass fuels are briefly covered.

Even within the targets set by the Paris Agreement, some non-zero sources of GHGs are likely to be unavoidable. This has led to the concept of negative emissions strategies or technologies (NETs), where the net result of the process should be to remove CO_2 from the atmosphere. NETs are part of all pathways to net zero CO_2 by 2050 that were modelled for the 2018 Intergovernmental Panel on Climate Change (IPCC) report on achieving only 1.5 °C of warming by 2100. The slower GHG emissions are reduced, the larger such technologies feature in the proposed pathways.[27] In pathway P4, where a resource- and energy-intensive scenario is assumed, bioenergy, carbon capture and storage (BECCS) technologies feature very heavily by the middle of the 21st Century, sequestering 20 $GtCO_2$/yr. Within BECCS, the carbon is assumed to be fully absorbed during biomass growth, and is captured pre- or post-combustion and stored underground indefinitely. If only the combustion process is considered, then producing a kWh of electricity from wood causes significantly more CO_2 than from using coal. The assumption of carbon neutrality stems from the assumption that the carbon released when biomass is burned, is sourced from forests that are sustainably managed such that it will be recaptured by a new stock of trees in the future.[28] In the domestic sector, the use of wood has been marketed as renewable, since it is at least possible that the source of wood may be renewed or replanted, thus over time sequestering the CO_2 that is released on its combustion. That time however, may be decades,[29,30] when we in fact require very short time scale emission reduction strategies in order to avoid a 1.5 °C overshoot. The carbon reduction benefits of bioenergy compared to the use of fossil fuels are thus time-dependent[29] and supply to both the domestic market and large scale power producers leads to complex CO_2 accounting issues.

A valid question is whether the potential for reliance on NETs in current plans is potentially locking us into energy and resource intensive lifestyles both now and in the near future, based on the premise of speculative technologies further down the line.[26] The scale of biomass resource requirement assumed in the more BECCs intensive pathways put forward by the IPCC, is typically one to two times the area of India, which raises questions about land availability, biodiversity loss, competition with food production, and competing demands for bioenergy resources from other sectors such as transport, heating, and industrial processes.[26]

2.4 The Use of Forestry Products

Globally, the estimate for wood pellet use in 2018 is > 50 million tons per annum, with Europe in particular forming the largest consumer region.[31] The total consumption of wood pellets in the EU was estimated to be 20.3 million metric tons in 2015 with 70 % of this supply sourced from the US.[32] By country, the UK was the top consumer of wood pellets in the EU region in 2018, with 8 million metric tons used. This has occurred as a result of the large scale switching of power generation from coal to biomass at the UK's largest power station, Drax (4 GW), causing the UK to become the world's largest importer of wood pellets over a period of just 5 years.[33] The supply is predominantly sourced from the US, Canada and Latvia. Drax is mandated under law to include lifecycle emissions from the cultivation, harvest, processing and transport of wood pellets, and claims that most come from waste products such as thinnings, forest and sawmill residues. The methodology potentially however, ignores large changes in forest carbon stocks and indirect effects caused by increased demand for wood products. The approach was subsequently challenged by a counter factual model which asks the question, what will happen to this waste wood if it is not pelletised and exported to a distant power station? The report found that a wide range of scenarios could result, from wood burning having the potential to reduce atmospheric CO_2, all the way to it being worse than burning coal.[33] The differences arise from the vastly differing CO_2 emissions that can arise when in reality, using these seemingly waste products, could actually be diverting them from other end uses. The distinction for forest residues is between fine woody materials, which may be better burnt in a power station instead of being left on the forest floor to rot and release methane, compared to larger diameter wood that could potentially store its carbon on the forest floor for many years.[29,33] Giuntoli et al. suggest a high sensitivity of the mitigation potential of using forest residues to decay rates of material on the forest floor, concluding that power generation from forest logging residues is an effective mitigation solution compared to the current EU mix, only when decay rates are above 5.2% a^{-1}.[21] A perhaps more controversial area is the harvest rates that are used to supply the increased demand for pellets in forests that are typically naturally regenerating. According to the counterfactual model, if forests are harvested every 60 years instead of every 70 years, then the amount of carbon locked in the forest is reduced, and the CO_2 emissions from the combustion of the resulting thinnings exceeds that of coal usage. The general principle for the sustainable use of wood should be that stocks (and their embedded carbon) increase more rapidly due to the development of a market that uses the wood, than they would have in its absence.[28] If forests are cultivated on marginal lands this may well be the case, but where biomass is derived from waste products of industries primarily concerned with producing other wood products such as timber or wood pulp for paper, or when land with high carbon stocks is converted to low productivity bioenergy plantations, this cannot be taken for granted.[28,29] Whether stock regeneration management practices are adjusted to suit the waste wood market also muddies the waters as highlighted above. Drax on the other hand argues that “forests are growing, not shrinking, in all of the US states that Drax sources from” and hence that carbon stocks are increasing as a result of this new market.[33] Biodiversity is also a key factor that must be considered if we are to seek win-win solutions, as discussed in a recent report on the use of biomass for power in the EU by the European Commission Joint Research Centre (JRC).[34] The JRC report also highlights accounting anomalies in reported usage of woody biomass across the EU compared to sources. The issues around carbon accounting are clearly complex, which makes trading in waste wood resources difficult to manage from a sustainability accountability and enforcement perspective. What looks like an NET on paper may or may not play out that way in the real world. The use of detailed LCAs and counterfactual models will be required to adequately assess the overall GHG savings from different feedstocks and utilisation strategies.

2.5 Dedicated Energy Crops

Dedicated energy crops are grown specifically for their utilisation in energy production in ways that do not displace food production. They include short rotation wood crops such as willow, and perennial grasses such as miscanthus and reed canary. In principle they avoid the time-scale issues that forestry based biomass presents, in that they tend to grow and be cropped on annual cycles. When directly compared with alternative land use options their effective CO_2 per area of land can be low, as long as indirect emissions of GHGs such as N_2O from fertiliser use are avoided. However, the carbon impacts from indirect land use change when biomass crops replace other crops are uncertain and potentially could be quite large requiring further consideration. There are also potentially substantial implications for biodiversity which are not covered here but were, for example, addressed in a recent review for the South Eastern US region.[35] They do not currently form a large part of the power production or the domestic heating market but could have potential for application in CHP or boiler applications. In Europe for example, solid biomass energy crops covered about 50,000 – 60,000 ha of land in 2007 which is significantly lower than the 2.5 million ha used for energy crops grown for the production of transportation biofuels such as cereals and rape.[36] A range of other second and third generation biomass feedstocks exist which include straw, bagasse, jatropha, waste vegetable oil, municipal solid waste and algae. Many of these may be processed to liquid biofuels rather than having large global usage as a direct source of energy and thus are not the focus of this review. Excellent reviews on the production of liquid biofuels from 1st to 3rd generation sources can be found in Nigam and Singh[37] and Fivga et al.[38]

2.6 Peat

Peat is a low calorific fuel that is still used in both domestic heat and electricity generation sectors. It is sometimes considered a “slowly renewable energy” and thus is mentioned briefly here. It is, however, classified along with solid fossil fuels rather than biomass fuels by the IPCC since its emissions are considered to be comparable to coal. Peatlands in their natural state act as a sink for atmospheric CO_2, and the carbon content of the world's peatlands represents around a third of all terrestrial soil organic carbon, despite covering only 3% of the earth's surface.[39] However, whilst pristine peatlands are generally a net carbon sink, they can also be a significant source of methane and there are significant uncertainties in estimating the carbon storage in peatlands under current and future climate scenarios.[40] Disturbed peatlands (e.g. through drainage for forestry, agriculture and peat removal) can alter the balance of CO_2 vs. CH_4 emissions but it is suggested that subsequent to the harvesting of peat for energy on a peatland, the remaining cutaway persists as an atmospheric source of CO_2 as the residual peat continues to decompose. The extraction of pristine peatlands for energy (as has been the case in Ireland) rather than drained peatlands, leads to the highest net GHG emissions from energy production; greater than the use of coal per unit of energy produced. A life cycle impact assessment of peat use for energy generation in Ireland, including processes such as peatland drainage and industrial extraction, transportation, combustion, and subsequent use of the cut-away land[41] found that the combustion phase contributed over 95 % of the total life cycle GHG emissions. To a certain extent, the emissions could be mitigated by the restoration of the cutaway areas, through for example, afforestation of cutaway peatlands.[42] However, there can also be considerable losses of soil carbon from residual peat that continues to decompose.[41,42] Time-scales again are critical, since the return of the land to a carbon sequestration state may take many years. A further controversial aspect of peatlands in tropical regions of Southeast Asia, is that they are being cleared to create land for industrial palm oil and pulpwood plantations, a bi-product of which could be used as forestry wastes for biomass energy production. The IPCC estimates that tropical peatland cultivation generates the equivalent of 440 million tonnes of CO_2 per year.[43]

Overall, the literature suggests that appropriate methods of carbon accounting using a common set of assumptions and system boundaries[32] will be a vital part of using biomass effectively, both as a renewable energy resource and within NET strategies. In particular, issues such as land-use change, definition of waste products, harvest rates, timescales of forest residue decay and carbon sequestration as well as soil based carbon should be considered.[32] Counterfactual models have been suggested as a means of developing more realistic net GHG changes for particular utilisation strategies.

3. AIR QUALITY STANDARDS AND ATTAINMENT WITHIN EUROPE

The AQ impacts of biomass combustion must be considered when defining effective strategies for its future use. This must include assessing the current state of AQ in a particular country or region and whether the future use of biomass will impact on that region's ability to meet AQ standards. Many nations have ambient standards for several of the pollutants that can result from biomass combustion. Whilst it is not possible in such a short review to consider the AQ standards in all countries, Europe is covered here as an example of an industrialised region with a well-established framework for monitoring AQ and the attainment of ambient concentration standards. WHO standards are also addressed since often they provide more stringent targets than national limits, and should in principle underpin long term health based targets for all countries.

Nitrogen oxides (NO + NO_2) are acidic gases and ozone precursors and can affect human health and vegetation. NO_2 is thought to have both acute and chronic effects on airways and lung function, particularly in people with asthma. As a result, both hourly and annual standards have been set within Europe for NO_2 to protect human health and vegetation within the Directive on Ambient Air Quality and Cleaner Air for Europe (2008/50/EC). An hourly standard of 200 μgm^{-3} should not be exceeded more than 18 times per calendar year and an annual standard of 40 μgm^{-3} has been set for the protection of human health, consistent with WHO guidelines. The EU standard for ozone is 120 μgm^{-3} as a maximum daily 8 hour mean which is slightly higher than that set by the WHO of 100 μgm^{-3}.

Air quality standards for particulates within Europe are based on controlling concentrations of PM_{10} to below 50 μgm^{-3} for a running 24-hour average, with 35 annual exceedences permitted per year. There is also an annual standard of 40 μgm^{-3}. The term PM_{10} refers to the mass of particles that pass through a size-selective orifice with a 50% collection efficiency cut-off at 10 µm aerodynamic diameter. This metric was chosen for the development of air quality and emissions standards as it best represents those particles most likely to reach the lung acinus.[44] An annual limit value of 25 μgm^{-3} has also been set for $PM_{2.5}$ although this is significantly higher than the annual mean target set by the WHO of 10 μgm^{-3}. In terms of impact, a distinction is made between PM_{10} which are termed “thoracic” particles, $PM_{2.5}$ termed “respirable” particles that can penetrate into the gas-exchange region of the lung, and ultrafine particles which are smaller than 100 nm. Exposure reduction targets have also been set within Europe for $PM_{2.5}$ based on a percentage reduction from the average exposure indicator (AEI) in 2010, although no standards have yet been set for exposure to ultrafine particles.

A variety of legislation and technological developments have led to reductions in emissions of major pollutants and therefore improvements in AQ in Europe. NOx emissions decreased by 44% and PM_{10} by 24 % between 1990 and 2011. As a result, the percentage of urban citizens exposed to pollutant levels above standards fell between 2000 and 2018. However, poor AQ remains a problem and in 2018 the percentage of citizens exposed to levels above EU limit values was estimated to be 34 % for O_3, 15 % for PM_{10}, 3.8 % for $PM_{2.5}$, 3.6 % for NO_2 and 15-20 % for Benzo-a-Pyrene (BaP).[45] The percentage for PM_{10} has in fact increased in recent years. Hence there remains a challenge for all pollutants, even with respect to EU Directive values. WHO recommendations are more stringent and estimates of exceedances of these run to 73.6 % of the population of Europe for $PM_{2.5}$ and 98.6 % for O_3. Europe is an example of a region with a well-established legal framework which aims to reduce the exposure of its population to harmful pollutants, but which is a long way off reaching the goal of complete compliance with the WHO’s AQ guidelines. This reaffirms the point that any policies chosen for the reduction of GHGs need to consider AQ impacts very carefully.

4. PHYSICAL AND CHEMICAL CHARACTERISTICS OF EMISSIONS FROM BIOMASS COMBUSTION

Emissions from the combustion of biomass within both large scale power generation processes and residential and commercial heating appliances will include a complex mixture of gases and particles. Gaseous emissions will contain differing levels of carbon monoxide (CO) and carbon dioxide (CO_2) depending on the completeness of combustion, NOx from both fuel, and to a lesser extent thermal sources, a wide range of volatile organic compounds (VOCs), sulphur oxides (SOx) and a range of trace species potentially including heavy metals and polyaromatic hydrocarbons (PAHs), furans and dioxins; as well as organic and inorganic aerosol particulates.[46] A schematic diagram summarising the potential variety of emissions is shown in Figure 3.

Wood consists primarily of the polymers cellulose, hemicellulose and lignin. Other biomass fuels such as grasses, and wheat stubble also contain these polymers, although in different relative proportions. Small amounts of low-molecular-weight organic compounds such as resins, waxes, sugars and inorganic salts are also present in wood. During combustion, pyrolysis occurs producing a variety of smaller molecules. Biomass combustion is typically inefficient, and as a result, a variety of partially oxidised organic chemicals are generated in particulate emissions from biomass. PM emissions are perhaps of the greatest concern since, particularly for domestic use of biomass, they are difficult to control for several reasons that will be discussed later in the review. Of the PM produced during wood combustion in an enclosed residential wood burner, 96% was suggested to be in the PM_{10} size fraction and 93% in $PM_{2.5}$ by Houck and Tiegs.[47] A study of PM emissions from a fixed grate domestic stove using a range of raw and torrefied biomass fuels, found over 96% of PM mass to be in the submicron (PM_1) range for all fuels tested.[48] This puts the majority of emissions into the respirable fraction of PM and is also of significance in terms of meeting AQ and exposure standards for $PM_{2.5}$.

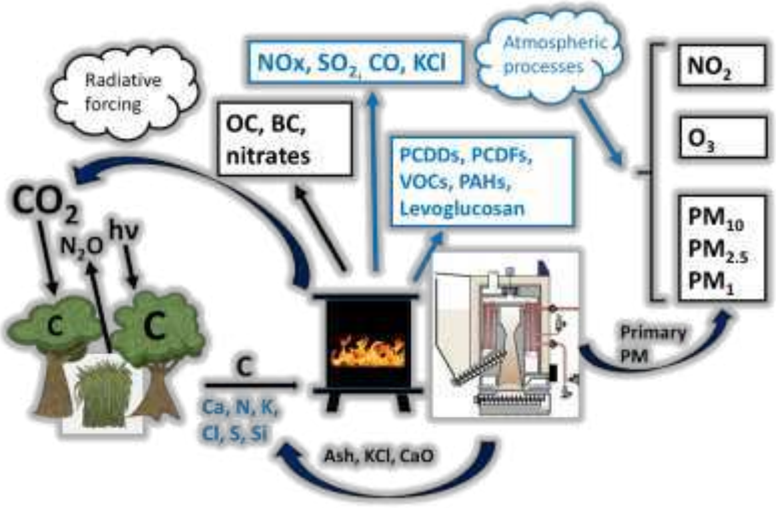


Figure 3. Schematic diagram illustrating the wide variety of emissions that can result from the combustion of biomass within both residential and commercial appliances.

The chemical characteristics and PM size distribution of the emissions are likely to vary depending on the type of appliance, burn rates, fuel moisture content, and the type of biomass used. The chemical components prevalent within each size fraction are also highly variable and may affect their toxicological impacts.[49] Fine et al. suggested that as much as 74% of the $PM_{2.5}$ emitted from domestic fireplaces burning wood was OC and 1-18% was elemental carbon (essentially BC).[50,51] Depending on the wood species burned, salts and minerals present within PM could include Na, Mg, K, Ca, Zn, ammonium, sulphates, chlorides, carbonates and nitrates.[52] Potassium was found to be the most abundant elemental species contributing up to 1.5 wt % of fine particle mass.[51] Its presence in biomass fuels has been found to lead to severe deposit formation, thus causing potential corrosion problems.[53] The organic compounds present in wood smoke will also vary depending on combustion conditions and wood characteristics such as lignin and resin content. McDonald et al., in a study of emissions from a range of wood burning stoves and fireplaces, suggested that most of the aliphatic and olefin hydrocarbon VOC content is made up of C1-C6 compounds.[54] Dienes, especially 1,3-butadiene and 1,3-cyclopentadiene, were also found to be enriched in wood combustion emissions deriving from the decomposition of terpenoids.[55] Significant concentrations of dicarboxylic and diterpenoid acids were found in wood smoke in a study by Rogge et al.[56], particularly from the burning of pine woods. A comprehensive review of the emissions of various aromatic compounds and PAH from a range of different wood-types is given in Fine et al. for various species of wood grown in the Northeastern United States.[51] They found that the pyrolysis product of cellulose (levoglucosan) was by far the most abundant component of the fine particle organic emissions (3%-12%). The high emission rates of levoglucosan and its uniqueness to biomass combustion make it an important candidate as a marker for biomass combustion in general, that can be used in ambient air pollution studies for source apportionment of PM (see section 5). Other compounds include a variety of aldehydes, phenols, alcohols, ketones, carboxylic acids, methane, ethane, ethanol, formaldehyde, acetaldehyde, acrolein, cresol, formic acid and acetic acid.[52,54,57] Dioxins (polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs)) are also present in emissions from wood burning.[58,59] Furans, formed from the decomposition of wood cellulose upon heating, were found in significant quantities from softwood and hardwood emissions by McDonald et al.[54] A comprehensive review of PCDDs and PCDF emissions from biomass burning is provided in Lavric et al.[58]

Mixing between fuel and oxygen within the combustion chamber has a strong role to play in the level of emissions of carbonaceous pollutants, since the formation of soot and PAH may be enhanced in fuel rich zones where incomplete combustion occurs. The conclusion of an extensive review of Williams et al.[46] was that increased biomass use in small units such as for space heating or cooking could lead to ineffective mixing and potential problems arising from emissions of particulates, particularly carbonaceous smoke. They suggest that biomass burned in larger combustion units leads to better mixing thus reducing carbonaceous emissions, which can be additionally removed from the flue gases using a range of different flue gas control options. They do however conclude that there remains a potential health problem from submicron inorganic particles from devices at all scales.

Secondary pollutants formed from gaseous emissions of VOCs from biomass combustion are also of concern. Tropospheric O_3 is formed in the atmosphere from chemical interactions of VOCs, NOx, and a radical source such as OH, which is formed from the photolysis of O_3, aldehydes, hydroperoxides, or nitrous acid (HONO). Biomass burning is a potential source of VOCs, NOx, and HONO and this may contribute to tropospheric ozone formation. However, VOC species are emitted at varying relative ratios depending on the fuel type and burn conditions making it difficult to predict the overall impact of emissions from biomass combustion on O_3 formation.[60] In addition, primary emissions of organic species can go on to form Secondary Organic Aerosol (SOA) which is particulate mass formed in the atmosphere from a complex series of reactions of a large number of organic species resulting in the formation of low volatility and/or high solubility oxidation products that partition to the particle phase. SOA formation from biomass emissions is also highly variable.[60,61]

It is clear that to fully understand the impacts of biomass burning, a wide range of compounds needs to be considered from the point of view of emissions factors for both gas and aerosol phase species, and their impact on atmospheric concentrations of pollutants with potential health risks. These issues will be covered in sections 7 and 5 respectively.

5. IMPACTS OF BIOMASS BURNING ON AIR QUALITY IN DEVELOPED COUNTRIES

As suggested in the previous section, emissions arising from the combustion of biomass are likely to be a significant contributor to atmospheric concentrations of, in particular, NO_2 and $PM_{2.5}$, with domestic burning of wood, pellets, briquettes and peat of particular concern. Identifying the contribution of biomass burning to atmospheric PM_{10} and $PM_{2.5}$ can be approached using source apportionment methods based on measurements at a receptor site (receptor-oriented models), but it is not a straightforward task. Such methods are commonly based on the use of chemical tracers. While there are no absolutely specific chemical tracers for the burning of wood, levoglucosan and water soluble fractions of organic carbon, potassium and rubidium are potential candidates, with the ratio of the concentrations of different tracers used in some studies and principal component analysis (PCA) of multiple tracers used in others.[51,62,63] The ratio between levoglucosan and mannosan concentrations has also been shown to indicate a preference for softwood or hardwood particulates within measured samples.[64] The ratio of levoglucosan to wood smoke mass and the ratio of levoglucosan to potassium concentrations within atmospheric PM can be highly variable, according to the type of wood being burnt and the combustion conditions, which makes the conversion process more difficult.[65,66] A third method for estimating wood smoke concentrations is based on the use of multi-wavelength aethalometers[67] and depends upon the assumption that elemental carbon arises from only road traffic and wood smoke, and that the two sources of black carbon may be differentiated from their Angstrom coefficients.[66] Many studies use more than one approach in order to reduce measurement based uncertainties.

Measurements of the contribution of wood smoke to overall PM concentration are highly variable and depend on season, location and the conversion factors used. Critical reviews of the suitability of specific organic markers and biomass emissions factors are given in Vicente and Alves[68] and Puxbaum et al.[65] A range of values for the conversion from levoglucosan to woodsmoke concentrations have been suggested within the literature from 6.0–23, with lower factors more typical for softwood fires in stoves and fire places, and higher factors typical for hardwood types such as beech.[51,65,69] Bari et al., for example, suggest a higher value of 22.87 for hard woods and a lower value of 10.7 for soft woods.[69] Schmidl et al. on the other hand, suggest relatively similar values for a range common woods growing in mid-European Alpine regions, apart from beech which had a lower value below 5.[70] The use of a conversion factor allows estimates for the contribution of biomass burning to $PM_{2.5}$ and PM_{10} to be made possible, but also means they are subject to uncertainties, which makes the comparison of data between different sites rather tricky. Fuller et al. compared aethalometer and levoglucosan estimates of mean PM across London from wood burning and found them to be in good agreement, with estimates of winter contributions to PM_{10} of 1.8 $\mu g\ m^{-3}$ (levoglucosan) and 2.0 $\mu g\ m^{-3}$ (aethalometer), representing between 7% and 9% of mean PM_{10} across the London transect.[71]

Some authors have suggested that within urban areas the contribution of wood smoke is generally lower than at rural sites, and Caseiro and Oliveira suggested that the lack of difference between urban roadside and urban background levoglucosan concentrations points towards the regional nature of woodsmoke pollution.[64] They estimated that wood burning contributed to about 3.1% of the winter PM_{10} mass in Oporto, and to 3.7% in Copenhagen. Using multi-wavelength aethalometers within an Alpine valley in Switzerland, Sandradewi et al. estimated that 51% of the BC was due to wood burning and 49% due to traffic emissions, suggesting a high contribution of woodsmoke to PM in rural mountain locations.[67]

Vicente and Alves provide a wide ranging review of available studies of outdoor AQ data[68] related to biomass burning in developed countries. They do not specifically suggest contributions to PM concentrations, rather providing a summary of levoglucosan concentrations from different studies. However, using an average conversion factor of 7.35 as suggested in Puxbaum et al.,[65] allows the range of $PM_{2.5}$ concentrations from biomass burning to be estimated. For winter concentrations within European cities these range from 2.8 $\mu g\ m^{-3}$ in Porto up to 7 $\mu g\ m^{-3}$ in Milan. The highest measured urban concentrations presented in the review are around 20 $\mu g\ m^{-3}$ in Fresno and Libby in the United States and in Geeveston, Tasmania. These concentrations are twice the $PM_{2.5}$ exposure levels recommended by the WHO of 10 $\mu g\ m^{-3}$.

Karagulian et al. compiled a source apportionment database for PM in global cities with domestic fuel burning emerging as the main contributor to PM mass concentrations in Africa (34%), and in Central and Eastern Europe (32%), and still a highly significant source in the Americas (25%), Northwestern Europe (22%), the Southern China region (21%), South Eastern Asia (19%), and India (16%).[72] This is consistent with the review of Vicente and Alves within Europe,[68] where accounting for seasonal variation, the percentage contributions of biomass burning to atmospheric PM mass concentrations could be as low as 1 % in the summer, but as high as 50% in winter samples for some regions. Bari et al. suggested that during winter months, as high as 59% of ambient PM_{10} pollution could be attributed to residential wood-fired heating in Stuttgart, Germany.[69] It is clear that elevated fine particulate levels due to biomass burning are a significant problem in developed countries and affect both rural and urban areas.

The reasons for burning biomass domestically can vary, with it being the only source of heating in some homes, to being an occasional secondary source of heat in others. Kalogridis et al., used an absorption based method to assess the contribution of biomass burning to winter BC in Athens.[73] They found that 30% of winter time BC came from biomass combustion in 2013-2014 with a clear diurnal pattern, showing highest concentrations during the night-time, supporting the theory of local domestic heating being the main source. Fuller et al. suggest that an increase in the contribution of biomass burning to PM concentrations over weekends in London indicates that the reasons for burning wood are mainly discretionary.[71] On the other hand, Saffari et al., reported that AQ in the Greek city of Thessaloniki worsened during the recent economic crisis due to residents burning more wood/biomass rather than using the more expensive options of oil and gas.[74] As a result they reported a 30% increase in $PM_{2.5}$ concentrations associated with residential wood combustion in 2012 and 2013 which was concurrent with 30–40% lower concentrations of V and Ni, associated with the combustion of residential fuel oil and industrial activity. In Denmark, a doubling of wood stoves and boilers over a ten-year period was partially attributed to increased fossil fuel costs.[75] However, a recent survey by the UK Department for Environment, Food and Rural Affairs (DEFRA) provides a quantification of different solid fuel use in UK domestic settings from April 2018-March 2019, based on data collected by the market research group Kantar.[76] Emissions inventories estimate that domestic wood burning in the UK contributed 38% of primary emissions of $PM_{2.5}$ in 2019, with emissions from this source having more than doubled since 2003.[77] This is consistent with the 40% suggested for Denmark.[75] The Kantar study found that just 8% of people in the UK burned solid fuels indoors, with two-thirds of them living in urban areas. One third had open fires as opposed to modern stoves, and 96% had alternative sources of heating such as gas or electricity. This suggests that, at least in the UK, discretionary burning as suggested by Fuller et al.[71] is prevalent.

Sample location can be an issue in terms of obtaining an accurate picture of the contribution of biomass burning to PM mass in urban areas. Typically, long term sample networks have been designed on the basis that traffic emissions are likely to dominate, and are often therefore at roadside locations or at urban background sites. Fewer sample sites tend to be located in residential areas away from major roads, which could lead to a poor representation of domestic sources of emissions within available data sets. In a hybrid approach, Larson et al. used a combination of fixed and mobile nephelometer monitoring, coupled with a spatial buffering procedure to estimate the spatial patterns of woodsmoke in and around Vancouver during 19 cold, clear winter evenings from November, 2004 to March, 2005.[78] Measurements at the fixed monitoring site were correlated with the measured average levoglucosan concentrations at the same locations producing an R^2 of 0.66, thus to a certain extent validating the mobile nephelometer data. The use of this type of hybrid approach allows for better spatial coverage of the city, allowing the identification of regions with relatively high woodsmoke levels within an urban airshed. Such "hotspots" were found in residential neighbourhoods, and thus in areas that would not normally be monitored using spatially fixed sites. This is illustrated in Figure 4, reproduced from ref.,[78] and showing large regions of high woodsmoke concentrations which are at substantial distances from the $PM_{2.5}$ regulatory monitoring sites. In some residential regions, the two-week average $PM_{2.5}$ concentrations well exceeded the 10 μgm^{-3} recommended by the WHO.[78] The study suggests that were the WHO limit values for $PM_{2.5}$ to be adopted by different countries, that monitoring strategies suitable for assessing attainment may need to be developed.

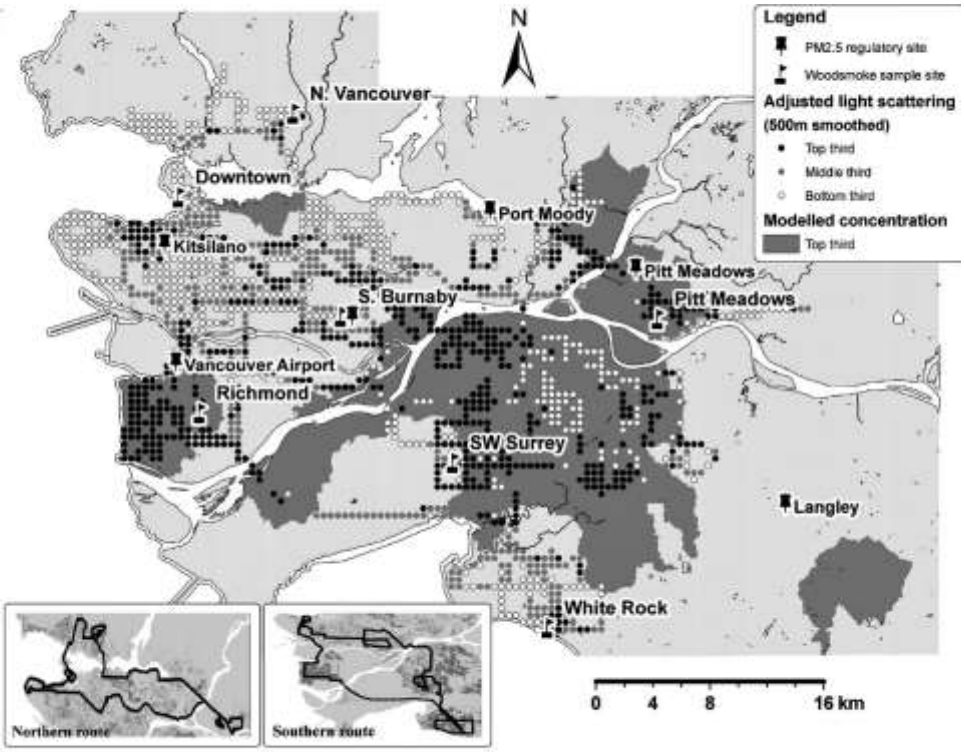


Figure 4. Adjusted light scattering values from measurements in the Greater Vancouver Regional District (GVRD). The locations of the regulatory $PM_{2.5}$ samplers, the fixed filter-based sampling sites, and the two major GVRD mobile sampling routes are also shown. The dark shaded regions show the highest woodsmoke levels as predicted by the "best" model. Reproduced from Larson, T.; Su, J.; Baribeau, A.-M.; Buzzelli, M.; Setton, E.; Brauer, M. A Spatial Model of Urban Winter Woodsmoke Concentrations, Environ. Sci. Technol*,* 2007, 41 (7), 2429–2436. Copyright [2007] American Chemical Society.

Clearly, woodsmoke concentrations can vary over quite short distances. Allen and Rector conducted a rural study in the Adirondacks of NY.[79] They found that increases in concentrations were mainly driven by local sources, as demonstrated by the lack of correlation of the paired sites used in the study. One location 40 metres from a residence with a wood stove, experienced repeated episodes of elevated $PM_{2.5}$ concentrations, with a maximum 3-hour average of 150 μgm^{-3}, and a maximum 24-hour rolling average of 64 μgm^{-3}, despite being in compliance with the United States Environmental Protection Agency (US EPA) National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$. This highlights both the localised and episodic nature of elevated PM from domestic wood burning.

Another important feature to note about available studies is that those based on chemical tracers tend to be short term in nature, perhaps being gathered during a specific measurement campaign. The use of chemical tracers and the analytical burden that they pose, makes it difficult to make spatially detailed analyses of the long term trends in PM from biomass burning. A recent report suggested that increased coverage of aethalometer networks could enhance our ability to track long term changes in PM from biomass burning, since they can be run in a more automatic way and in general have reasonable correlations with chemical tracer methods.[80] Several examples of long term trend analysis of atmospheric particulates using aethalometers exist in the literature.[81–83] Such studies have facilitated the separation of the effects of localised emissions from wood burning from particulate episodes which occurred due to long range transport.[81,83] They have also provided data on the diurnal and seasonal variability in ratios of BC to OC, which is important for understanding the optical properties of carbonaceous aerosols, thus potentially reducing uncertainties within climate models.[84,85] From a policy perspective, availability of long term trend data is incredibly useful, since the impacts from wood burning on ambient AQ are likely to be less important for breaches of specific concentration targets, but will have greater effects with respect to $PM_{2.5}$ exposure reduction targets within for example, EU AQ directives. Long term aethalometer data can be used to determine the time dependence of source contributions to carbonaceous PM.[86] Monitoring long term trends that can be linked to sources, is necessary to tackle those that could impact most on exposure reduction. So far, the WHO annual mean guidelines for $PM_{2.5}$ have not yet been adopted as AQ targets within Europe or the US. If they are in the future, then better spatial coverage of $PM_{2.5}$ measurements will be required to assess the attainment of these targets, which will be challenging if there are continued increases in domestic wood burning.

Estimating the contributions of biomass burning to atmospheric gaseous pollutants such as NO_2 has to be tackled in a different way since there is no means of identifying the source of individual NO_2 molecules from atmospheric sampling. However, receptor modelling can be carried out by looking at both receptor and source profiles for multiple species within a Positive Matrix Factorisation (PMF) based approach.[87] In such an approach, source types are identified by comparing them to measured profiles providing fingerprints, and source weightings are used to determine how much each source contributed to a measured atmospheric concentration sample. Liu et al. for example, applied a PMF approach to estimate source contributions to a range of pollutants, including NO_2, in the Chinese Mega City of Tianjin.[87] The biomass factor was identified by higher loadings of K, OC and BC and by means of this footprint, the contribution of biomass burning to atmospheric concentrations of a range of pollutants including NO_2 could be determined. The performance and uncertainty of PMF and other receptor models for source apportionment was reviewed by Belis et al.[88] who concluded that the accuracy of receptor models is sufficient for the needs of air quality management. PMF approaches can be combined with trajectory modelling based approaches (source-oriented models) in order to identify the geographical source regions that contribute to pollutant concentrations at the selected measurement sites.[89] This type of approach is more commonly used to evaluate the long range transport of emissions from agricultural residue burning and outdoor fires rather than more localised sources such as domestic wood burning.[90,91]

A limited number of studies have examined the effects of stove usage on levels of indoor air pollution under real world usage in developed countries.[92–95] Semmens et al. studied $PM_{2.5}$ and particle number concentrations and infiltration in 96 homes in Northwestern US and Alaska that were using older model wood stoves as the primary source of heating.[93] During two forty-eight hour sampling periods they found daily average $PM_{2.5}$ concentrations of around 29 μgm^{-3} suggesting significant infiltration of PM with average indoor $PM_{2.5}$ concentrations exceeding WHO ambient AQ guidelines and approaching the US EPA 24-h standard equivalent (25 μgm^{-3} annual average). Piccardo et al. found installation and operational issues impacted on indoor concentrations and that following education, indoor exposures could be reduced.[92] Allen et al. also examined the impact of stove upgrades at 15 homes in northern British Columbia, Canada over a 6-day sampling period, but they did not find a consistent relationship between stove technology upgrades and indoor AQ improvements in homes where stoves were exchanged.[95] A recent 30 day study within the UK city of Sheffield, showed that even modern UK DEFRA-certified residential stoves can lead to short term peaks in exposure to PM_1 and $PM_{2.5}$ within the indoor environment.[93,94] These transient peaks in $PM_{2.5}$ could reach up to hourly indoor means of 47.60 μgm^{-3} and were linked to refuelling frequency and length of burn period, indicating that opening of the stove door was a primary mechanism for introducing PM into the room. These short term exposures to high concentrations may be significant from a health perspective and the systematic review of Orellano et al. found a positive relationship between short term PM exposure and cardiovascular, respiratory, and cerebrovascular mortality.[96]

6. HEALTH IMPACTS OF PM FROM BIOMASS BURNING

It is clear from these studies that particulates from biomass burning are likely to impact on human health, although separating out the health effects of particulates from biomass burning compared to other pollutant sources within epidemiological studies, is even more difficult than obtaining spatially and temporally detailed concentration profiles. As Naeher et al. point out, exposure to woodsmoke is as old as humanity itself, but the notion that woodsmoke, being a natural substance, must be benign to humans is far from the truth.[57] Based on epidemiological data, Sigsgaard et al. estimate that the contribution of biomass smoke to premature mortality in Europe amounts to at least 40 000 deaths per year as well as negatively affecting respiratory and, possibly, cardiovascular health.[10] However, they do highlight the difficulties in assessing overall population risk which relies on the use of risk coefficients from large scale meta-analyses involving a range of particle concentration metrics based on PM mass (PM_{10}, $PM_{2.5}$), particle number or absorption based BC concentrations. Gu et al. suggest that estimated premature deaths are approximately ten times greater when using a BC-specific risk coefficient compared to using $PM_{2.5}$ based coefficients, and as a result estimate 171,000 premature mortalities in China attributable on an annual basis due to exposure to rural residential BC, mainly from the domestic combustion of solid fuels.[97] However, significant uncertainties remain in determining composition based risk factors.

As pointed out by Zelikoff et al., a range of health effects can result from different pollutants emitted from biomass burning at levels measured within indoor environments of households using wood fires and stoves.[98] CO has been shown to produce carboxyhemoglobin and to increase the incidence of angina among people with cardiac disease. NO_2 can bind to haemoglobin, affecting several enzyme systems, causing vascular membrane injury and edema, and may produce bronchoconstriction in asthmatics at low levels. PAHs may be immunosuppressive and carcinogenic.[99] Aldehydes are associated with upper airway irritation, headaches and other neurophysiologic dysfunctions, exacerbation of bronchial asthma, and possibly cancer. PM is perhaps the component of most concern within emissions from biomass burning, since it is relatively much higher than when combusting gaseous fuels for cooking and heating, and potentially has a range of effects due to both particle size and chemical composition.

There have been few controlled human exposure studies to PM within woodsmoke but that of Sällsten et al., found evidence of woodsmoke-associated systemic inflammatory effects in study participants.[100] Most of the available evidence on health effects in humans is epidemiological in nature, particularly documenting respiratory effects in children. Zelikoff et al., summarises that exposure of preschool children living in homes heated with wood burning stoves or open fireplaces causes a range of effects including decreased pulmonary lung function in young asthmatics and increased incidence and severity of acute bronchitis and acute respiratory infections.[98] Wider epidemiological studies reviewed in Boman et al.[101] and Naeher et al.[57] have evidenced the link between short-term exposures to PM from wood smoke and adverse health effects such as asthma, respiratory symptoms, daily mortality, and lung function. Boman et al. in particular, compared the relative risks of wood smoke with general estimations for ambient PM and adverse health effects, and found that they were even stronger in studies where residential wood combustion was considered a major source of PM. Evidence of associations with cardiac events are suggested to be weaker than for impacts on respiratory symptoms.[57] A possible weakness of wider epidemiological studies, is the lack of information on personal exposures. Many epidemiological studies are based on ambient concentration measurements of total $PM_{2.5}$ or PM_{10}, and as suggested above, these are often from fixed monitoring sites which might not be representative of human exposure, particularly when people move between different exposure environments during their daily routine and spend large amounts of time in indoor environments.

The conclusions from epidemiological studies are however, supported by the outcomes of toxicological studies. Many toxicological studies on animals have used acute single-exposure regimes, since they are often carried out to support investigations of the potential impact of smoke on fire fighters or smoke inhalation victims from fires. Such studies, as summarised by Naeher et al.[57], identified the irritant effects of woodsmoke through the generation of oxidative stress producing an inflammatory response. In some cases, acute lung injury and pulmonary lesions were found that are typically seen in smoke injured victims from fires. Of most relevance to ambient exposures however, are more prolonged exposure studies at lower concentrations and these are much scarcer within the literature. However, an acute exposure study by Fick et al. on rabbits showed that woodsmoke had the potential to alter pulmonary immune defence mechanisms potentially creating increased susceptibility to infectious lung disease.[102]

Lal et al. reported from a long term exposure study on rats, that pulmonary lesions were progressive with repeated smoke exposures.[103] Zelikoff et al., following several 1 h per day exposures of rats to smoke concentrations more typical of concentrations found indoors during the operation of a poorly vented fireplaces, found suppression of bacterial clearance from the lungs.[98] This was particularly related to particle concentrations, although they did not find evidence of lung pathology or lung cell damage.[98] This suggests that prolonged exposure to woodsmoke may compromise pulmonary immune mechanisms that are important for protection against infectious lung pathogens. Longer term studies on rats of up to 6 months exposure to smoke from an uncertified wood stove burning mixed oak species found that effects on general indicators of toxicity, bacterial clearance, cardiac function, and carcinogenic potential were mild. [104] They were however, similar to those found in diesel exposure studies and included increased platelet number, reduced liver weight and increased spleen weight.[104] Animal studies have suggested that whilst lung tumour prevalence in mice exposed to woodsmoke was higher than in the control group, woodsmoke is a weaker carcinogen compared to smoke from coal burning which was coincident with lower concentrations of BaP in woodsmoke compared to that from coal.[105]

A number of in vitro studies as summarised in Naeher et al. indicate the generation of reactive oxygen species (ROS) as a result of exposure to woodsmoke, which are known to be linked to the progression of inflammatory disorders and potentially pulmonary fibrosis.[57] Several studies also indicated that woodsmoke extracts were mutagenic in bacterial systems.[106] Correlation analysis carried out by Saffari et al., using $PM_{2.5}$ samples obtained during a winter sampling campaign in Thessaloniki, also indicated a strong association between ROS activity in rat alveolar macrophage cells and the PM bound concentrations of levoglucosan, galactosan, and potassium, highlighting the potential impact of wood smoke on PM-induced toxicity.[74]

7. EMISSIONS FACTORS AND MITIGATION STRATEGIES FOR BIOMASS COMBUSTORS: ISSUES OF SCALE AND BIOMASS COMPOSITION

7.1 Fuel Pre-Treatment Methods for Biomass Resources

Biomass resources available for energy production are diverse in their physical and chemical characteristics with direct consequences on air pollutant emissions when combusted. Clean woody biomass with relatively low amounts of moisture and ash would be favoured over potentially variable lower grade fuels which may have higher N or ash fractions, high moisture content, be larger in terms of fuel particle size or contain unwanted trace components such as heavy metals.[107] The pretreatment of solid raw biomass can be used to upgrade the properties of the fuels prior to use in combustion systems. Methods include washing, drying, sieving, leaching, densification technologies such as biomass chipping, pelleting and briquetting, torrefaction, and hydrothermal carbonisation (HTC), or wet torrefaction. During torrefaction, the biomass is heated to 225-275˚C driving off water and semi-volatiles which can be used as a fuel to drive the process. Torrefaction also contributes to structural changes in the fuel through breaking hemicellulose, lignin, and cellulose chains at elevated temperatures.[108] The result is a high energy density fuel with up to 50% mass loss, but with little total energy loss. Wet torrefaction can be used to process low-grade, chipped, biomass feedstocks using pressurised steam, which after rapid decompression, results in breaking apart biomass material, with the released moisture removing part of alkaline and chlorine components from the fuel. Following pelletisation, the resulting fuel is hydrophobic, with high energy density. Leaching with water can also reduce fuel K and chloride concentrations by up to 80% and 90% respectively. Pretreatment methods can therefore help to diversify potential bioenergy resources to lower grades of biomass, which may help to increase the use of waste derived products, broadening the resource base, as well as providing better defined fuel specifications to assist with the design and management of clean combustion devices. Excellent reviews on pretreatment methods can be found in refs[109,110] and the IEA Policy report.[107]

Pretreatment methods are not however, without impacts on AQ. A number of studies have shown there to be the potential for VOC, CO and CO_2 emissions from the storage of torrefied fuels, which needs to be carefully managed from an AQ and safety perspective during transport and storage.[111,112] Composition, and thus potential toxicity of the VOC mix, has been shown to depend on a variety of factors such as treatment methods and temperatures, moisture content and storage time.[112] Pelletisation is also not without impacts on AQ and could potentially be a major source of airborne VOCs and PM unless adequate controls are implemented. Drax were recently served with a $2.5 million fine for emitting three times the regulated limit for VOCs from a pellet plant in Mississippi, indicating the difficulties in ensuring that effective control equipment has been regulated and installed.[113] A recent study suggests that in the South Eastern US, pellet plants are significantly more likely to be placed within vulnerable and economically depressed communities[114] and often have not been subjected to appropriate permit regimes.[115]

Pretreated fuels can be potentially used in all scales of biomass combustion. Pelletised and briquetted fuels are more likely to be utilised in medium to large scale systems, or potentially even domestic scale boilers where automatic fuel feeding systems are part of the operation principle. As an example, the total consumption of wood pellets in the EU in 2015 has been estimated as 20.3 million metric tons, of which 36% was used for power generation and the rest to meet heating demand.[32] Pellet baskets also allow the conversion of fireplaces or wood-burning stoves for use with pellets. For manually fuelled systems such as wood stoves and cook stoves, pretreated fuels can also be manufactured into firelogs or briquettes to be used in place of traditional wood logs. These can be made from a variety of waste starting materials including: paraffin wax and sawdust; woodchips; agricultural residues such as olive stones, ground nutshells and seeds, bagasse, corn cob, palm fibre and cotton plants; cardboard; and coffee grounds. The physical and chemical characteristics of such manufactured logs will clearly differ from traditional wood logs and will influence their combustion characteristics and emissions.[116] There have been some limited studies on emissions profiles from the use of such manufactured fuels that will be briefly covered in the following sub-sections.

7.2 Emissions Factors and Abatement Methods for Larger Scale Devices

The available technologies for the utilisation of solid biomass for energy and heat provision include pyrolysis, gasification, and combustion. In future scenarios these could be coupled with CCS in order to provide NETs as discussed in section 2.3. Among these, combustion is the most commonly used technology for heat and power production. Gasification is also of interest for future applications because of its potential to reduce emissions of relevance to AQ, particularly PM, in comparison to conventional combustion. Biomass combustion systems are available from a few kW up to several 100 MW.

Unabated, biomass combustion systems are likely to exhibit relatively high emissions of NOx and particulates in comparison to furnaces using natural gas or light fuel oil.[117] As a result they can contribute significantly to atmospheric PM, as well as to ozone, and NO_2 formation in the ambient air via secondary atmospheric processes. An LCA indicated that 38.6% of the environmental impact of a modern automatic wood furnace is attributed to NOx and 36.5% to PM_{10}.[117] Hence effective abatement technologies are required to reduce AQ impacts, although the technology readiness and cost effectiveness of such technologies vary substantially for different systems. The scale of appliance, as well as whether it is operated automatically or manually, have potentially the largest impact on the level of emissions of the major pollutants for reasons explained in this section. Table 1 summarises the different scales of appliances with their operation principle, as well as the legislative regime in Europe that specifies limits on their air pollutant emissions.

Type of Appliance	Energy Output	Operation principle	Power	Legislative regime
Wood stove	Heat	Manual	2-15 kW	EDD
Log wood boiler	Heat	Manual	5-50 kW	EDD
Pellet stove or boiler	Heat	Partially automatic	2-25 kW	EDD
Commercial boiler	Heat or CHP	Automatic (e.g. moving grate, understoker)	Up to 20 MW	EDD < 1 MW MCPD > 1 MW
Large Combustion Plant	Power	Automatic (e.g. stationary/bubbling or circulating fluidised bed, entrained flow)		MCPD < 50 MW_{th} IED > 50 MW_{th}

Table 1. Different scales and modes of operation of appliances as well as the legislative regime in Europe that specifies limits on air pollutant emissions. EDD (Ecodesign Directive), MCPD (Medium Combustion Plant Directive), IED (Industrial Emissions Directive).

Biomass combustion is a complex process involving both heterogeneous and homogeneous reactions. The main steps are drying, devolatilisation, gasification, char combustion, and gas phase oxidation.[117] Pollutant formation from biomass combustion devices occurs for a variety of reasons. Poor mixing and thus incomplete combustion can lead to high emissions of pollutants such as CO, volatile organic compounds (VOC), soot, condensable organic compounds or tar, and PAHs. A variety of technologies have been developed for improved mixing, particularly in large scale devices, which have evolved from those developed for the combustion of other solid fuels like coal and coke.[118] Fuel pulverisation can be used to help ensure complete burnout of the fuel in within short residence times of a few seconds, and is typically used in large power stations, requiring similar particle sizes to coal of less than 1 mm in size. Fluidised bed systems are able to utilise larger pelletised or chipped fuels with the aim of improving the contact of the fuel particles with air in comparison to packed bed systems. This reduces the processing requirement compared to pulverised fuels and may facilitate the use of more fibrous types of biomass. Both pulverisation and fluidisation can help to reduce the products of incomplete combustion, and when coupled with flue gas clean up technologies can facilitate achievement of the stringent emission limits for large scale devices set out within the Industrial Emissions Directive (IED, 2010/75/EU) and the Medium Combustion Plant Directive (MCD, 2015/2193). As we move down the scale to smaller devices, the combustion systems are increasingly likely to become manually operated and to utilise larger fuel units such as logs and briquettes. This affects mixing and therefore the level of unburnt products produced by the system.

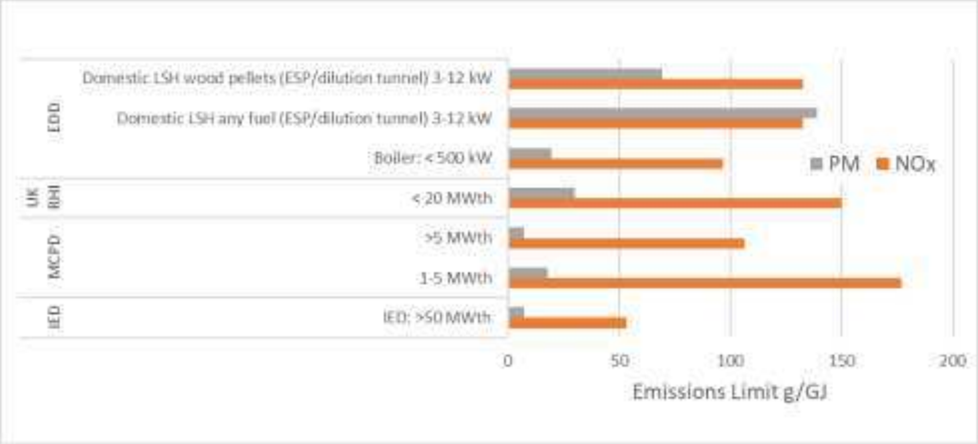


Figure 5. Comparison of European PM and NO_x emissions limits for different scales of combustion devices based on an equivalent energy basis (g/GJ) net heat input. The chosen test method within the EDD is the ESP/Dilution tunnel method.[119] LSH – Local Space Heater. Conversions are based on an assumed stoichiometric dry flue gas volume of 253 m^3/GJ net heat input adjusted for reference oxygen content and a fuel CV for dry wood of 17.3 GJ/tonne (dry basis) based on the Digest of UK energy statistics from 2014.[120]

The European regulated emissions levels for PM and NOx for different scales of combustion device are summarised in Figure 5. For PM there is about a factor of 20 difference between the limits for Local Space Heaters (LSH) controlled by the EDD, compared to large combustion plant which is controlled under the IED, when considered on an equivalent energy input basis. On an equivalent fuel input basis then, the utilisation of biomass within large scale units will lead to substantially lower AQ impacts than its use at the domestic scale. The residence times and improved mixing possible in larger combustion units allow the burnout of the carbonaceous part of the particulates and hence prior to flue gas clean up particulates from larger units will mainly consist of submicron particles of inorganic material[118] formed as a result of fuel constituents such as N, K, Cl, Ca, Na, Mg, P, S. Sippula et al. showed that $PM_{1.0}$ mass concentration correlated linearly with ash content and composition of the fuels studied, particularly for the major PM forming elements K, Na, Cl, S.[121] Nussbaumer states that native wood is usually the preferred biomass source due to low contents of ash and nitrogen, whereas herbaceous biomass such as straw, miscanthus, switch grass, etc., have higher contents of inorganics such as N, S, K, Cl, leading to higher emissions of NOx and particulates, increased ash, corrosion, and deposits.[117] For large combustion plant, submicron inorganic particles may still remain a challenge for pollution controls since particle control methods such as cyclones, electrostatic precipitators and baghouse/fabric filters tend to have lower efficiencies for smaller particles.[118] Ceramic filters may have potential for removing smaller size fractions and have high thermal durability, although in smaller devices could cause issues related to pressure drop.[122,123]

7.3 Emissions Factors and Abatement Methods for Medium Scale Plant and CHP

Medium combustion plant and large pellet boilers also have low limits on PM emissions controlled in Europe by the MCPD and EDD, necessitating the use of advanced combustion systems and potentially flue gas clean-up methods. For larger boilers, emissions can be partially controlled by the design of the combustor, for example using two-stage combustion with injection of secondary air prior to a hot combustion chamber. Electronic combustion control and forced ventilation is also recommended in order to ensure optimum operation in practice, and to enable good mixing with less dependence on ambient climate conditions such as air temperature and external air flows.[124] Smaller appliances are less likely to use automatic systems, and although under typical operations they can achieve lower PM emissions than manual stoves, variations in operational conditions due to variable loads may lead to poor combustion conditions and consequently higher PM emissions than for automatic systems.[123] For example, Poláčik et al. compared an automatic boiler fed by spruce pellets with a manual stove, fed by beech logs, and found fine particle emissions (in the size range of 17–544 nm) of 173 mg kg^{-1} and 1043 mg kg^{-1} respectively during a similar test cycle.[125] Lower grade pellets may also have higher ash content and the review of Rabaçal and Costa concludes that PM emissions from pellet boilers are strongly correlated with the ash content and ash composition.[123] In the UK, the Renewable Heat Incentive (RHI) scheme has supported the expansion of both commercial and domestic biomass boilers by providing financial support at a set rate per unit of renewable heat produced per kWh to the owner of the heating system, for a period of seven years. RHI systems are subject to emissions limits as shown in Figure 5, and fuel must be sourced from an approved supplier which should in theory provide some consistency in fuel quality. This is important as several studies have shown significant variation in PM emissions and size fractions for different pellet types with an example shown in Figure 6.[126]

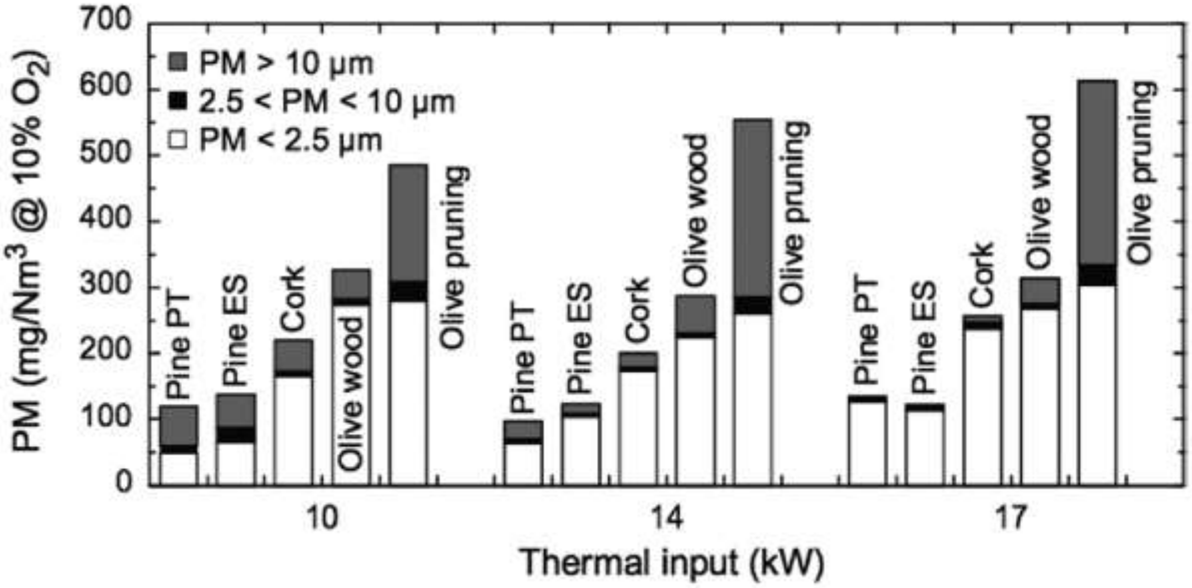


Figure 6. PM emissions of the different types of pellets for all operating conditions from a domestic pellet-fired boiler[126]. Reprinted from Fuel, 119*,* Garcia-Maraver, A.; Zamorano, M.; Fernandes, U.; Rabaçal, M.; Costa, M., Relationship between Fuel Quality and Gaseous and Particulate Matter Emissions in a Domestic Pellet-Fired Boiler, 141–152, Copyright (2014), with permission from Elsevier.

NOx emissions from different biomass sources and combustion devices can also vary greatly, depending on many factors including the N content of the fuel and combustion temperature. However, the variation with scale of combustor is much less severe for NOx than for PM as illustrated in the closer agreement between NOx emissions limits shown in Figure 5. Air staging and fuel staging have been developed as primary measures for NOx reduction, particularly within automatic systems, and can offer potentials of 50% - 80% reduction.[117] If further mitigation is required, post combustion technologies are available such selective non-catalytic reduction (SNCR) and selective catalytic reduction (SCR) which rely on the reaction $NO + NH_2 \rightarrow N_2 + H_2O$ with urea or ammonia injected as a source of NH_2. At the smaller scale, downdraft boilers with inverse flow have also been introduced which apply the two-stage combustion principle used in larger systems.[117] In a review of abatement technologies for biomass systems, Nussbaumer concludes that effective operation at optimum excess air and maximum efficiency is best achieved with advanced process control requiring the application of sensors for λ (air–fuel equivalence ratio), temperature, and unburnt substances across all device scales.[117]

CHP using biomass sources is one potential means of providing low carbon electricity and heat through, for example, the further development of heat networks. However, the overall impact of CHP on AQ may be complex. On the one hand, CHP provided using large scale boilers would provide overall efficiencies greater than those of conventional power generation through combustion, and could replace domestic scale heat provision by local boilers, potentially leading to lower overall emissions of pollutants. However, this could be combined with local increases in key pollutants such as NO_x and PM, since CHP is also more likely to be located in regions of high population compared to conventional power plant, due to the need to supply urban heat networks. The overall uncertainties in providing projections in potential AQ benefits or dis-benefits of an expansion in CHP from biomass are large, as pointed out by Apsimon and Oxley.[127] Guidance planning from Environmental Protection UK encourages the use of larger plant, for example in conjunction with the development of heat networks, rather than a larger number of small plant, since abatement technologies become more cost effective for larger plant sizes and are currently more highly developed for larger systems than for smaller community and domestic scale boilers. The impact and cost implications of a range of control measures for biomass boilers ranging from SCR of NO_x, to cyclones and ceramic filters for PM reduction, were discussed in a review of the AQ impacts biomass combustion by the UK Air Quality Expert Group (AQEG). Whilst there are small CO_2 penalties involved in the use of such mitigation technologies and a range of potential costs involved, the potential for reductions in AQ impacts may be significant as highlighted in Table 2. Relative costs per unit energy benefit from efficiencies of scale for larger devices. High efficiency methods such as ceramic filters may merit the costs involved for new installations in areas with existing poor AQ, suggesting more localised policies on emissions limits may be useful, as part of planning or permit applications.[124]

Proposed control technology	Estimated reduction efficiency %	Indicative emissions factor g/GJ	Estimated annualised costs as a % of initial

			investment where stated
Selective non catalytic reduction (SNCR) for NOx	40	70	
Selective catalytic reduction (SCR) for NOx (>1 MWth)	80	23	
Multi-cyclone	50	17	1-2%
Ceramic filter	99	2	< 10%

Table 2. Effectiveness of post combustion emissions reduction strategies for solid biomass boilers (adapted from AQEG)[119]

Gasification and pyrolysis techniques perhaps offer the best potential to lower particulate emissions from biomass utilisation since they are based on converting solid fuels into more uniform gaseous (or liquid) fuels, thus improving mixing and lowering the potential for particle formation. Currently such technologies are largely used in plant > 2 MWth. Further developments are required at smaller scales and a sustainable route for the utilisation of the remaining char material should be developed. A recent study by Pröll and Zerobin[128] compared several methods for biomass utilisation in a CHP plant with a fuel power input of 66 MW_{LHV}. These included fluidised bed and chemical looping combustion connected to a steam cycle CHP plant, and rotary kiln slow pyrolysis of biomass for biochar soil storage with direct combustion of volatiles supplying the steam cycle CHP plant: all with and without CO_2 capture. The purpose of this work was not to study AQ related pollutants but to compare overall efficiencies and CO_2 emissions per unit energy, against the least CO_2-intensive fossil fuel, namely natural gas. The study shows however, that low emission technologies do exist at this scale and furthermore, the study makes recommendations for global climate change mitigation strategies, concluding that both BECCS and the use of biochar for soil enhancement may be effective NETs when coupled with CCS. The decision on which technological path to follow will depend on the type of available biomass, energy markets, and ecosystem demands. Further studies covering wider aspects of sustainability including AQ impacts would be useful.

7.4 Emissions Factors for Domestic Scale Devices

In 2017, 43% of solid biomass energy in the EU28 was used in the residential sector.[129] Small domestic boilers and local space heaters (LSH) such as stoves and open fireplaces have significantly higher regulated limits than for larger plant, driven by the lack of cost effective pollution control methods at this scale (Figure 5). For LSH, the PM emissions limits within the Ecodesign Directive (EDD, 2009/125/EC) are 20 times higher than those in the IED for large plant when compared on the basis of equivalent net heat input[119] (~7 g/GJ for large combustion plant vs. ~140 g/GJ for a modern, clean domestic stove as shown in Figure 5). The short residence time and poor mixing of the fuel and air in small units leads to significantly higher emission rates of particulates than for larger systems where combustion technologies such as fuel pulverisation and fluidised beds may be utilised along with flue gas control methods.[46] From an AQ perspective therefore, small devices such as domestic stoves and boilers are expected to pose a larger problem than large combustion plant.

Mitchell et al. compared NOx and PM emissions from the combustion of a range of biomass fuels with coal and smokeless coal in a fixed bed 5.7 kW stove, that in this case was not designed to meet the EDD limits, but does allow for a comparison between fuels.[130] Cycle average NOx emissions ranged from 85 gGJ^{-1} for torrefied wood up to 259 and 438 gGJ^{-1} for smokeless fuel and peat briquettes respectively. Peat was the worst of the fuels in all respects. The fuel-N content of the latter two fuels was shown to be significantly higher than for the biomass derived fuels, with fuel-N shown to be more important than combustion temperature and residence time for NOx emissions in this case (Figure 7). The phase of combustion was also shown to be of importance with flaming combustion producing higher NOx emissions than smouldering combustion within the overall cycle. Ozgen et al., recently provided an overview of NOx emissions from biomass combustion for domestic heat production, including a useful summary of emissions factors found in the literature which are shown to be highly variable. They also suggest a strong relationship between NOx emissions and fuel-N content, although the conversion efficiency is suggested to exponentially decline with increasing fuel-N.[131]

The Mitchell study showed that flaming combustion also produced significantly higher emissions of PM than the smouldering phase, although the difference was reduced in the case of ‘smokeless’ fuels including torrefied wood.[130] Emissions factors for $PM_{2.5}$ ranged from 32 gGJ^{-1} for torrefied briquettes up to 90 gGJ^{-1} for commercially available seasoned mixed hardwood and 210 gGJ^{-1} for peat briquettes when using a heated filter based measurement method. Fuel type as well as the morphology of the fuel are clearly critical, and relationships were found between average total particulate emissions and volatile matter within the fuels. The surface area of the fuel particles determines the rate of initial devolatilisation as well as the progress of the flame front and combustion of the char formed.[130] These influence the burning rate and consequently the temperature in the combustion chamber impacting on emissions.

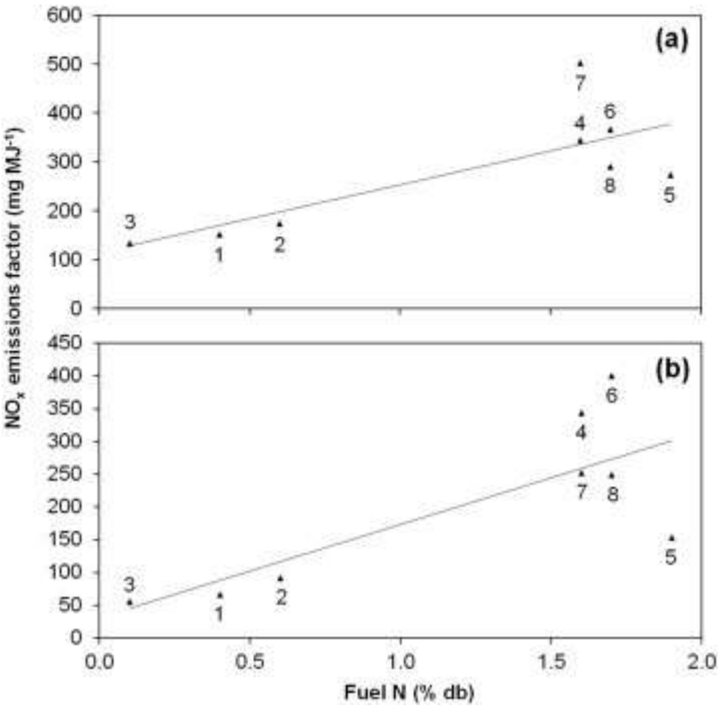


Figure 7. NO_x emissions for various fuels plotted against fuel-N content for (a) flaming, and (b) smouldering phases. Fuel specifications: 1. Commercially available seasoned mixed hardwood; 2. Air dried hardwood (silver birch); 3. Torrefied spruce wood (bark-free) briquettes; 4. Peat briquettes; 5. Bituminous coal; 6. Biomass/coal blend; 7. Low smoke anthracite; 8. Smokeless fuel. Reproduced under the terms of the Creative Commons CC-BY license from Mitchell at al.[130]

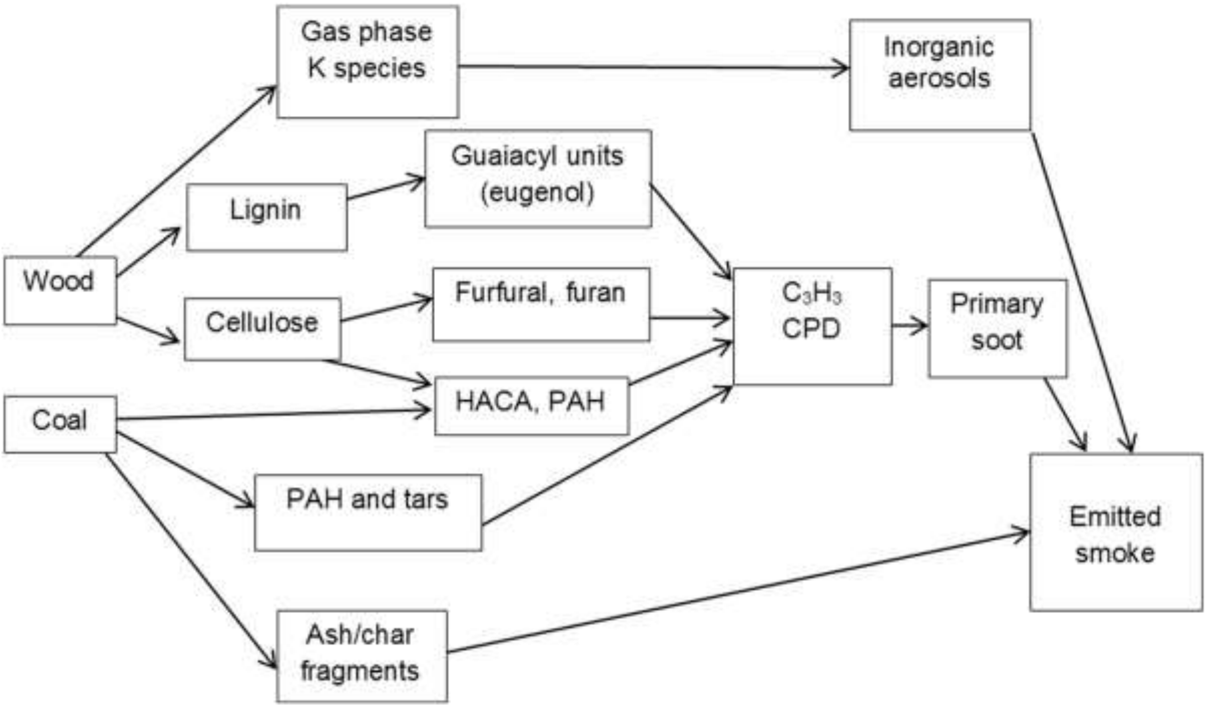


Figure 8. The routes to the formation of smoke from biomass or coal, or a blend. Reproduced under the terms of the Creative Commons CC-BY license from Mitchell at al.[130]

Several studies have linked the footprint of pyrolysis products from biomass sources with mechanisms for soot formation. Decomposition products, such as furfural and eugenol which are formed from cellulose and lignin decomposition are highly sooting since they can contribute to the main routes of soot formation, namely the HACA (hydrogen abstraction, carbon addition) mechanism which dominates in the case of furfural, and via cyclopentadienyl formation and reaction in the case of eugenol (Figure 8).[48,132] Carbonaceous particles can agglomerate to form chains together with inorganic aerosol such as KCl and fragments of char.[130,132] A recent study by Du et al. suggested that that emission factors of condensed organic compounds in a domestic stove increased with increasing KCl content for KCl impregnated fuels due to increased heterogeneous condensation of semi-volatile hydrocarbons on pre-existing ash particles.[133] Elsasser et al. found that the organic fraction of PM emitted during the ignition phase was the main contributor to overall emissions in their logwood stove study.[134] Torvela et al. found that impaired combustion conditions increased the level of condensable organics within the PM in a 40 kW grate combustion unit with PM_1 emissions of up to 160 gGJ^{-1}.[135] Pettersson, also reported significantly higher emissions of total PM for a log burning stove under conditions of poor air supply and high firing intensity. PM was dominated by fine particles (200-320 nm) and emissions factors varied in the range 38−350 g/GJ^{-1}. Trubetskaya et al. recently studied PM emissions factors from the combustion of a range of fuel types in both a conventional and an EDD compliant wood stove.[108] They found that the use of secondary air could reduce PM from wood logs by about two-thirds due to improved mixing of fresh air with preliminary combustion products. Vincente and Alves[68] reviewed a wide range of studies to compare derived PM emission factors from biomass combustion based on different measurement techniques. The values for fireplaces ranged from 161-1610 gGJ^{-1} and from 20 up to 1433 gGJ^{-1} from woodstoves, indicating an improvement for stoves over open fires. Pellet boilers were again an improvement over domestic stoves (6-377 gGJ^{-1}).

All of these studies highlight the huge variability of emissions factors depending on fuel type/quality, and appliance design and operational practice. This also suggests that real world emissions may deviate from those obtained under operating conditions during test cycles obtained for compliance and research purposes. Scott, for example,[136] compared emissions from the same appliances under "real world" conditions as simulated within a laboratory, and under real world conditions where the wood burners were operated normally by the householder using their own firewood supply. Their results suggested that real world emissions may be up to 4-5 times those achieved within legislated test cycles and laboratory "real world" studies. Coulson et al. suggested that in situ studies which take account of the "operator" variable are essential for understanding real world emissions of manually controlled devices.[137] They presented a review of such in situ studies for New Zealand which highlighted the variability in emissions factors, and showed that only 22% of stoves met their legislated limits in the real world. Wood moisture, flue temperature and oxygen supply were the major factors affecting emissions rate variability. Fachinger et al., conducted a real world comparison study of a residential pellet stove and a wood stove using 11 hardwood species. They suggested that real world emissions factors from pellet stoves can be expected to be closer to laboratory-derived emission factors than those for wood stoves, due to lower dependence on user operation.[138] Typical ranges of emissions factors from average to best to worst as reported by Nussbaumer are shown in Figure 9 for smaller devices such as pellet boilers, pellet stoves, log wood boilers, and wood stoves. It is clear that domestic stoves present the highest emissions factors, often reaching several hundred g/GJ and far in exceedance of the EDD limits. Such stoves have long lifespans and are likely to remain in operation for many years unless schemes incentivising replacement are offered.

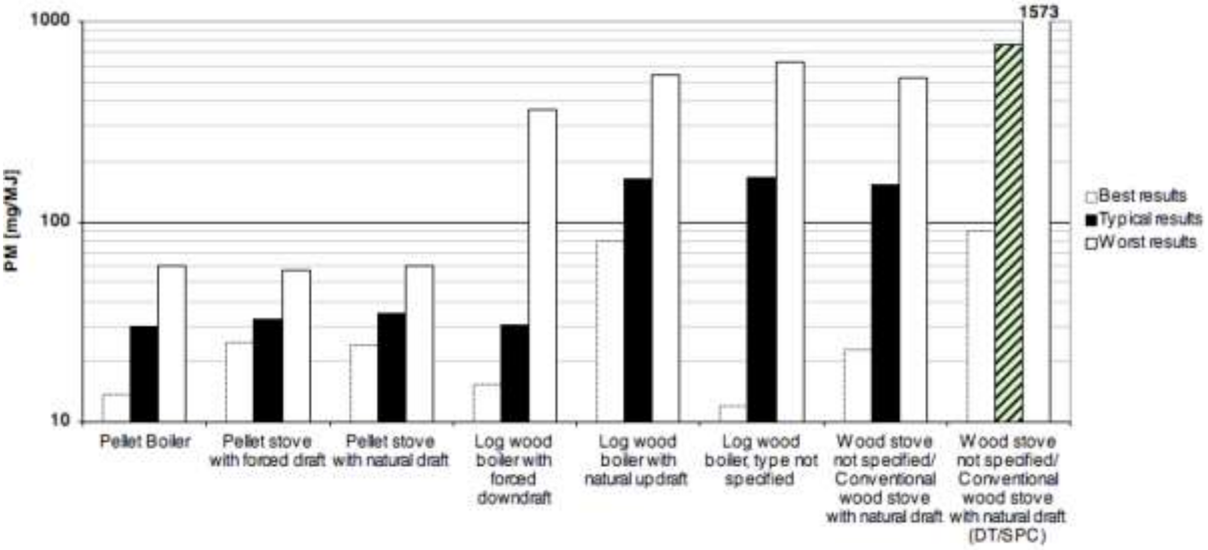


Figure 9. Average emission factors (best, typical, worst) for pellet boilers, pellet stoves, log wood boilers, and wood stoves. All data indicated as solid particles except for wood stoves, where data including condensables (SPC) or measured in dilution tunnel are given in the last column. Reproduced from Nussbaumer et al., Overview on Technologies for Biomass Combustion and Emission Levels of Particulate Matter, Proceedings of the 16th European Biomass Conference & Exhibition, Vol16, OA 9.2, Copyright (2008) with permission from ETA Florence Renewable Energies.[124]

Nussbaumer suggests that wood stoves can be developed with designs that provide a hot combustion chamber with good mixing. However, stoves are partially designed to provide visible flames in order to achieve the “homely” feel desired by the customer. Operation without forced ventilation therefore gives limited possibilities for improved combustion principles. He suggests that designs should be developed that prevent improper operation of the stove such as over-fuelling and the possibility to reduce the air inlet.[124] Elsasser et al. for example, showed that an overloaded stove emitted roughly 4 times the mass of PM compared to a stove operated according to the manufacturer’s recommended conditions.[134] The same stove also showed much higher emissions factors for PAHs for over fuelled operation.

7.5 Influence of Fuel Quality

Fuel quality has also been shown to influence PM emissions. Emissions of PM and to a lesser extent NOx from a fixed bed 5.7 kW stove with a single combustion chamber were found to be lower from the combustion of torrefied fuels compared to raw biomass from spruce, willow and olive stone sources by Maxwell et al[48]. However, even for the torrefied fuels, NOx emissions ranged from 45-90 g/GJ. Moisture content has been shown to particularly influence PM emissions with freshly cut, high moisture woods giving higher PM emissions than kiln dried woods (small logs) for the soft wood spruce within a study of a 5.7 kW domestic stove.[139] Kiln dried fuels were shown to lead to higher combustion temperatures and rates of combustion, particularly during the early phases of combustion. However, oxygen supply was also shown to be critical, and insufficient air supply led to high PM emissions for logs of the kiln dried hard wood beech. Calvo et al. also found higher PM emissions from hard woods (eucalyptus), compared to pine soft woods.[140]

There have been relatively few emissions studies of manufactured fuel logs and briquettes in domestic scale devices. However, Mitchell et al. recently compared emissions from briquetted bagasse, miscanthus, wheat and barley straw with low moisture wood logs and wood briquettes. PM emissions factors for the bagasse and miscanthus briquettes were found to be lower than those for the straws and comparable to the wood briquettes. The study of Trubetskaya et al. compared PM emissions of torrefied olive stone briquettes, smokeless coal, sod peat, wood logs and bituminous coal.[108] The torrified olive stones and smokeless coal gave lower emissions factors than the other fuels but the differences were dependent on the test method used (hot filter sampling of the raw flue gas vs. sampling of cooled, diluted flue gas). A large scale study of spatially resolved emissions factors for a region in rural China suggested slightly lower $PM_{2.5}$ emissions per Kg of fuel burned from processed biomass pellets than from uncompressed straws, although overall the emissions factors for biomass were slightly higher than those for coal.[141]

The UK implemented a “Ready to Burn” certification scheme in May 2021 through the Air Quality (Domestic Solid Fuels Standards) (England) Regulations 2020, which bans the sale of wood with a moisture content of > 20% in amounts of two cubic metres or more. France is also planning a similar labelling scheme certifying wood of low moisture wood and sustainable forestry origins.[142] The US EPA manages the Burn Wise program with the aim of providing information to the public on how to store and dry wood, as well as lists of EPA certified appliances and appropriate installation and maintenance regimes.[143] Schemes such as these, and the penetration of stoves built to EDD/EPA specifications may partially offset the increase in stove sales that has occurred over the last decade. However, as appliances are slowly replaced by newer EDD compliant versions, it will be important to conduct real world emissions studies to establish whether they meet regulatory limits during every day usage and for typical ranges of fuel types used.

7.6 Abatement Methods for Small Scale Devices

Strategies for the mitigation of emissions from small scale devices are focussed in two main areas: i) improvement of the combustion process and/or fuel quality improvement, or ii) flue gas cleaning technologies. Primary measures address issues around air supply through, for example, staged combustion, or by pre-heating the primary combustion air.[68] Fuel improvement by processing as discussed above, or through the use of additives can also be used to reduce alkali compounds, decreasing the formation of inorganic particles.[144] Several options exist for flue gas removal such as electrostatic precipitators (ESPs), condensing scrubbers[145] and catalytic converters.

The difficulties of controlling PM from domestic scale biomass combustors were highlighted in a report prepared as part of the IEA Bioenergy project.[146] The report compares efficiencies of particle collection devices for residential biomass systems (<50 kW) including ESPs, catalytic converters, ceramic filters and condensing heat exchangers. The scarcity of available PM control systems at the domestic scale that can be retro-fitted is of particular concern. The challenge is technically more difficult than, for example, the use of diesel particle filters, due to the fact that small scale domestic appliances tend to rely on natural draughts for air supply. Pressure drops can be induced by flue inserts such as catalyst carriers which can affect primary combustion conditions and thus influence emissions.[147] IEA Bioenergy Task32 found no promising results from catalytic converters for wood boilers and stoves. The high flue gas temperatures required for catalytic oxidation, mean that these devices are typically not operating effectively during start-up where the highest emissions often occur. They can also suffer from fouling and clogging due to tar and soot emissions and hence cleaning regimes would be vitally important.[68] The pressure drop of ceramic filters or catalytic converters was found to negatively influence the combustion behaviour of natural draught systems. A recent study by Klauser et al. did however, see an effect of flue gas catalytic removal in reducing emissions of high molecular weight PAHs from burning beech wood in a domestic room heater, albeit at the expense of slightly increased total PM emissions.[148] It was postulated that the catalysts might have led to oxidative formation of semi-volatile species, thus contributing to overall PM emissions. Wöhler et al. compared the use of a foam ceramic element, a catalytic active coated foam ceramic element, and a honeycomb catalyst for the potential reduction of emissions of CO, PM and OC from a domestic wood stove.[149] The honeycomb catalyst showed the highest potential of systems tested, leading to up to a 33 % reduction in PM emissions, although the effectiveness of all devices was found to be lower in the start-up phase of the stove. Reichert et al. recently demonstrated that the use of honeycomb catalysts could be effective at achieving emissions rates stipulated for wood stoves in the EDD [147] under real-life related testing conditions. Hence more recent studies are starting to show some promising results for the use of catalysts, but the literature remains inconsistent and further studies are required.

IEA Bioenergy Task32 found ESPs to be the most promising technologies for boilers, with collection efficiencies for PM of up to 85%. Costs can be reduced by utilising a metal chimney tube as the collecting electrode, but particle removal would need to be done manually by the operator or a chimney sweeper for most of the examples considered, and hence efficiencies will depend on maintenance regimes. Most of the devices considered, seemed to be effective for the removal of PM_1 which is promising in terms of reducing respiratory effects of PM. However, where there are high emissions of condensables, the aerosol load can be higher at the filter outlet compared to the inlet, and the use of ESPs may therefore alter the PM composition[68] potentially changing the toxicity of the emitted particles.[150] At an estimated capital cost of 1000 to 3000 €, ESPs would approximately double the price of a domestic stove installation which may impact on voluntary uptake. More recent examples of ESPs (e.g. Exodraft[151]) offer chimney integrated ESP particle filters with a chimney fan, and are starting to reach near market status. These have the aim of ensuring a constant up-draught in the chimney, thus improving air supplies and preventing the backflow of smoke into the room. Automatic particle removal is also claimed which would potentially reduce the influence of poor maintenance regimes.

Without such mitigation options, domestic stoves are likely to continue to make a substantial contribution to particulate emissions in both urban and rural areas and research into cost effective retro-fit PM control systems at this scale is greatly needed. Stricter emissions limits tend to accelerate technological development and would encourage the market introduction of, for example, particle precipitation or catalytic devices. A common international approach regarding PM emission measurements is also urgently needed to assess the comparative efficiency of control technologies, particularly in real world settings.[146]

7.7 PM Composition Based Emissions Factors

As discussed in section 4, PM emissions from biomass burning are compositionally complex including a range of organic and inorganic species such as heavy metals, PAHs, dioxins and furans. There is a substantial literature which shows that the combustion of biomass, and wood in particular, is a rich source of PAHs, both in the gas phase and bound to PM, which may arise via pyrosynthesis. PAHs are of particular concern because of the known carcinogenicity of the mixture. McDonald et al.[54] derived an emissions estimate for total PAH emission rates from wood stoves as 74.7 mg/kg fuel. Predominant species were found to be acenapthylene, naphthalene, anthracene, phenanthrene, benzo(a)pyrene and benzo(e)pyrene. Rogge et al. found that the smoke from the burning of synthetic logs had the highest total PAH emission rate and the most diverse range of PAHs present when compared to burning pine and oak.[56] The review of Vicente and Alves[68] and the detailed wood stove study of Pettersson et al. using birch, spruce and pine logs, provide a detailed discussion and emissions estimates for PAHs respectively.[152] One striking conclusion of Pettersson's study is the high variability of emissions factors for PAH, with values ranging from 1.3 to 220 $mgMJ^{-1}{}_{fuel}$. In most cases the mass of PAH was dominated by phenantrene, fluoranthene, and pyrene, and mostly found in the particle phase.[152] Factors influencing the level of PAH emissions included wood type, and the same types of operation conditions that also affected overall PM emissions such as oxygen availability and burn rates.

The review of Lavric et al. provides a summary of emissions of dioxins (PCDDs) and furans (PCDFs),[58] concluding that as complete as possible combustion is critical for a low emission level of dioxins, mainly depending on combustion chamber temperature[146], turbulent burning rates, residence time and excess oxygen. Zhang et al. showed that combustion mode: either flaming or smouldering had a considerable influence on the amounts of dioxins emitted from the system.[59] An analysis of soot scrapings from the chimneys of wood-burning stoves operating in various regions of the United States showed that average total PCDD/F levels in the chimney deposits were 8.3 ng/kg in the eastern region, 42.1 ng/kg in the central region, and 10 ng/kg in the west[153] with differences attributed to the wide variability in device design affecting sampling points and/or possible contamination of the fuel wood. The burning of waste wood found in urban areas has been highlighted as leading to significantly higher levels of dioxins in the fly ash from a range of combustors, likely due to the use of treated, varnished or PVC-coated wood.[58] Tame et al. reviewed the role of preservatives in the formation of dioxins from the domestic combustion of wood. They found that preservatives such as copper could influence dioxin formation via acting as catalysts, as well as promoting the smouldering of wood char following the cessation of flaming, proving the low temperature environment needed for dioxin formation. Chlorinated organics, added as secondary preservative components, were also found to yield dioxin precursors on thermal decomposition.[154] The disposal of preservative impregnated timber via combustion, through for example domestic bonfires, is clearly hazardous, even when arsenic is not present.

The ratio of Black Carbon (BC) to Organic Carbon (OC) within total carbon (TC) emissions is also important in relation to the relative impacts on climate forcing, since BC is a major warmer, but OC aerosol is thought to have a cooling effect on the climate as discussed in section 2.1.[16] Mitchell et al. showed that the BC/TC ratio varied during the combustion cycle for a 5.7 kW domestic stove burning a variety of biomass fuels. A higher ratio of ~0.4 was found during the flaming mode for typical wood logs, with a lower value during smouldering combustion.[130,155] They also point out the importance of measurement technique for determining this ratio, since measurement temperature clearly affects the collection of semi-volatile organics. Fine et al. found that the BC content of fine particle emissions for woods burned in the NE US, ranged between 3-7% although, white pine and paper birch produced much higher ratios due to high sap and bark content respectively.[51] Fernandes et al. found that the BC content of PM from an energy-efficient "chimney type" logwood stove was substantially higher than from a conventional cast iron stove and fireplace, with the opposite observed for the OC fraction.[156] Sun et al. present BC and OC emissions factors for a range of biomass fuels for PM emissions from mini-boiler stoves under close to real world operation, finding a dominance of OC, but variability between fuel types.[157] Clearly there is a large variability in BC/TC ratios of PM from different biomass sources and appliances which would affect their climate impacts.

Variability in emissions of the GHG methane was also noted in the simulated real world study of several residential heating biomass appliances (6-11 kW) by Ozgen and Casarini.[158] Using a GWP based method, they compared the CO_2 equivalent emissions of methane with those of N_2O, CO, NOx, VOC, and BC emitted by the same appliances, and showed that the warming impact of methane was lower than that of BC and CO, but was still an important fraction of the CO_2 avoided by the substitution of fossil fuels with biomass. Importantly, they concluded that within a 20-year time frame, the $CO_{2\ eq}$ for all the non-CO_2 forcers offset the CO_2 benefits of biomass use.[158]

7.8 VOC Emissions Factors

As discussed in section 4, VOC emissions can also affect the formation of secondary smog components such as O_3 and NO_2 as well as leading to the formation of SOA. Sun et al., based on a study of traditional and clean domestic heating appliances in northwest China, suggest that the compounds with the greatest contribution to O_3 formation tend to be alkenes, and to SOA, aromatics.[159] They found significant differences in emission profiles between coal and biomass, with wood giving the highest mass based emissions of non-methane VOCs (NMVOCs) and anthracite the lowest. Volatile matter content and combustion efficiency of the appliances were the two biggest factors affecting NMVOC emissions factors. Poor air supply and poor mixing were prevalent in traditional stoves and significant improvements were seen for new devices. NMVOCs yielded by traditional stoves were five times higher than that from semi-gasifier stoves. Pettersson et al. found that emissions for NMVOCs varied from 17 to 2300 $mgMJ^{-1}{}_{fuel}$ in their domestic stove study, with the highest concentrations being of ethene and acetylene generally followed by benzene, propene, ethane, and toluene.[152] Emissions were highest during intensive flaming combustion. Emissions for 1,3-butadiene, of relevance to AQ target compliance due to being an identified carcinogen, varied in the range of 0.1−61 mg/MJ and were again highest in the intensive combustion phase. McDonald et al. also found elevated emissions of 1,3-butadiene and 1,3-cyclopentadiene, for wood combustion relative to other sources and suggested that they probably arise from the decomposition of terpenoids.[54] McDonald et al. carried out a highly detailed study of both particle and gaseous phase emissions from the combustion of US softwoods, hardwoods, and synthetic logs in fireplaces and hardwoods burned in a wood stove, providing emissions data for over 350 elements, inorganic compounds, and organic compounds.[54] They found high emission rates for alcohols, particularly methanol, alkenes, C2-C9 carbonyls and aromatics. Evtyugina et al., in their study of VOC emissions from the combustion of European woods in fireplaces and wood stoves, found aromatics to be the most abundant group, followed by oxygenated compounds and aliphatic hydrocarbons. The relative emissions factors for individual VOCs were found to be strongly affected by the wood composition, the type of burning device and operating conditions.[160] Many previous studies of solid fuel combustion have focussed on VOC emissions from coal as a fuel source and more information is needed on VOC emissions from biomass, particularly since emissions profiles seem to be very variable depending on fuel type and combustor design and operation.

8. ISSUES OF FUEL SWITCHING

From an AQ perspective, the impact of a new biomass burning device installation will very much depend on what it replaces. Figure 10 shows emissions factors for the domestic combustion of various fuels used within the 2018 version of the UK National Atmospheric Emissions Inventory (NAEI). The PM emissions for wood and peat are particularly high, and are representative for the types of stoves prevalent in the UK for this year i.e. in advance of the adoption of limits set in the EDD. The values are estimates, but are used here to illustrate the potential influence of fuel or device switching. If a modern EDD compliant device is replacing an open fireplace or an old stove for the delivery of the same heat, then improvements in emissions, particularly for PM are likely to result. Comparison of the emissions factors for wood in Figure 10 with those from the EDD in Figure 5 suggest at least a halving of PM emissions. Similarly, switching from peat to wood as a fuel would have large benefits for PM emissions. However, in many cases wood stoves and/or domestic boilers could be replacing gas fires or gas/oil boilers as a source of secondary heat, and in this case there are likely to be dis-benefits for both NOx and PM. A comparison of NOx emissions factors within the literature for various biomass fuels compared to fossil fuel alternatives shown in Figure 11 also reinforces this point, particularly for non-woody biomass.[131] This recent review of NOx emissions also highlights that not all of the tested appliances in the literature meet the EDD regulation limit, i.e. that there will continue to be a problem with legacy devices, unless suitable retro-fit mitigation options are developed.

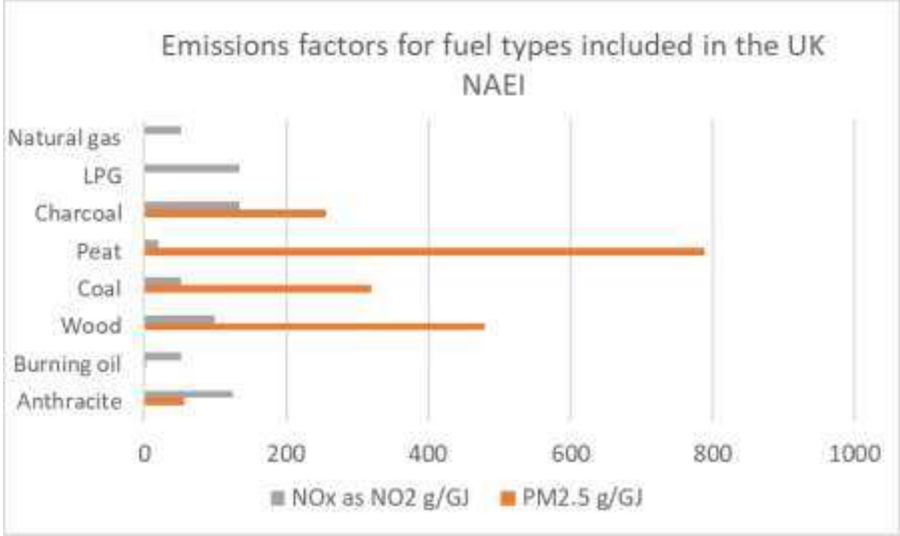


Figure 10. Emissions factors expressed in g/GJ (net) for different fuel types as included in the 2018 National Atmospheric Emissions Inventory (NAEI) for the UK. Note, although the majority of NOx mass emissions are likely to be NO, since NO is subsequently converted to NO_2 in the atmosphere, they are often reported as NO_2 as is the case in the NAEI.

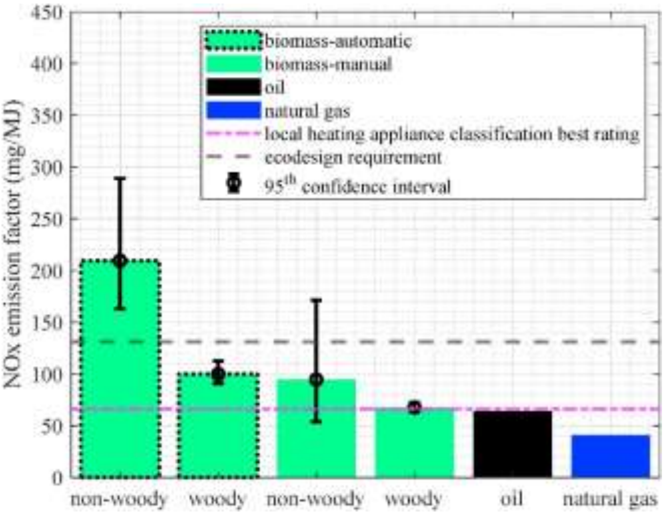


Figure 11. Comparison of the literature residential heating experimental emission factors for biomass combustion and common fossil fuels.[131] Reprinted from Renewable and Sustainable Energy

Reviews, 135*,* Ozgen, S.; Cernuschi, S.; Caserini, S.*,* An overview of nitrogen oxides emissions from biomass combustion for domestic heat production, 110113, Copyright (2021), with permission from Elsevier.

9. IMPACTS ON INDOOR AIR QUALITY AND HEALTH IN DEVELOPING COUNTRIES

9.1 Indoor Concentrations

It is perhaps not surprising that most technologies and strategies proposed for the reduction of GHG emissions are aimed at the industrialised or rapidly industrialising world. However, tackling problems related to the domestic use of solid fuels in developing countries could provide significant benefits to both health-related air pollution problems, as well as climate change. It was estimated in 2000 that approximately 50% of the global population, mostly in developing countries, relied on coal and biomass in the form of wood, dung and crop residues for domestic energy.[14] The IEA suggests some progress since 2000, with the number of people in developing countries with access to clean cooking (LPG, natural gas and electricity), growing by 60% compared to 2000. [161] However, they state that a third of the world's population – 2.5 billion people in mainly sub-Saharan Africa and Asia – still rely on the use of solid biomass for cooking. Data from the 2017 IIASA Gains model suggests that anthropogenic BC emissions within the African continent are dominated by biomass usage for cooking (>50 % of the total). Within Asia, a mixture of biomass cook stoves and coal stoves used for cooking and heating dominate the BC emissions profile. Much of this usage is likely to involve fuels being burnt in simple, often open stoves, although they can vary greatly in terms of design and construction material.[162] Traditional cooking systems are based on burning coal or firewood on stoves with cooking pots placed on top, resulting in low thermal efficiencies and therefore excessive fuel consumption and high emissions.[163] They are usually naturally ventilated, without scope for controlling air supply, leading to incomplete combustion in unvented indoor environments. As a consequence, women and young children are exposed to high levels of indoor air pollution on a daily basis. Although uncertain, estimates suggest that exposure to indoor air pollution may be responsible for nearly 2 million excess deaths in developing countries. Causes of death include COPD, tuberculosis, stroke, heart disease, respiratory infections, pneumonia and lung cancer.[14,164–166] The same studies estimate that over 3% of the global burden of disease can be attributed to indoor solid fuel burning, a level comparable with tobacco smoke and higher than the 1% attributed to urban outdoor pollution.[57] The WHO estimate that close to half of deaths due to pneumonia among children under 5 years of age, are caused by PM inhaled from household air pollution.[165]

Indoor exposures from cook stove emissions can far exceed even heavily trafficked outdoor environments. A study of the impacts of traditional stove usage on lung functions in adult women of a rural Indian village, found average indoor levels of $PM_{2.5}$ as high as 728.90 ± 50.20 μgm^{-3} from the use of biomass compared with 99.76 ± 41.80 when using LPG.[167] This led to respiratory symptoms such as wheezing and chronic cough being significantly more common in the biomass group compared to the LPG control group. Concentrations of up to 2000 μgm^{-3} were found during cooking in biomass-using households in studies of rural households of southern India[168] and in Mexico.[169] Measurements of indoor AQ in 23 houses in Costa Rica[170] using wood burning stoves indicated that average daily PM_{10} levels reached up to 132 μgm^{-3} (c.f. European standard of 50 μgm^{-3}). Peak PM_{10} levels during cooking reached 18,900 μgm^{-3}. The effects of such extreme levels of short term exposure are not well understood. However, some studies suggest that high numbers of particles may overload the lung macrophage system and thus short terms peaks may be as important as lower exposures over longer time periods.[44] Ventilation conditions were found to be the most significant factor determining exposure by Nayek and Padhy[171] implying that huge benefits to global health could be achieved by improvements in stove technology and building ventilation. Jaakkola and Jaakkola suggest that sustainable development calls for interventions that replace traditional biomass fuels with more processed, cleaner fuels, and that improve stoves, ventilation, and housing conditions in an affordable way that is easy to implement.[172] In the context of the Paris Agreement however, cleaner fuels should not be fossil based going forward.

9.2 Mitigation Measures in Developing Countries

Within developing countries, particularly in rural environments, domestic fuel burning for cooking and heating is likely to remain a significant environmental challenge that must be addressed in order to improve public health. Reductions in air pollution-related disease burden (both for household and outdoor) is being used to monitor the progress towards attaining the Sustainable Development Goal on Health (SDG 3)[165] and mitigation strategies are vitally needed. In addition, ensuring universal access to clean fuel and technologies is a target of the Sustainable Development Goal on energy (SDG 7). Whilst electric based cooking might be the cleanest alternative, it is rarely used in the developing world due to the high costs of electricity and limited access in rural areas. Hence, where biomass fuel sources are to be used, research priorities should focus on vastly improving both efficiency and AQ impacts. Large scale penetration of efficient small scale technologies for the effective burning of biomass fuels is therefore required with potential benefits to health and GHG equivalent emissions such as BC. Bhattacharya and Salam for example, suggest that the use of wood stoves with improved efficiency could reduce GHG emissions by at least a factor of two over traditional stove technologies[162] as well as relieving pressure on vital biomass fuel resources.

The Clean Cooking Alliance was established in 2010 and works across a global network of partners with the aim of making clean cooking accessible to millions of households.[173] Simple mitigation measures such as encouraging a more open ventilation environment and using forced draught stoves or fan assisted stoves[174] with mixed biomass solid fuel sources can lead to improvements in indoor levels of $PM_{2.5}$ compared to traditional stoves in more enclosed spaces, with 20–80% reductions in concentrations measured in a rural Indian study.[175] The use of pellet stoves, as oppose to burning logs and unprocessed biomass, has also been shown to lead to improvements in indoor AQ, although a study in Chile suggested that social barriers to such interventions exist, even when financial burdens are removed.[176] Switching to gaseous fuels would also lead to substantial reductions in PM as indicated in the study of Pathak et al. mentioned above.[167] However, LPG is a petroleum derived fuel and thus a poor choice from a climate perspective. The use of biogas rather than solid fuels has been suggested as a possible route for rural communities. This could be based on micro-gasification technologies at the individual stove level,[163,177] on anaerobic digestion or on community scale biogas production which has become relatively well established in China[178] and India.[179] Stoves with up to 55% efficiency are available for biogas combustion offering lower GHG emissions than traditional designs. Gasification also offers a potential route to the reduction in open agricultural waste burning and would potentially reduce unnecessary sources of PM and CO_2 emissions if such fuels could be burned to provide useful energy.[163] Biogas can also be produced from the anaerobic fermentation of organic matter from waste products such as agricultural manure, food waste as well as landfill gas.[180]

The potential success of using biogas as a fuel depends on the availability of appropriate gasification or anaerobic digestion technologies. However, barriers to wide scale implementation need to be addressed which are not necessarily technical in nature, and can include issues related to infrastructure, financial and perceptual barriers.[181] Economic viability is also a key issue which is in part related to gasification and combustion efficiencies. Several studies identify solving problems related to secondary environmental pollution as key to the wide spread implantation of biomass gasification in developing countries.[178] Methods for improved tar removal could remove the need for waste water treatment methods, improving efficiency and reducing costs. There have been a number of studies on the use of catalysts for the removal of tar during gasification with scope for further research at the practical scale.[182,183]

There are a growing number of studies investigating the development of gasifying stoves, although it is claimed that such designs may have already existed 300 years ago.[184] Such stoves are based on two stage combustion with a fraction of air introduced into the lower part of the reactor in order to gasify solid biomass into syngas. Secondary air is introduced at the top of the reactor, to ensure complete combustion. Both natural draft or forced draft designs have been developed. Perez et al. investigated a 3 kW forced air multi-fuel gasification stove using a range of solid fuels including briquettes made from the densification of agricultural solid wastes.[163] They found significant reductions (84-93% for high and low power operation) in fine particulates compared to traditional stove designs. They also noted a decrease in fuel usage of around 60% due to the improved efficiency of the stove design. Gitau et al., found reductions in indoor concentrations of CO, CO_2, and $PM_{2.5}$ by 57%, 41%, and 79% respectively when using a gasifying stove compared to using a three-stone open fire.[185] Mitchell et al. compared emissions from a raised grate charcoal stove, a rocket stove using dry wood, wet wood and straw, and a gasifier 'top lit' down-draft stove using wood pellets.[186] The gasifier-type stove was found to emit the lowest levels of all mass based pollutant emissions, although high particle numbers were found which may require further investigation. A more recent study of this gasifying stove found a low signal from Laser induced incandescence (LII) measurements, indicating that the ultrafine particles seen using a fast particle analyser, were composed of inorganic or tar particles that would vaporise at temperatures too low to generate an LII signal, rather than of soot.[187] KCl is suggested as the most likely component, but clearly further work on ultrafine PM from gasifier stoves for a range of fuel types would be useful, since composition would clearly affect potential toxicity. A number of studies have addressed design principles of gasifying cook stoves with Kirch et al. for example[188] indicating that low primary air supply rates with utilisation of the resulting char for subsequent applications, may be beneficial from an air pollution point of view. [188]

10. FINAL DISCUSSION AND FUTURE DIRECTIONS

The AQ, epidemiological and toxicological evidence raises questions as to whether the domestic burning of biomass is environmentally sustainable, since without suitable mitigation technologies, unacceptable exposure to harmful pollutants can result both within indoor and outdoor settings. In terms of combustion efficiency and the ability to control harmful emissions, especially of PM, large scale combustion of biomass in power stations or CHP units makes more sense. However, such large scale utilisation puts pressures on biomass supply chains, which must utilise carbon neutral sources involving sustainable replanting or the use of genuinely waste materials. Even then, the timescales involved in carbon sequestration of large trees pose problems for carbon accounting and for achieving shorter term carbon reduction goals.

In terms of domestic use, even in industrialised countries, it is difficult to envisage legislation that severely limits an individual's choice to burn solid fuels. Indeed, attempts made to ban wood burning in Paris in 2014 met with fierce public opposition and plans were scrapped only days before the ban was due to come into force.[189] Other mitigation measures such as the prohibition of wood burning on open fires, tightened emissions limits for new stoves, and regulations on fuel quality may have some impact, but their success will depend on the degree of enforcement, as well as turnover rates for old appliances and the energy system that they are replacing. Turnover rates could potentially be encouraged through grants and scrappage schemes such as those introduced in Denmark and Norway[190], but even an ultra-modern domestic stove meeting the EDD emissions limits, is still likely to emit as much PM per hour as 6 Euro VI heavy goods vehicles.[119]

If we are not as a society willing to ban the use of solid fuels in domestic settings, then the development and mandatory use of both primary and secondary control technologies will be vital to limiting AQ impacts from biomass burning. These could include primary measures such as restrictions on fuel quality, education programmes on appropriate device usage, and designs that limit poor operation, as well as secondary measures such as domestic scale ESPs and catalytic removal methods. Fuel treatment methods provide further scope for reducing emissions by removing moisture, semi-volatiles and inorganic components from the fuel, but emissions from treatment plants themselves need to be properly regulated. Further research is necessary to establish the effectiveness of all these mitigation options for a range of biomass fuel types, in terms of speciated emissions both within gas and particulate phases, and within **real world** as well as in laboratory based settings. Again, we can look to the automotive sector to realise the importance of real world testing, where based on a programme of real world tests, the US EPA revealed that Volkswagen had intentionally programmed diesel engines to activate NOx emissions controls only during regulatory laboratory emissions testing.[191] As a result, NOx emissions in the real world were substantially higher and did not meet regulatory limits. Amongst other things, the diesel scandal highlights the challenges in designing pollution mitigation systems that simultaneously reduce emissions of relevance to both climate change and AQ.

For the biomass sector, further challenges remain for the mitigation of semi-volatile emissions and ultrafine particles, as well as for gaseous emissions that lead to the formation of atmospheric ozone and SOA. Previous research has demonstrated an extremely high level of variability in emissions factors for manually operated small scale devices, although there are clearly some trends related for example to fuel quality/moisture content, ash, trace species and volatile content, wood contamination, air supply, combustion mode, fuel supply rates and firing intensity. A better understanding of speciated emissions factors under different mitigation options used over a range of operational conditions and fuel qualities, could assist the development of guidelines on fuel quality, as well as localised emissions limits or "no burn days" that could potentially be used in areas with existing poor air quality, or during periods of unfavourable meteorological conditions such as low wind speeds or limited vertical mixing due to temperature inversions.[190]

Enforcement of any future policies is a key issue and requires sufficient resources in order to be carried out effectively. Most urban areas in the UK for example, have been designated as smokeless zones as part of the Clean Air Act. This means that indoor burning of wood on open fires is already prohibited. However, the recent survey carried out on behalf of DEFRA, suggests that this practice is still widespread, implying that insufficient resources are available within local government to enforce the ban.[76]

Long term ambient monitoring strategies that can identify the contribution of biomass burning, particularly to PM, are needed within residential areas, and would help to develop a better understanding of concentration trends, as well as the impact of any technological and policy interventions. Improved monitoring is also needed within indoor environments, as well as at the level of personal exposure, in order to support epidemiological studies on health outcomes that are able to couple exposure measurements with symptoms tracking to establish causal links. An improved understanding of exposure to pollution from biomass burning and the resulting health impacts may also help to promote behavioural change. A substantial part of biomass burning in industrialised countries is currently providing secondary heat, and therefore alternative options exist in many households. Over the next few decades however, large changes to the provision of domestic heat will be required, which may involve switching to electricity based systems, expanded use of heat networks and/or the use of non-carbonaceous fuels such as hydrogen. How biomass fits into future low carbon strategies for heat, requires careful consideration of resulting potential changes to AQ and pressures on supply chains. System wide studies including LCAs with a scope broad enough to include both climate and AQ relevant emissions could help to support overall strategy development for the domestic heat sector.

Within developing countries, particularly in rural environments, domestic fuel burning for cooking and heating is likely to remain a significant environmental challenge that must be addressed in order to improve both public health and climate impacts. Here though, there are opportunities to improve sustainability goals in both these areas, since currently used inefficient stoves lead to over usage of precious fuel resources, as well as to high emissions of CO_2, NOx, PM and VOCs. Gasification technologies do exist for domestic cook stoves, and interventions are required to promote their large scale uptake, potentially providing positive benefits to both human health and GHG emissions, although further work may be needed to investigate ultra-fine particle emissions. In all areas of biomass usage for energy, we should be aiming for such win-win strategies that combine improvements for climate and air quality outcomes and thus human health.

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TABLE OF NOMENCLATURE

AQ	Air Quality	NAEI	National Atmospheric Emissions Inventory
AQEG	Air Quality Expert Group	NET	Negative Emission Technology
BC	Black Carbon	NMVOC	Non-methane Volatile Organic Compound

BECCS	Bioenergy Carbon Capture and Storage	NOx	Nitrogen Oxides ($NO+NO_2$)
CCS	Carbon Capture and Storage	OC	Organic Carbon
CHP	Combined Heat and Power	PAH	Polyaromatic Hydrocarbon
DEFRA	Department for Environment, Food and Rural Affairs	PCDD	Polychlorinated Dibenzo-p-dioxin
EDD	Ecodesign Directive	PCDF	Polychlorinated Dibenzofurans
ESP	Electrostatic Precipitator	PM	Particulate Matter
GHG	Greenhouse Gas	$PM_{2.5}$	Mass of particles that pass through a size-selective orifice with a 50% collection efficiency cut-off at 2.5 µm aerodynamic diameter
GWP	Global Warming Potential	PM_{10}	Mass of particles that pass through a size-selective orifice with a 50% collection efficiency cut-off at 10 µm aerodynamic diameter
IEA	International Energy Agency	PMF	Positive Matrix Factorisation
		ROS	Reactive Oxygen Species
IED	Industrial Emissions Directive	SCR	Selective Catalytic Reduction
IPCC	Intergovernmental Panel on Climate Change	SNCR	Selective Non-Catalytic Reduction
LCA	Lifecycle Analysis	SOA	Secondary Organic Aerosol
LPG	Liquid Petroleum Gas	US EPA	United States Environmental Protection Agency
LSH	Local Space Heater	VOC	Volatile Organic Compound
MCPD	Medium Combustion Plant Directive	WHO	World Health Organisation
NAAQS	National Ambient Air Quality Standards		

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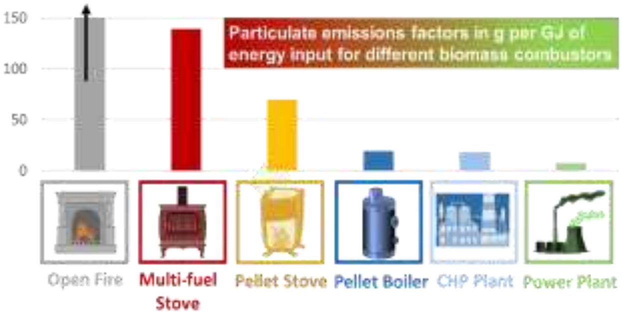
TOC Graphic


150
100
50
0
Particulate emissions factors in g per GJ of energy input for different biomass combustors
Open Fire
Multi-fuel Stove
Pellet Stove
Pellet Boiler
CHP Plant
Power Plant