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Impact of residential wood combustion on ambient benzo(a)pyrene levels

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supervised by

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LIST OF ABBREVIATIONS

%	percent
℃	degree Celsius
µg	microgram
BaP	Benzo(a)pyrene
BTX	Benzene/toluene/xylene
CO2	Carbon Dioxide
e.g.	Exempli gratia
EC	Elemental Carbon
EC	European Commission
EEA	European Economic Area
et al.	Et alii / aliae / alia
etc.	Et cetera
ETIA	MSc Program Environmental Technologies and International Affairs
EU	European Union
Fig.	Figure
IIASA	International Institute for Applied Systems Analysis
m^3	Cubic meter
MT	Metric Ton
Mtoe	Million tons of oil equivalent
MW	Megawatt
ng	Nanogram
no.	Number
OC	Organic Carbon
PAH(s)	Polycyclic Aromatic Hydrocarbons
PM	Particulate matter
pp.	Pages

Rep.	Report
TPM	Total particulate matter
TSP	Total Suspended Particles
UN	United Nations
UNECE	United Nations Economic Commission for Europe
UNEP	United Nations Environment Programme
vol.	Volume

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ABSTRACT

In the EU “Integrated Climate and Energy Policy” the increased use of biomass fuels is promoted to reduce the emissions of fossil fuel derived greenhouse gases. An unexpected side effect, the increased emission of gaseous and particulate organic compounds associated with incomplete combustion (e.g. as occurring during the ignition and the final smouldering phase of solid fuels) has been overlooked in its importance until recently. In the thesis the impact of wood smoke on the air quality of European sites is investigated with special emphasis on fine particles (PM) and the polycyclic aromatic hydrocarbon benzo(a)pyrene (BAP). With a BaP/PM ratio method it was derived, that for regions with wood smoke derived PM10 levels above 5 µg/m^3 on an annual basis the EU target value for BaP of 1 ng/m^3 is likely to be exceeded. The results of this study show that wood smoke is the predominant source of BaP in Europe. Elevated PM and BaP levels from wood smoke are to be expected for densely populated European rural and semi rural environments, where wood combustion is a major source of residential heat generation. It appears that larger urban centres are not typical wood smoke combustion areas, while settlements in suburbs and rural areas seem to use fuel wood more frequently, in particular in areas with frequent inversions. Based on the investigation it was concluded that a violation of the EU BaP target value of 1 ng/m^3 is likely to take place at numerous sites in Europe, namely at rural and semi rural settlements in valleys and basins with reduced aerial ventilation and effectual wood supply.

1 INTRODUCTION

Biomass exists in various forms (such as waste, wood, bio-diesel fuels, alcohol fuels), all of which can be combusted: for 90% of the biomass, combustion is the process where it is converted to energy (Janasek, 2008).

The household sector is one of the largest users of energy in the EEA, consuming 29% of the final energy consumption (excluding energy used for transport) which in 2009, amounted to EU27 1825.2Mtoe (EEP, 2009). Space heating accounts for most energy consumption per dwelling in the EU (69 %) (EEA, 2009).

The energy types typically used in European homes are remote heat, electricity, and fuels such as coal, oil distillates, natural gas, and biomass based fuels, mostly wood or from woody material (EEP, 2009).

As fuel prices rise and the international community tackles the challenges of climate change, the world is turning towards renewable energy sources as the ideal providers (Summa, 2008a). Wood - despite its long lead times (Reitze, F. et al, 2008) in comparison to other renewable energy sources (e.g. wind, solar) – is a relatively CO2 neutral renewable source, and until now, it has supplied 80% of the biomass used for EU energy (Summa, 2008a).

Liquid and gas fuels are burning with very low particle emissions, while solid fuels (e.g. coal and wood) combustion in particular in smaller units like small stoves and fire places are prone to considerable particle emissions. These particle emissions contain inorganic and organic constituents. Among the organic constituents are polycyclic aromatic hydrocarbons, a group of compounds where some members exhibit carcinogenic activity. The main carcinogenic PAH is benzo(a)pyrene, for which a target value of 1 ng/m^3 in the PM 10 annual mean has been included in the fourth EU daughter directive 2004/107/EU to be implemented in national legislation by European nations (UBA, 2006).
While in the past 4 decades a trend from the solid fuel coal towards liquid and gaseous fuels had been occurred, increasing prices for fossil fuels are favouring a switch to woody fuels in countries with ample wood supply and specifically in rural areas.
A further impact on increased wood fuel use originated from the EU Integrated climate and energy policy. In order to mitigate the warming effect from greenhouse gases one of the proposed measures is to replace fossil fuels by renewable biogenic material.

The Austrian National Inventory report states that ‘The average surface temperature of the earth has risen by about 0.6-0.9℃ in the past 100 years and, according to the fourth assessment report of the IPCC, will rise by another 1.8–4.0℃ in the next 100 years, depending on the emission scenario’ (UBA, 2008a). This temperature increase will alter the hydrologic cycle, as well as the Earth's Albedo ('total reflectivity of the earth') and the atmospheric circulation, which is the driving force behind temperature, wind and rainfall at a regional level. Combined, these changes are considered to bring about increased occurrences of extreme weather events (i.e. hurricanes, tornadoes, storms, droughts, floods) (UBA, 2008a).

The EU integrated Climate and Energy Policy established GHG emission reduction targets of 20-30 % below 1990 levels by 2020. It also established energy targets for energy efficiency (consumption reduced by 20% compared to projections), and aims that 20% of consumer energy in 2020 will come from renewable sources (Summa, 2008a). To promote this idea the switching from use of fossil fuels for residential heating to wood based fuels, various economic policy measures are being used, including price setting, tax breaks and investment subsidies (Eder et al, 2005).

The EU policy for renewable energies has as objectives environmental sustainability and combating climate change, competitiveness, ensuring availability of affordable energy, and securing energy supply (Summa, 2008b). The EU Council estimates that benefits of reaching this 20% target in 2020 include annual CO2 emission savings of 600/900 MT from 2020 onwards, as well as a net increase in employment (300 000 jobs created), and an annual reduction in fossil fuel demand of 252 MToe from 2020 and onwards (Summa, 2008b). The two most important EU directives promoting renewable energy sources are Directive 2001/77/EC (on the promotion of electricity produced from renewable energy sources in the internal electricity market) and Directive 2003/30/EC (on the promotion of the use of biofuels or other renewable fuels for transport) (Eder et al, 2005). Although the EU has legislative framework for renewable energies in regards to electricity production and the use of biofuels for transport, it has no legal framework for promotion in the heating and cooling sector (Summa, 2008b). However, it does have support instruments for biomass production, which are included in the Common Agricultural Policy (CAP), like the cultivation of non-food crops on land specifically limited to this purpose (Summa, 2008b).

As most of the homes with direct use of fossil fuels are operating oil or gas stoves and boilers, the switch to fuel wood is connected to a potentially higher PM emission. The problems that stem from increasing PM emissions from wood stoves have been addressed by several studies in recent years (Caseiro et al, A.E. Schimdl et al). An unexpected side effect, the increased emission of organic compounds associated with incomplete combustion (e.g. as occurring during the ignition and the final smouldering phase of solid fuels) has been overlooked in its importance until recently (Papers from Scandinavia, San Francisco, Baumbach paper from Germany). Some reports from Air quality problems in wood burning communities (Scandinavia, Libby Ward Montana study,) were already aware that wood combustion has the potential to raise ambient PM10 and PAH levels above national air quality standards.

The aim of this thesis is to investigate the potential impact of wood combustion emissions on rural and urban sites in Europe in particular on wood smoke derived fine particles levels and on the potential levels of the main PAH compound benzo(a)pyrene in the ambient air. In this thesis, a BaP/PM10 ratio method is introduced to assess the potential impact of wood combustion on ambient BaP levels. The "prognostic ratio" is used to estimate BaP levels for selected European sites, where information about wood smoke PM10 or PM2.5 levels are available. The results are verified by cross checks from ambient data where BaP/PM ratios from ambient measurements are available.

The results of this study show that wood smoke is the predominant source of BaP in Europe. Elevated PM and BaP levels from wood smoke are to be expected for densely populated European rural and semi rural environments, where wood combustion is a major source of residential heat generation. It appears that larger urban centres are not typical wood smoke combustion areas, while settlements in suburbs and rural areas seem to use fuel wood more frequently, in particular in areas with frequent inversions. It can be anticipated, that in densely forested countries more fuel wood is used than in countries with sparse wood areas. Based on the BaP/PM ratios investigated it was derived, that a violation of the EU BaP target value of 1 ng/m^3 is likely in European rural and semi rural settlements in alpine and pre-alpine valleys and other regions with reduced aerial ventilation and ample wood supply.

2 EMISSIONS

Energy trends nowadays point towards a demand for renewable and affordable energy like biomass, solar, wind, etc. Wood is one of the oldest renewable energy sources, and is used worldwide for household heating, which gives often a false sense that it is a clean and safe combustion method; however, wood smoke emits harmful compounds and can cause serious health problems.

Smoke is made up of “airborne solid and liquid particulates and gases emitted when a material undergoes combustion or pyrolysis, together with the quantity of air that is entrained or otherwise mixed into the mass” (Wikipedia article on smoke).

The process that forms the particles is similar in all instances of fuel types and combustion conditions, although the emission rates vary based on fuel type and combustion efficiency (for example, if the fuel contains a lot of volatile compounds that are not combusted, then the amount of PM formed will be high) (Kupiainen and Klimont, 2006).

Wood smoke is proven to contain more than 100 chemicals, such as lead, cadmium and arsenic, and wood combustion is one of the most significant emitters of a great number of pollutants such as PM, benzene, toluene and xylenes (BTX), polycyclic aromatic hydrocarbons (PAHs) and particulate matter (PM). A great number of substances emitted by wood combustion are detrimental to human health (some are even classified as carcinogenic((Herdberg et al, 2002). Also, wood emissions are considerably greater than emissions from natural gas and oil combustion. Emission control technologies exist, but they are not uniformly used (Ries et al., 2009). In areas where wood is regularly used for heating, wood combustion contributes up to 25% of airborne PM and 10% of the carbon monoxide in the air (Health Canada, 2004).

Particulate matter (PM) and polycyclic aromatic hydrocarbons (PAHs) are emitted during wood combustion (particularly incomplete wood combustion), and there is strong evidence that PAHs (like benzo(a)ppyrene – BaP) are toxic constituents of fine PM (Fierro, 2000). Particulate matter (PM) is formed in the atmosphere by transformation of gaseous emissions; the term refers to “a mixture of solid particles and liquid droplets suspended in the air; their chemical and physical compositions depending of location, time of year, and weather” (Fierro, 2000).

PM is composed by coarse and fine particles (Fierro, 2000). “Coarse particles (PM10) have an aerodynamic diameter between 2.5 μm and 10 μm”. PM10 lasts in the atmosphere from minutes to hours, and can travel from less than 1km to 10 km.

Fine particles (PM2.5) refers to “particles with an aerodynamic diameter of less than or equal to 2.5 μm, that are collected by measuring devices with 50% collection efficiency” (UBA, 2008b). They are formed during combustion, “from gas and condensation of high-temperature vapors” (Fierro, 2000), they last anywhere from a few days to weeks, and can disperse for up to thousands of kilometres. Fine and coarse particles differ in chemical composition and formation (see Table 3).

The Formation of PAH and PM 2.5 revolves around “the oxidation of benzene, the formation of cyclopentadienyl radicals, and reaction with C3H3 radicals. PAHs can subsequently form soot via hydrogen abstraction and acetylene-addition. Larger molecular-weight PAHs, such as BaP, are semivolatile organic compounds” (Klimont et al., 2002).

2.1 Emissions of PM

The climatic and health effects of PM are a principal political and environmental concern.

Starting with 2008, EU legislation requires PM2.5 and PM10 levels to be reported. However, in order to implement policies and decrease emissions, PM formation and important sources need to be understood (Caseiro et al, 2009).

PM arises from combustion processes (industrial or domestic) and during mechanical processes (like surface abrasion) and secondary formation (from SO2, NOx, NMVOC or NH3) (UBA, 2009); amongst these, domestic combustion is one of the major sources (Kupianien and Klimont, 2004).

The process that forms the particles is similar in all instances of fuel types and combustion conditions, although the emission rates vary based on these two factors (for example, if the fuel contains a lot of volatile compounds that are not combusted, then the amount of PM formed will be high) (Kupianien and Klimont, 2004).

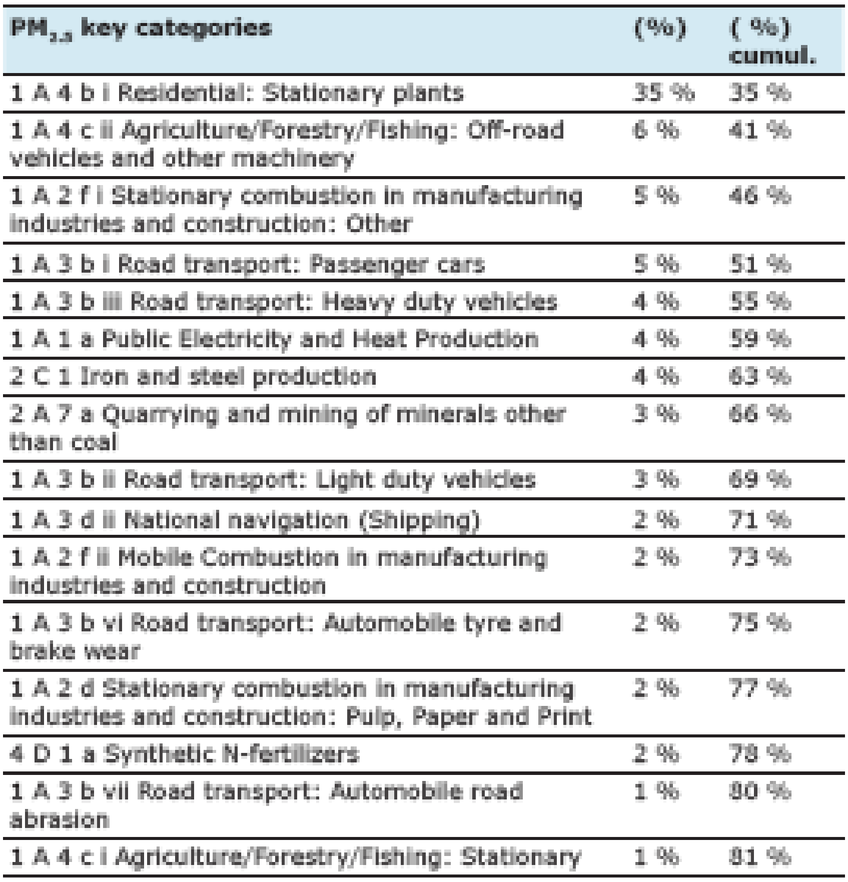
Particulate Matter is usually categorized as TPM (particulate matter with a diameter less than 100 microns), PM10 (diameter less than 10 microns), PM2.5 (diameter less than 2.5.

microns), and submicron (diameter less than one micron).

Particle size from wood combustion depends on the type of wood (moisture content, tree species, etc) and type of combustion appliances. Some studies show that emissions from wood combustion are up to 95% fine particulates (Klimont et al., 2002).

Submicron particles have a long atmospheric residence mode (days to weeks), as they are immune to “removal mechanisms like diffusion or gravitational settling” (Kupianien and Klimont, 2004), which makes them particularly burdensome polluters.

PM emissions stemming from wood combustion are overall significant because it is a very popular domestic energy source worldwide. In Europe, up to 40% of PM2,5 emissions result from domestic combustion (typically biomass), which is more than emissions caused by vehicles (Salonen, 2008). This makes household energy use the greatest source of PM2.5 emissions (EEA, 2009). In 2008 for example, the most important emission source in the EU 27 was Domestic fuel use ('1 A 4 b i – Residential: Stationary Plants' in Table1), responsible for 35% of total fine particulate emissions (UNECE and FOAO, 2008).



$PM_{2,5}$ key categories	(%)	(%) cumul.
1 A 4 b i Residential: Stationary plants	35 %	35 %
1 A 4 c ii Agriculture/Forestry/Fishing: Off-road vehicles and other machinery	6 %	41 %
1 A 2 f i Stationary combustion in manufacturing industries and construction: Other	5 %	46 %
1 A 3 b i Road transport: Passenger cars	5 %	51 %
1 A 3 b iii Road transport: Heavy duty vehicles	4 %	55 %
1 A 1 a Public Electricity and Heat Production	4 %	59 %
2 C 1 Iron and steel production	4 %	63 %
2 A 7 a Quarrying and mining of minerals other than coal	3 %	66 %
1 A 3 b ii Road transport: Light duty vehicles	3 %	69 %
1 A 3 d ii National navigation (Shipping)	2 %	71 %
1 A 2 f ii Mobile Combustion in manufacturing industries and construction	2 %	73 %
1 A 3 b vi Road transport: Automobile tyre and brake wear	2 %	75 %
1 A 2 d Stationary combustion in manufacturing industries and construction: Pulp, Paper and Print	2 %	77 %
4 D 1 a Synthetic N-fertilizers	2 %	78 %
1 A 3 b vii Road transport: Automobile road abrasion	1 %	80 %
1 A 4 c i Agriculture/Forestry/Fishing: Stationary	1 %	81 %

Table 1: PM 2. 5 Key emitting categories in Europe (UBA, 2009)

In Central and Western Europe, biomass smoke contributes 10 to 30 % of organic matter annually. In winter, the share increases to between 20 and 50%. Although usually high 2.5 PM emissions are associated with urban industrialized areas, during winter, rural areas usually emit just as high, due to the frequent use of woodstoves, thus making biomass smoke 'the predominant organic aerosol constituent in winter time mid and western Europe' (these calculations do not take into consideration the emitted secondary organic aerosols) (Puxbaum et al., 2007).

Although PM 2.5 emissions have been reported to have overall decreased since 2000, PM emissions reports from member states are not as accurate or complete as for other emissions (EEA, 2009). In terms of biomass information availability, the National Wood Resource Balance states that there is a serious need for 'up-to-date', reliable statistical information on wood sources and uses', but that this information is either missing or insubstantial in numerous states (UNECE and FOAO, 2008). The Joint UNECE/FAO/IEA/EC Wood Energy Enquiry began to accumulate and assemble accurate data on supply and uses for wood energy at a European level. This has allowed for the foundation of 'national and European-level wood balances (study on "wood resources availability and demands") (UNECE and FOAO, 2008).

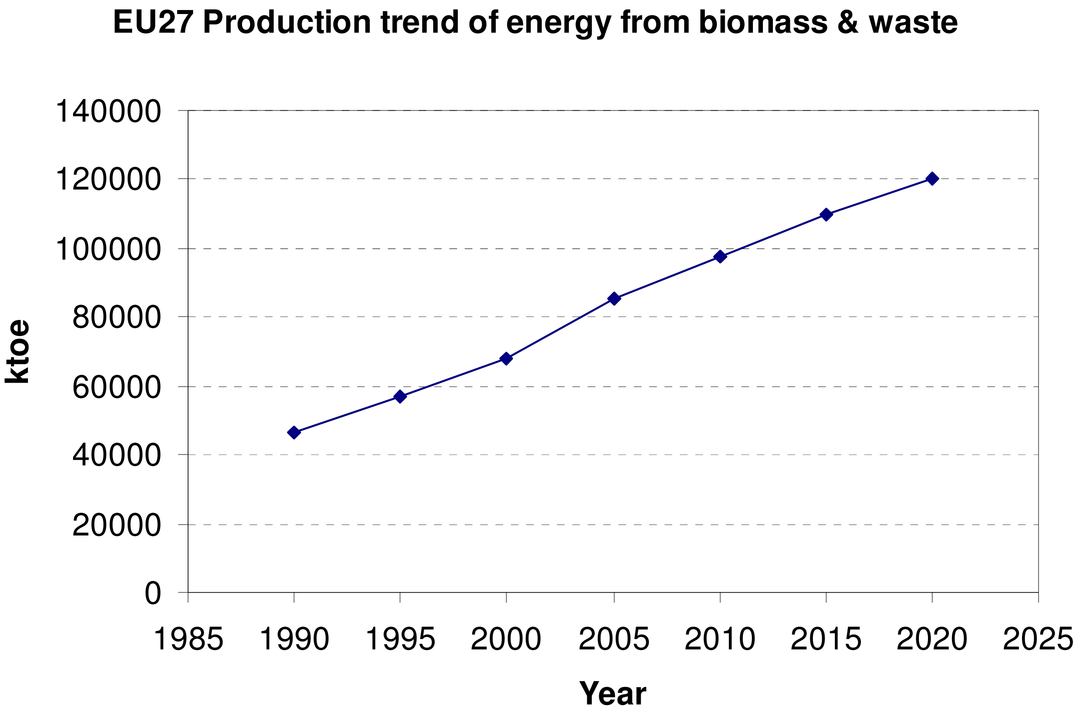
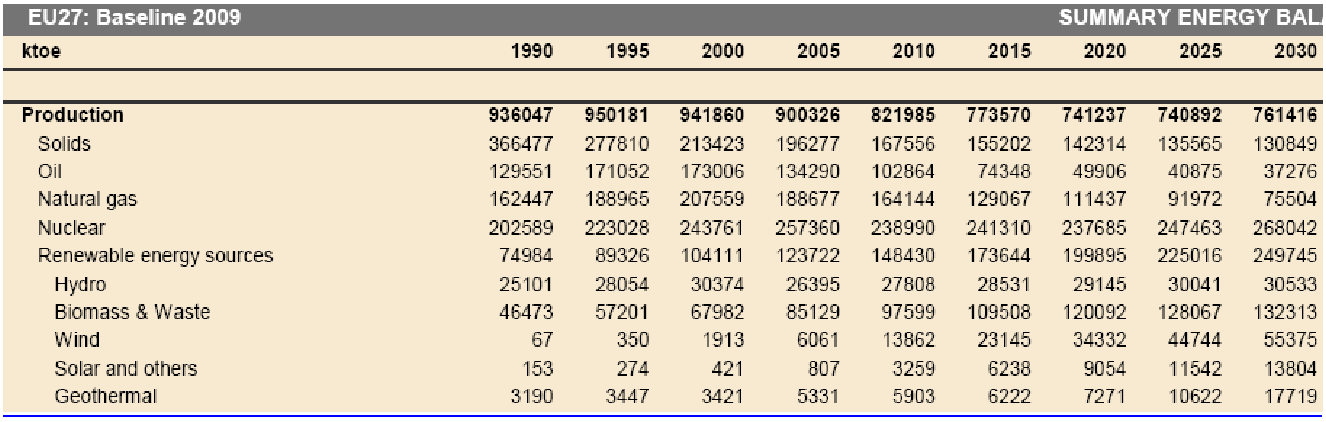


Figure 1: EU27 Energy Balance Summary (based on Table 2)



EU27: Baseline 2009									SUMMARY ENERGY BAL
ktoe	**1990**	**1995**	**2000**	**2005**	**2010**	**2015**	**2020**	**2025**	**2030**
Production	**936047**	**950181**	**941860**	**900326**	**821985**	**773570**	**741237**	**740892**	**761416**
Solids	366477	277810	213423	196277	167556	155202	142314	135565	130849
Oil	129551	171052	173006	134290	102864	74348	49906	40875	37276
Natural gas	162447	188965	207559	188677	164144	129067	111437	91972	75504
Nuclear	202589	223028	243761	257360	238990	241310	237685	247463	268042
Renewable energy sources	74984	89326	104111	123722	148430	173644	199895	225016	249745
Hydro	25101	28054	30374	26395	27808	28531	29145	30041	30533
Biomass & Waste	46473	57201	67982	85129	97599	109508	120092	128067	132313
Wind	67	350	1913	6061	13862	23145	34332	44744	55375
Solar and others	153	274	421	807	3259	6238	9054	11542	13804
Geothermal	3190	3447	3421	5331	5903	6222	7271	10622	17719

Table 2: EU27 Energy Balance Summary (EC, 2007)

Table 2 and Figure 1 show an increasing trend in the use of biomass and waste energy in Europe, with a threefold increase between 1990 and 2030.

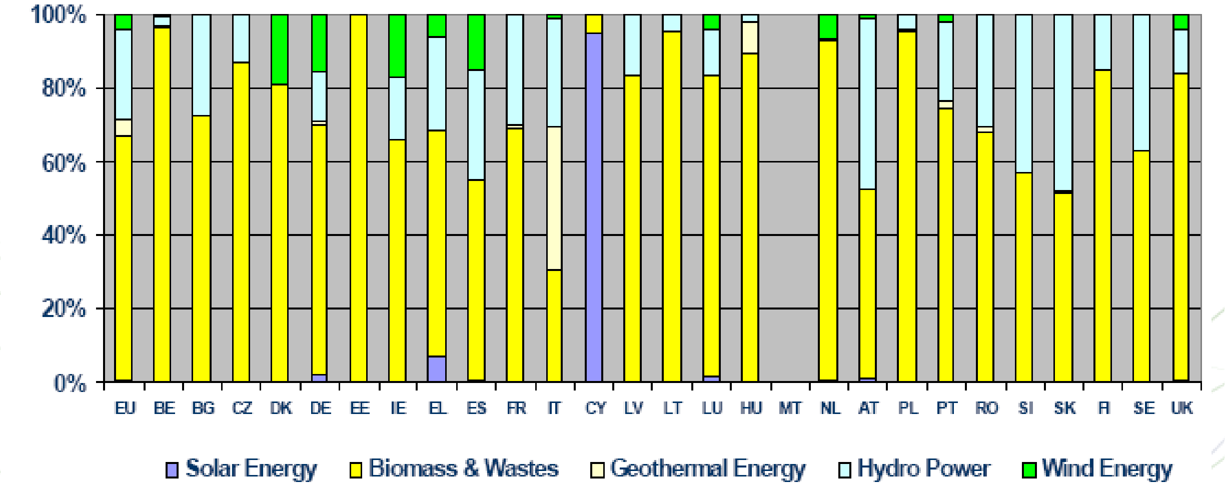


Figure 2: Energy production from renewables – Share by source (Roubanis, 2008)

2.1.2 PM Emission Factors based on Technology

Combustion technologies nowadays offer different combustion conditions that lead to different chemical and physical properties in the particles emitted. *Bølling* et al. (2009) reviewed literature referring to wood combustion particles, and observed that although there is data on particles emitted under different combustion states, there is a lack of thorough data on particle variation from precise combustion appliances.

Wood can be combusted in different categories of combustion appliances, such as stoves, fireplaces and boilers. Since the most important feature that influences biomass carbonaceous emissions is combustion temperature, it follows that the “air supply, burn cycle, stove design, burn rate, species and moisture content of the fuel” are directly related to these emissions and influence the amount and type of particles emitted in the wood smoke (including the probability to cause health damage) (Kupiainen and Klimont, 2007).

Fine particles (PM2.5) released through biomass combustion can be divided into 3 categories (depending on physical and chemical characteristics): spherical organic carbon particles, aggregated soot particles and inorganic ash particles (*Bølling* et al., 2009). All 3 types can be emitted and can interact in the same combustion environment.

Table 3 describes the main conditions for formation and particle characteristics for the 3 categories. Carbon emitted during combustion is organic or elemental: “Organic carbon (OC) comprises hundreds to thousands of organic compounds, whereas elemental carbon (EC) is defined as the carbon that is not organic, but EC can also be characterised as refractory carbon” (*Bølling* et al., 2009). When burning biomass under low temperature, PM emissions contain mostly organic carbon.

When there is not enough air, but high temperature combustion occurs, soot (“elemental carbon aggregates”) compose the majority of emissions, with less than 1 percent of emissions mass attributed to organic and elemental carbon (*Bølling* et al., 2009)

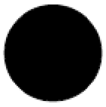
	Spherical organic carbon particles	Soot (elemental carbon aggregates)	Inorganic ash particles
Schematic drawing			
Diameter measured by electron microscopy*	50-600 nm [52, 53]	20-50 nm [52, 73]	50-125 nm [97]
Mobility diameter	100-300 nm [68-70]	50-300 nm [68, 76]	50-125 nm [69, 98, 99]
Internal turbostratic microstructure	No [61]	Yes / No [81-83]	No
Solubility (H_2O)	Depends on ageing [61]	Insoluble	Soluble
Main chemical characteristic	Organic carbon [62, 64, 67] (Most abundant organic compounds: metoxyphenols and monosaccaride anhydrides) [57-60]	Elemental carbon with variable amounts of organics condensed on the surface [12, 62, 81] (Most abundant organic compounds: hydrocarbons and polycyclic aromatic hydrocarbons) [84, 85]	Alkali salts (mainly KCl and K_2SO_4 with small amounts of trace elements (e.g. Zn)) [78, 92]
Combustion conditions	Low-temperature, incomplete combustion [11, 52-56]	High-temperature, incomplete combustion [52]	High-temperature, complete combustion [120]
Possible sources	Air starved combustion or start-up phase of batch wise combustion in conventional stoves, open fireplaces [58,62,64,67]	Combustion in conventional stoves, open fireplaces, boilers for wood, wood chips and pellets [14, 52, 75-79]	Combustion in pellets stoves, boilers for wood, wood chips and pellets [69, 120]

Table 3: The physicochemical characteristics of the three classes of wood combustion particles (Bølling *et al, 2009).* The numbers refer to the references used in the text. * For the aggregated soot particles the listed diameter refers to the primary particle diameter.

Implementing successful measures to cut down emission levels requires in-depth information on the extent of individual sources and their respective emissions. Table 4 shows 4 categories of wood combustion appliances and their emissions. Open fireplaces emit mostly a combination of soot and OC. Conventional wood stoves emit principally soot, as well as OC condensed onto the soot particles or as individual particles. Conventional wood log boilers and masonry heaters are old categories of appliances, with very little data available on carbon content or physical properties of emissions. Bølling et al. however, deduce that emissions will be mostly soot and organic carbon, although in different proportions based on combustion conditions, which also vary with the type and make of appliance (Bølling et al., 2009). Modern stoves, masonry heaters and boilers for wood logs are modern appliances, with improved combustion technologies and therefore cleaner emissions, mostly made up inorganic ash. However, emissions of OC and soot can increase during certain combustion phases (like during star-up), or if they are not properly operated. Pellets stoves and boilers are also modern appliances, and are technologically designed to provide ideal combustion conditions, so that the main emissions are mostly inorganic ash. However, just like with modern stoves, heaters and boilers, if inappropriately operated, emissions will result in higher soot or OC.

Emissions data for different combustion appliances exists, but lacks when it comes to modern wood stoves and boilers for logs. However, descriptions of particle properties and respective emission conditions are lacking; it is important to gather more information about the different particle properties, since the extent of health effects depends on particulate size and chemical properties (for example, EC is linked with cardiovascular mortality) (Kupiainen and Klimont, 2007).

Type of combustion appliance	Reported emission factors	
	Approximate range (mg/MJ)	Reported data (mg/MJ)
Open fireplaces	160 - 910	800 [a]
		160 - 447 [b,1]
		860 - 910 [b,2]
Conventional wood stoves	50 - 2100	700 [a]
		94 - 650 [b,1]
		50 - 1932 [b,2]
		100 [c]
		150 - 2100
Other conventional stoves, including masonry heaters and sauna stoves	30 - 140	140 [a]
		30 - 100 [c]
Conventional boilers for wood logs		

without accumulator tank	50 - 2000	700 [a]
		300 - 2000 [b,1 and 2]
		1300 [c]
		300-900
with accumulator tank	50 - 250	80 [a]
		50 - 300 [b,1 and 2]
		95
Modern wood stoves	34 - 330	34 [c]
		330
Modern boilers for wood chips or logs	5 - 450	5-450[b,1]
		20 - 25 [c]
		30-100
Pellet stoves and boilers	10 - 50	30 [a]
		10 - 50 [b,1 and 2]
		20 [c]
		30

Emission factors are reported as mg particles emitted per MJ of fuel burnt (MJ = Mega Joule)

[a] mean emission factors based on available literature.

[b] range of emission factors based on data from members of the International Energy Agency

[c] range of emission factors [120] .

Table 4: Emission factors for different types of residential combustion appliances (Bølling et al., 2009)

2.2 Emissions of PAH/Benzo(a)pyrene;

Definition: “The polycyclic aromatic hydrocarbons (PAH) are molecules built up of benzene rings” and form a group of approximately 100 compounds (UBA, 2008b). The majority of PAHs come from incomplete combustion of substances containing carbon (e.g. coal, wood, oil, waste); PAHs can also arise from forest fires. However, the majority of PAH emissions are due to anthropogenic activities. PAH concentrations vary significantly in various rural and urban environments and are mainly influenced by vehicular and domestic emissions (Ravindra et al., 2008). PAHs are released in particulate and gas forms, and they are a concern as many are known to be carcinogenic and mutagenic.

A principal source of gas PAHs in many countries is residential wood combustion. As biomass use is increasing in popularity, these emissions are likely to grow. Coal and oil combustion also results in PAH emissions. However, gas PAH emissions are most likely low, and there is very little information on residential oil heating (Bignal et al, 2008). Researchers found that wood emits more carcinogenic and genotoxic PAHs per mass of fuel burnt than bituminous coal and respectively, coal briquettes (Bignal et al, 2008).

32 PAH compounds, with concentrations varying between 0.06 to 7 mg/kg, made up between 0.12 and 0.38% of the PM mass, based on the wood and facility kind (Gullet et al, 2003).

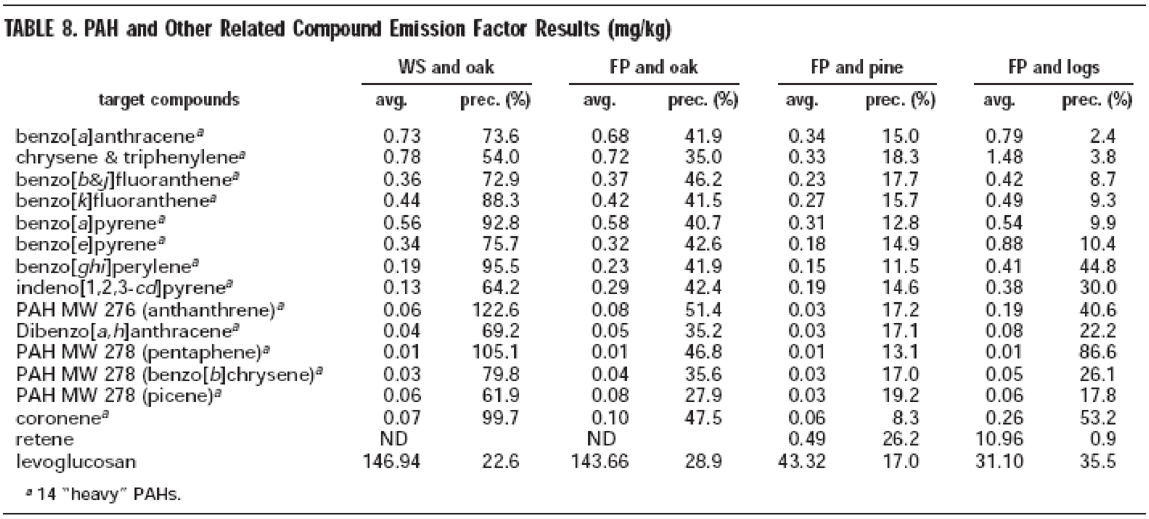


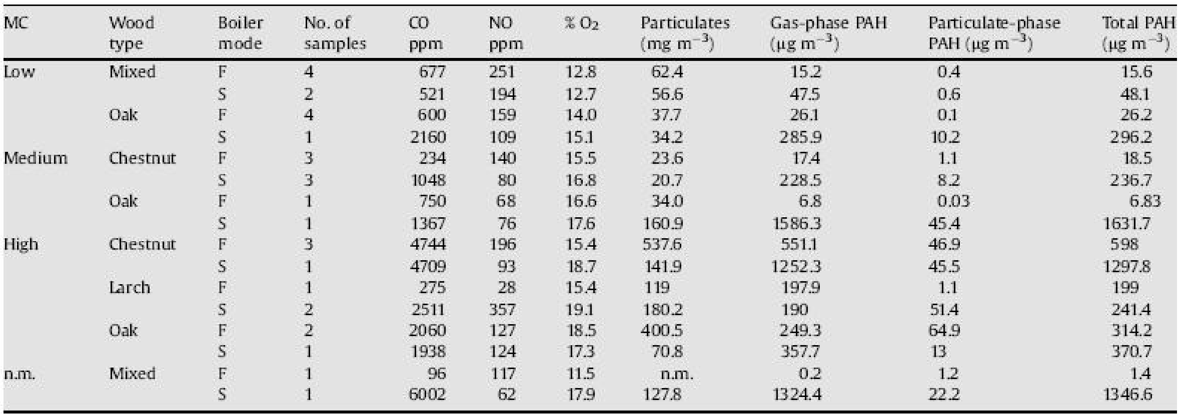
TABLE 8. PAH and Other Related Compound Emission Factor Results (mg/kg)

	WS and oak		FP and oak		FP and pine		FP and logs	
target compounds	avg.	prec. (%)	avg.	prec. (%)	avg.	prec. (%)	avg.	prec. (%)
benzo[*a*]anthracene[a]	0.73	73.6	0.68	41.9	0.34	15.0	0.79	2.4
chrysene & triphenylene[a]	0.78	54.0	0.72	35.0	0.33	18.3	1.48	3.8
benzo[*b&j*]fluoranthene[a]	0.36	72.9	0.37	46.2	0.23	17.7	0.42	8.7
benzo[*k*]fluoranthene[a]	0.44	88.3	0.42	41.5	0.27	15.7	0.49	9.3
benzo[*a*]pyrene[a]	0.56	92.8	0.58	40.7	0.31	12.8	0.54	9.9
benzo[*e*]pyrene[a]	0.34	75.7	0.32	42.6	0.18	14.9	0.88	10.4
benzo[*ghi*]perylene[a]	0.19	95.5	0.23	41.9	0.15	11.5	0.41	44.8
indeno[1,2,3-*cd*]pyrene[a]	0.13	64.2	0.29	42.4	0.19	14.6	0.38	30.0
PAH MW 276 (anthanthrene)[a]	0.06	122.6	0.08	51.4	0.03	17.2	0.19	40.6
Dibenzo[*a,h*]anthracene[a]	0.04	69.2	0.05	35.2	0.03	17.1	0.08	22.2
PAH MW 278 (pentaphene)[a]	0.01	105.1	0.01	46.8	0.01	13.1	0.01	86.6
PAH MW 278 (benzo[*b*]chrysene)[a]	0.03	79.8	0.04	35.6	0.03	17.0	0.05	26.1
PAH MW 278 (picene)[a]	0.06	61.9	0.08	27.9	0.03	19.2	0.06	17.8
coronene[a]	0.07	99.7	0.10	47.5	0.06	8.3	0.26	53.2
retene	ND		ND		0.49	26.2	10.96	0.9
levoglucosan	146.94	22.6	143.66	28.9	43.32	17.0	31.10	35.5

[a] 14 "heavy" PAHs.

Table 5: PAH and Other Related Compound Emission Factor Results (mg/kg) (Gullet et al, 2003)

As PAH emissions result from inefficient combustion, residential boiler operating conditions and moisture content (MC) were proven to be more important emission concentration factors than tree species (Bignal et al., 2008). In order to reduce PAH emissions from boilers, woodchip fuel should be low in moisture content, and the boiler operation should avoid slumber modes (basically a smoldering flame), as PAH emissions are higher in slumber mode vs. full flame; this can be done by “increasing automatic operation capability of wood-fired boilers” (ex: auto-start capabilities, and automatic fire rate variations) (Bignal et al., 2008).

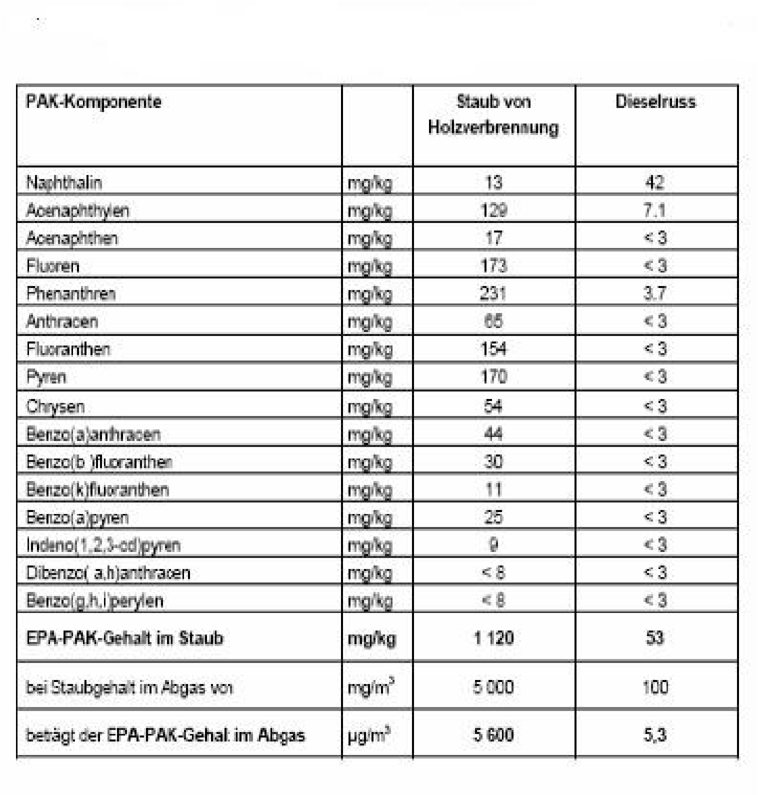


MC	Wood type	Boiler mode	No. of samples	CO ppm	NO ppm	% O_2	Particulates (mg m^{-3})	Gas-phase PAH (μg m^{-3})	Particulate-phase PAH (μg m^{-3})	Total PAH (μg m^{-3})
Low	Mixed	F	4	677	251	12.8	62.4	15.2	0.4	15.6
		S	2	521	194	12.7	56.6	47.5	0.6	48.1
	Oak	F	4	600	159	14.0	37.7	26.1	0.1	26.2
		S	1	2160	109	15.1	34.2	285.9	10.2	296.2
Medium	Chestnut	F	3	234	140	15.5	23.6	17.4	1.1	18.5
		S	3	1048	80	16.8	20.7	228.5	8.2	236.7
	Oak	F	1	750	68	16.6	34.0	6.8	0.03	6.83
		S	1	1367	76	17.6	160.9	1586.3	45.4	1631.7
High	Chestnut	F	3	4744	196	15.4	537.6	551.1	46.9	598
		S	1	4709	93	18.7	141.9	1252.3	45.5	1297.8
	Larch	F	1	275	28	15.4	119	197.9	1.1	199
		S	2	2511	357	19.1	180.2	190	51.4	241.4
	Oak	F	2	2060	127	18.5	400.5	249.3	64.9	314.2
		S	1	1938	124	17.3	70.8	357.7	13	370.7
n.m.	Mixed	F	1	96	117	11.5	n.m.	0.2	1.2	1.4
		S	1	6002	62	17.9	127.8	1324.4	22.2	1346.6

Table 6: Mean concentrations of various pollutants from different fuels under different boiler operation (Bignal et al., 2008)

Test studies on different boiler and stoves showed that PAH emissions were 30 times greater in furnaces employing older technologies versus newer ones. Therefore the technology used is an important factor in PAH emissions, alongside fuel type: for example, log emissions are much higher than woodchip emissions (Bignal et al, 2008). Biomass combustion is proven to emit a variety of gas (89%) and particulate PAHs. The slumber phase emits higher concentrations than full-flame (494 mgm┐ 3 versus 265 mgm┐ 3) (Bignal et al., 2008).Some researchers argue that although fuel type does influence PAH emissions, the most important aspect is combustion efficiency, whereby emissions varied from 5 to 683 mg kg┐ 1 (Bignal et al., 2008). Other studies concluded that the most important PAH emission factors were combustion technique (they tested wood furnaces, stoves, and boilers) and MC. For example, wet spruce logs emissions (MC higher than 30%) were 5 times greater than from dry spruce logs (MC 15-20%). In fireplaces, emissions from wood continued after the flame died, and 20-30% of emissions were produced during the smoulder phase (Dasch, 1982).

The best way to reduce PAH emissions is to use low MC fuel, and operate the boiler at full flame (avoid slumber modes). Several other ways to reduce emissions are: advanced combustion appliance design, prolonging combustion times, and altering combustion chamber temperature.



PAK-Komponente		Staub von Holzverbrennung	Dieselruss
Naphthalin	mg/kg	13	42
Acenaphthylen	mg/kg	129	7.1
Acenaphthen	mg/kg	17	< 3
Fluoren	mg/kg	173	< 3
Phenanthren	mg/kg	231	3.7
Anthracen	mg/kg	65	< 3
Fluoranthen	mg/kg	154	< 3
Pyren	mg/kg	170	< 3
Chrysen	mg/kg	54	< 3
Benzo(a)anthracen	mg/kg	44	< 3
Benzo(b)fluoranthen	mg/kg	30	< 3
Benzo(k)fluoranthen	mg/kg	11	< 3
Benzo(a)pyren	mg/kg	25	< 3
Indeno(1,2,3-cd)pyren	mg/kg	9	< 3
Dibenzo(a,h)anthracen	mg/kg	< 8	< 3
Benzo(g,h,i)perylen	mg/kg	< 8	< 3
EPA-PAK-Gehalt im Staub	**mg/kg**	**1 120**	**53**
bei Staubgehalt im Abgas von	mg/m^3	5 000	100
beträgt der **EPA-PAK-Gehalt im Abgas**	$\mu g/m^3$	**5 600**	**5,3**

Table 7: Poor wood combustion shows higher PAH levels than diesel soot. Clean wood combustion eliminates most PAH formation (Albrecht, R.J., 2007)

PAH emission data is incomplete and uncertain, principally because a focus on inorganic pollutants due to legal requirements, as well as feasibility (technical and financial) of measurements. PAH measurements are expensive and technically difficult ("and only a mean value over several hours can be obtained") (Bignal et al., 2008). Also, there is little data on the effects of fuel and MC in biomass boilers, which is very important in assessing emission quantity and quality. The European emissions of PAH – namely at least of 4 indicator-PAH, including BaP – is reported on a yearly basis by EU states to the UN ECE – EMEP – LRTRAP program (program on Long Range Transport of Air Pollutants).

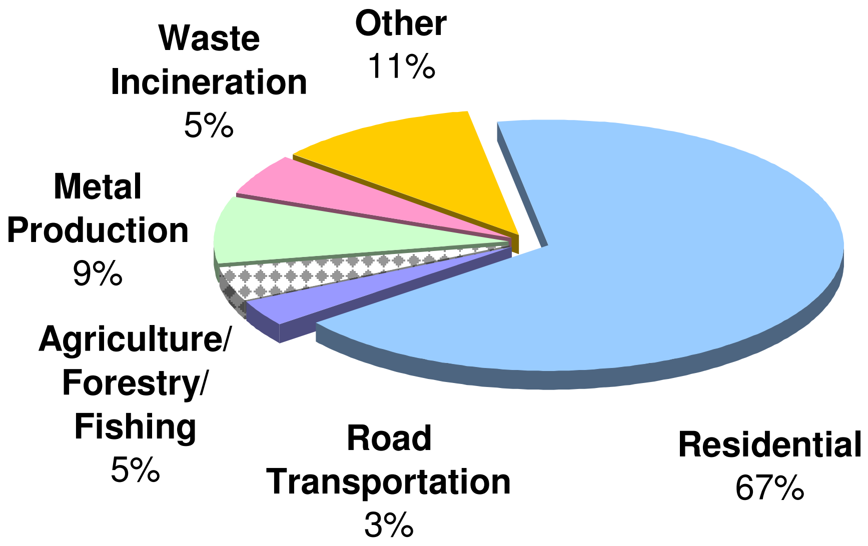


Fig. 3: Sector split for PAHs emissions in 2006 (23 countries), % (EMEP, 2008)

The EMEP Status report 3/2008 (EMEP, 2008) deals with the emissions and the transport modeling of POP's (Persistent Organic Pollutants) including BaP. The EMEP domain includes Europe, Turkey, Israel, Russia and further West- and Central Asian countries. The report states that

> "*the information on PAH emission spatial distributions was provided by 24 countries (Austria, Belarus, Belgium, Bulgaria, Croatia, Denmark, Estonia, Finland, France, Germany, Hungary, Iceland, Ireland, Italy, Latvia, Lithuania, Monaco, the Netherlands, Norway, Poland, Slovakia, Spain, Sweden, the United Kingdom). Among them, 16 countries (underlined) submitted gridded sector data. In comparison with the previous year, one more country – Croatia – reported gridded emission data*" (EMEP, 2008).

Official information on emissions of four indicator PAHs by sectors in 2006 is available for 23 countries. The sector split for PAH emissions for these countries is presented in Figure 3. The Residential sector is the major contributor to the total annual emission accounting for on average 67%. The predominant source in the Residential sector is combustion of wood. The second most important sector is Metal Production. This sector is a significant source of PAHs for Italy and Sweden."

“The spatial distribution of PAH emissions for 2006 is illustrated in Fig. 4 by taking of B[a]P as an example. It can be seen that elevated levels of B[a]P emissions (20 – 200 g/km2/y) [annual deposition of BaP per km²] are characteristic of the Central, Southern and Eastern parts of Europe. For the most parts of Northern and Western Europe, Russia, and the Central Asian countries emission fluxes do not exceed 20 g/km2/y”.

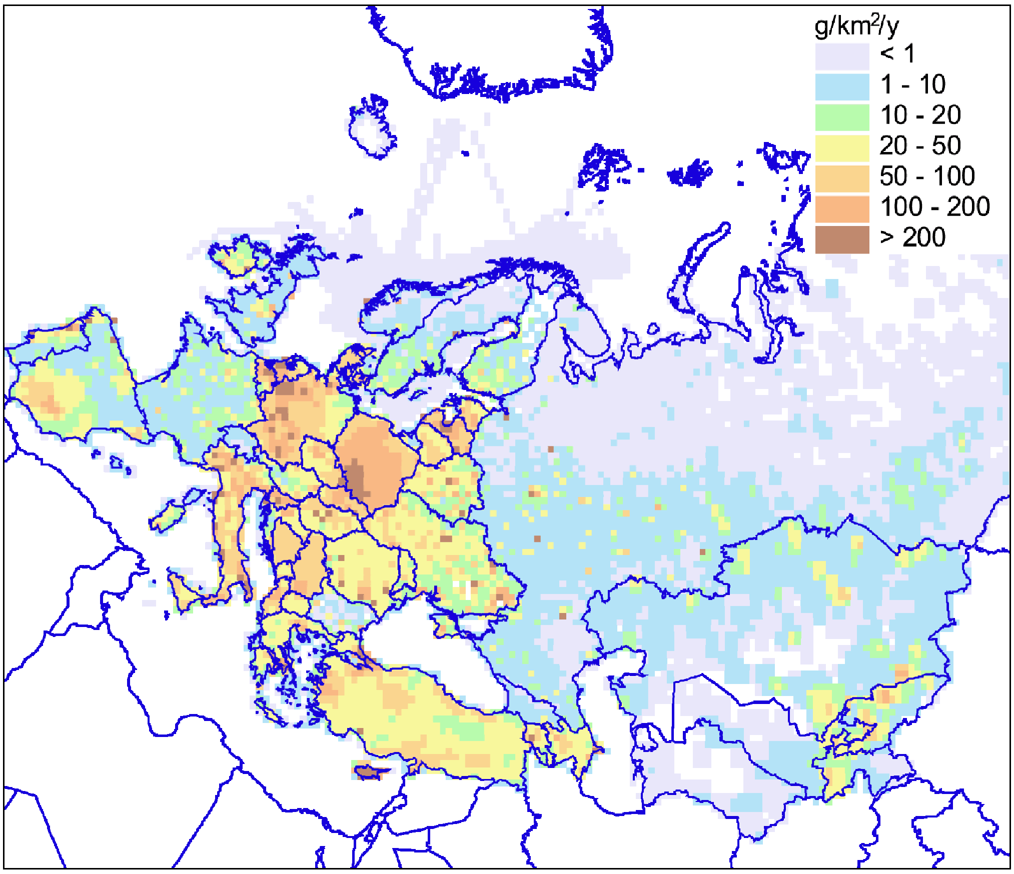


Fig. 4: Spatial distribution of B[a]P emissions in 2006 over extended EMEP grid with resolution 50x50 km2, g/km2/y (EMEP, 2008)

The report shows that emissions of the indicator PAHs within the EMEP domain decreased by 20% - 27% in the period from 1990 to 2006 (Fig. 5). Considering the emission trends it is further reported:

> “*Among the countries that submitted official data on PAH emissions for 2006, maximum emission reduction within the considered period took place in the UK (95%) and Republic of Moldova (68%). At the same time in Denmark, Italy, Latvia, Poland, Portugal, and Sweden PAH emissions were increased in comparison with the level of*

> *emission in 1990. The total emissions of B[a]P within the extended EMEP grid in 2006 used for model calculation amounted to 494 tonnes. Considering the B[a]P annual emissions of individual countries it can be noted that maximum contributions to total emission within the EMEP region were made by the Ukraine (20%), Poland (10%) and Turkey (8%).”*

The increase of PAH emissions in several European countries is likely a result of increasing use of biomass for residential heating.

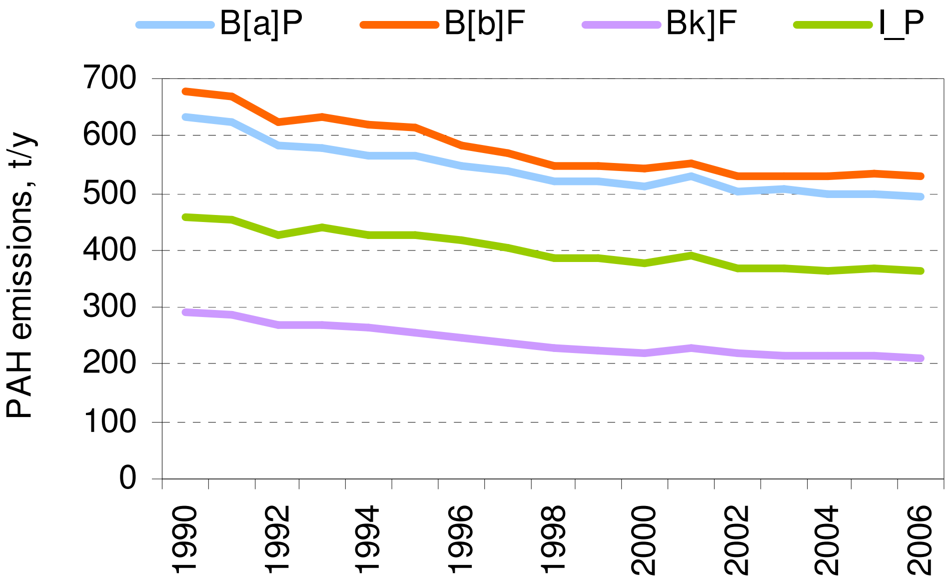


Fig. 5. Temporal trends of B[a]P emissions within the EMEP grid in 1990-2006, t/y (from EMEP 2008)

The sectoral split of PAH emissions in Austria is shown in Fig. 6, where 72.4 % of the PAH emissions originate from domestic sources.

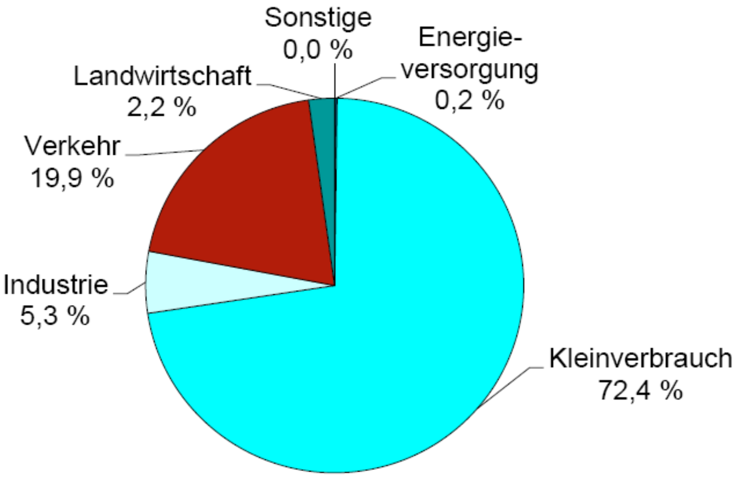


Figure 6: Emission sectors of PAH in Austria 2008 (UBA, 2010a)

Figure 6 shows contributions of different sectors to PAH emissions in Austria (2008): Small-Scale consumption contributes 72.4%, Industry 5.3 %, Traffic 19.9%, Agriculture 2.2 %, and Energy Supply 0.2%. The PAH emissions and emission trends in Austria are reported by Umweltbundesamt (UBA, 2010b). There a considerable reduction of the PAH emissions from 1985 (27 Mg) to 17 Mg in 1990 and 8 Mg in 2008 – 70% reduction from 1985-2010 is stated. In 1985, the main emission sources for PAH emissions in Austria were fuel combustion activities (44%), industrial processes (29%) and agriculture (26%). From 1990 to 2008, PAH emissions from agriculture decreased remarkably by 89% due to prohibition of open field burning; PAH emissions from the sector Industrial processes decreased by 97% due to the closure of primary aluminum production in Austria, which was a main source for PAH emissions, and of improvements in industrial processes (e.g. coke oven emissions). The contributions of the individual sources is depicted in Fig.6.

3 HEALTH EFFECTS OF WOOD SMOKE

The general public opinion in many countries is that wood smoke, since it is natural, does not cause any health problems. However, it is scientifically proven (and many toxicologic and epidemiologic studies show) that wood smoke contains thousands of chemicals, out of which many are harmful pollutants, and some are carcinogenic: more specifically, wood smoke contains at a minimum five chemical groups of substances that are ‘classified as human carcinogens by the International Agency for Research on Cancer’ (Naeher et al., 2007). Wood smoke is principally a strong source of fine particles (PM) and polycyclic aromatic hydrocarbons (PAH) (Salonen, 2008). According to Hellen et al., (2008) the most important pollutants from residential wood combustion with respect to health consequences are fine particles, particulate PAH and volatile organic compounds. Fine particles are considered to impose serious health effects (WHO, 2003) and many particulate and volatile organic compounds are harmful or even carcinogenic (U.S. EPA, 2005). The EU has set limit values for annual averages of PM2.5 (25 µg/m^3), benzene (5 µg m^3) and benzo(a)pyrene (1 ng m^3) in ambient air (Directives of the European Parliament and of the Council, 2000/69/EC and 2004/107/EC).

3.1. Health Effects from PM Emissions

Fine particles, specifically PM2.5, currently represent the largest health hazard amongst all the environmental pollutants in Europe. It is therefore imperative to reduce PM 2.5 emissions from small-scale biomass combustion. Particle size influences the gravity of the health problems; therefore legislation implements a size classification for PM (UBA, 2008b). Epidemiological studies show that dust particles promote respiratory and circulatory illnesses, especially in children, asthmatics, and seniors. The danger lies in the fact that they can penetrate deep into the respiratory system. In order to deal with this problem, the EU introduced limits to control them (UBA, 2008b). Studies have also indicated amplified lung inflammation, immunosuppression and weakened clearance of the PM from the lungs of the animals in areas with elevated PM levels. Statistics also indicated a higher number of hospital emergency room visits due to asthma attacks, and an increase in morbidity and mortality due to long-term or acute PM exposure (Salonen, 2008). In the EU programme CAFE (Clean Air for Europe), an assessment of the health impacts of fine particulate matter (PM2.5) has been performed. An overview of the impacts is compiled in Table 8.

Type of health effect	Impact
Premature death cases	347 900 / yr
Life years lost	3 618 700 / yr
Infant death cases	677 / yr
New cases of chronic bronchitis	163 800 / yr
Hospital admission cases, heart and lung	100 300 / yr
Lower respiratory symptom days 5-14 years	192 756 400 d
Restricted activity days 15-64years	347 687 000 d
Value of health damage	268-781 billion euros/year

Table 8: CAFÉ assessment of the health impacts of PM 2.5 in the EU 25 in 2000 (Salonen, 2008)

At the core of the biomass residential heating health problems lie 3 issues: 1) the release of flue gas is near ground level, and usually in highly populous areas; 2) most biomass residential heating occurs during winter, when pollutants are poorly diluted in the air; and 3) small-scale installations mostly do not have treatment for emissions, and their combustion is often not optimized (some studies reported up to 80% emissions of PM 2.5 during winter from wood smoke) (Salonen, 2008). Essentially, most biomass combustion occurs in winter, when people are at work places or homes, which leads in addition to a high chance of exposure to indoor emissions.

Most health effect studies on combustion fumes have been performed for Diesel-exhaust and for tobacco smoke. Dedicated studies on health effects of wood smoke have also been performed, however with less intensity. A range of specific investigations have been performed with persons affected by wood smoke from wild fires (reviewed by Naeher et al., 2007). Wildland fire fighting can lead to intermittent intervals of exposure of high concentrations of irritating gases and smoke particles, including CO, formaldehyde, acrolein and soot particles containing high amounts of toxic organics. Most cases concerned eye-irritations, and to a lesser extent respiratory symptoms (Naeher et al., 2007). Long term studies, however, are not reported.

In some Asian areas large scale haze from agricultural and residential biomass use has been observed and health symptoms associated with the elevated fine PM levels. Similarly, studies have been performed on effects from grass, straw and from sugar cane burning. For residential wood smoke one direct exposure study and a whole range of epidemiological studies have been reported from USA, Canada and New Zealand (reviewed in Naeher et al., 2007). The authors conclude that: "this short summary of published studies shows that significant exposures to ambient wood smoke do occur in developed countries and that important health effects have been demonstrated to result."

Modelling for biomass combustion aerosols has been thoroughly developed, and it is a useful tool in understanding and predicting aerosol formation and interaction with other particles in the atmosphere (International seminar on Aerosols in Biomass Combustion, 2005).

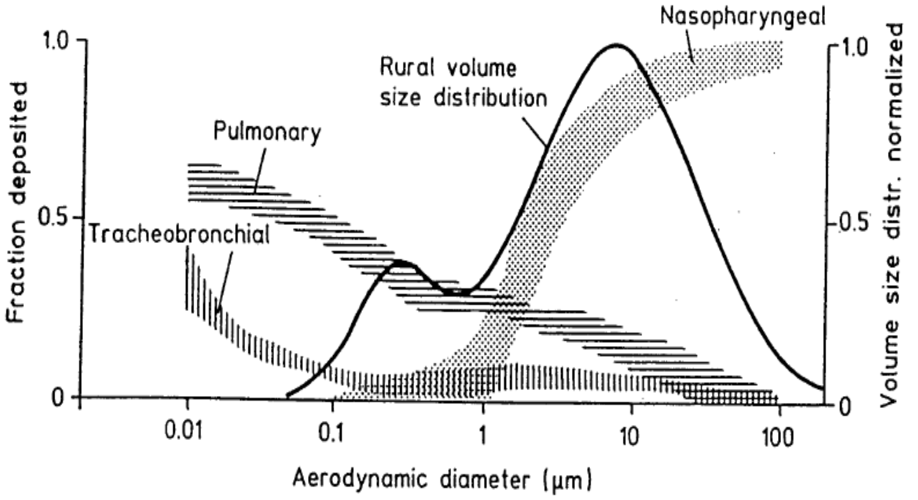


Figure 7: Atmospheric aerosol deposition – three compartment model of the respiratory system (Puxbaum, 2009)

3.2 Health effects of PAH/Benzo(a)pyrene from wood smoke

PAHs are also emitted through biomass combustion, and are a group of complex organic chemicals which are extensively dispersed in the atmosphere; they are also one of 'one of the first atmospheric pollutants groups to have been identified as suspected carcinogen' (Ravindra et al., 2008).

Given that PAH emissions are widespread, and that they are inherently dangerous to human health, there is growing concern with these emissions. Many states have proposed a legal or voluntary limit on PAH concentrations. Based on many studies, health researchers show that

PAH exposure poses a very serious threat, and suggest that PAH should be a top concern when it comes to air quality legislation (Ravindra et al., 2008).

In Europe, Directive 1999/30/EC classifies airborne fine particles that could be inhaled according to their damaging effects on health. Organic pollutants are an important category of contaminants, and it is vital to identify, classify and measure them accordingly, especially when it comes down to PAHs (Ravindra et al., 2008). Toxicological studies of wood smoke have been performed with animal and cultivated tissues exposures. Naeher et al. (2007) summarise the results:

> *"Most available animal studies indicate that exposure to wood smoke results in significant impacts on the respiratory immune system and at high doses can produce long-term or permanent lesions in lung tissues. Based on relatively few studies, these effects seem most strongly associated with the particle phase. Woodsmoke is also mutagenic and possibly carcinogenic in laboratory and field studies, but less so than coal smoke."*

Although several studies have been looking into the health effects of biomass combustion aerosols, the results are still incomplete and there is a need for more in-depth research into which factors (ex. particle size and composition) are more pertinent and under which conditions. Studies should also include cell and inhalation tests for an appropriate anatomical assessment (International seminar on Aerosols in Biomass Combustion, 2005). As Naeher et al. (2007) point out, there is still much research to be done in connection to wood smoke exposure assessments, its chemical composition, as well as toxicology and epidemiology aspects.

A compilation of the major health-affecting pollutants from biomass combustion is given in Table 9 (Naeher et al., 2007).

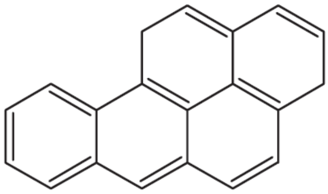
Compound	Examples	Source	Notes	Mode of Toxicity
Inorganic Gases	Carbon Monoxide (CO)	Incomplete combustion	Transported over distances	Asphyxiant
	Ozone (O3)	Secondary reaction of product of nitrogen dioxide and hydrocarbons	Only present downwind of fire, transported over long distances	Irritant
	Nitrogen dioxide (NO2)	High-temperature oxidation of nitrogen in air, some contribution from fuel nitrogen	Reactive	Irritant
Hydrocarbons	Many hundreds	Incomplete Combustion	Some transport-also react to form organic aerosols. Species vary with biomass and combustion conditions	
	Unsaturated: 40+, e.g. 1,3-butadiene			Irritant, carcinogenic, mutagenic
	Saturated: 25+, e.g., n-hexane			Irritant, neurotoxicity
	Polycyclic aromatic (PAHs): 20+, e.g., benzo(a)pyrene			Mutagenic, carcinogenic
	Monoaromatics, 28+, e.g. benzene, styrene			Carcinogenic, mutagenic
Oxygenated organics	Hundreds	Incomplete combustion	Some transport – also react to form organic aerosols. Species vary with biomass and combustion conditions	
	Aldehydes: 20+, e.g., acrolein, formaldehyde			Irritant, carcinogenic, mutagenic
	Organic alcohols and acids: 25+, e.g. methanol acetic acid			Irritant, teratogenic

	Phenols: 33+, e.g., catechol, cresol (methylphenols)			Irritant, carcinogenic, mutagenic, teratogenic
	Quinones: hydroquinone, fluorenone, antrhaquinone			Irritant, allergenic, redox active, oxidative stress and inflammation, possibly carcinogenic
Chlorinated organics	Methylene chloride, methyl chloride, dioxin	Requires chloring in the biomass		Central nervous system depressant (methylene chloride), possible carcinogens
Free radicals	Semiquinone type radicals	Little is known about their formation		Redox active, cause oxidative stress and inflammatory response, possibly carcinogens
Particulate Matter (PM)	Inhalable particles (PM10)	Condensation of combustion gases; incomplete combustion; entrainment of vegetation and ash fragments	Coarse + fine particles. Coarse particles are not transported far and contain mostly soil and ash	Inflammation and oxidative stress, may be allergenic.
	Respirable particles	Condensation of combustion gases; incomplete combustion	For biomass smoke, approximately equal to fine particles	(See below)
	Fine particles (PM2.5)	Condensation of combustion gases; incomplete combustion	Transported over long distances; primary and secondary production	Inflammation and oxidative stress, may be allergenic

Table 9: Major Health-damaging pollutants from biomass combustion (Naeher et al., 2007)

Toxicity and carcinogenic potential of BaP have been investigated thoroughly in the past decades and links between benzo[a]pyrene and cancer have been established. In 1996, a study provided clear evidence linking components in tobacco smoke to lung cancer (Denissenko et al., 1996). Benzo[a]pyrene in tobacco smoke caused genetic damage in lung cells that was identical to the damage observed in the DNA of most malignant lung tumours.

BaP in food was not as effective forming cancer since in most cases small amounts of benzo[a]pyrene were metabolized by gut enzymes prior to being passed on to the blood. The lungs are not protected in this manner. The actual toxic metabolic compound identified was benzo[a]pyrene -7,8-dihydrodiol-9,10-epoxide (Wikipedia article on Benzo(a)pyrene).



benzo[a]pyrene **(B2)**

$C_{20}H_{12}$

Figure 8: Chemical structure of BaP, a mutagenic and carcinogenic PAH (Wikipedia article on Benzo(a)pyrene)

4 AMBIENT WOOD SMOKE AND BAP LEVELS AT SELECTED EUROPEAN SITES

Measurements of PAH at European sites are still sparse, even sparser is the information about the fine particles levels originating from wood smoke. Information of wood smoke levels was derived for a few European sites by source analysis techniques. These data are listed in Table 10. Earlier information that wood smoke can be a major source of air pollutants originates from Nordic countries (Hedberg et al., 2002; Glasius et al., 2006; Hellén et al., 2006). Wood smoke levels for rural west- and central European sites have been reported by Puxbaum et al., 2007. Also in cities elevated levels of wood smoke were observed (Lanz et al., 2007, 2008, Schnelle-Kreis et al., 2007, Caseiro et al., 2009; Favez et al., 2009).

Simultaneous or near-simultaneous BaP and PM data available from European sites are listed in Table 10.

Site	**Sample**	**PM10 µg/m³**	**Source**	**“wood smoke” µg/m³**	**Year**	**Source**	**BaP ng/m³**	**Year**	**Source**
Austria urban sites									
Vienna RBS	PM10	33	UBA2005	1.7	2004	Cas2009	1.0	2007	UBA2008
Vienna KEN	PM10		UBA2005	1.5	2004	Cas2009			
Graz Süd	PM10	38	UBA2005	5.2	2004	Cas2009	2.3	2007	UBA2008
Graz DB	PM10		UBA2005	4.4	2004	Cas2009			
Salzburg RU	PM10	32	UBA2005	2.3	2004	Cas2009	0.9	2007	UBA2008
Salzburg LE	PM10		UBA2005	2.2	2004	Cas2009			

Linz RBG	PM10	45	Jank2006	3.1	2005/06*	Jank2006	0.9	2007	UBA2008
Austria suburban and rural sites									
Vienna Lobau	PM10	20	UBA2005	1.3	2004	Cas2009			
Styria Bockberg	PM10	21	UBA2005	2.7	2004	Cas2009			
Salzburg AN	PM10	16	UBA2005	1.7	2004	Cas2009			
Steyregg	PM10	33	Jank2006	2.6	2005/06*	Jank2006	0.8	2007	UBA2008
Other sites									
Aveiro PT	PM10			5.0 R	2002/04**	Pux2008			
K-Puszta HU	PM10			3.7 R	2002/04**	Pux2008			

Table 10: BaP and fine particles annual averages for selected European sites

R..reconstructed from OC value (PM[Biomass comb]=OC * 1.4 [conversion to OM] * 1.15 [accounting for EC]

RBS .. Rinnböckstrasse; KEN … Kendlerstrasse; DB … Don Bosco; RU ... Rudolfsplatz;
LE … Lehen; RBG Römerberg; AN … Anthering

* ...April 2005-March 2006

** June 2002 - May 2004

Cas2009: Caseiro et al., 2009	Jank2006: Jankowski et al., 2006
UBA2005: Umweltbundesamt, 2005	Pux2008: Puxbaum et al., 2008

Annually averaged wood smoke levels at urban sites in Austria ranged from 1.7-5.2 µg/m³, at a semi-rural site it was 2.6 µg/m³ (Table 10). Since wood smoke originates mainly from space heating, highest values occur in the fall and winter seasons. At Linz Römerberg the January monthly average was 8 µg/m³, compared to the annual average of 3.1 µg/m³ (Jankowski et al., 2006).

In the coastal Portuguese town Aveiro, the bi-annual average for wood smoke was 5 µg/m³ and 9.9 µg/m³ was the winter season average; at the Hungarian background site K-Puzta the bi-annual average for wood smoke was surprisingly high at 3.7 µg/m³ (Puxbaum et al., 2007). The contribution of wood smoke to the annual average PM10 ranged from 5 to 14% at the Austrian sites. For the winter season, wood smoke contributions to ambient PM10 levels increased to 10-20% (Caseiro et al., 2009) and thus formed the strongest source of primary particles in the wintry ambient air at those sites.

The wood smoke concentrations in Table 10 are annual or bi-annual averages. The wood smoke concentration however exhibits a considerable seasonal variation as shown for the site Graz Süd in Figure 9.

Figure 9: Seasonal variation of the wood smoke concentration at the site Graz Süd (after Caseiro et al., 2009)

Wood smoke is responsible for elevated PM10 levels during the cold season. The increase in wood smoke contribution from summer to winter conditions at one of the Austrian cities is shown in Figure 10. From the graph it can be derived that two major sources are responsible for increased PM10 winter levels. The secondary inorganic aerosol (sulphate and nitrate) increases mainly due to the physical property of the ammonium nitrate which exhibits a higher stability at cold temperatures. The second largest increase is due to the wood smoke contribution.

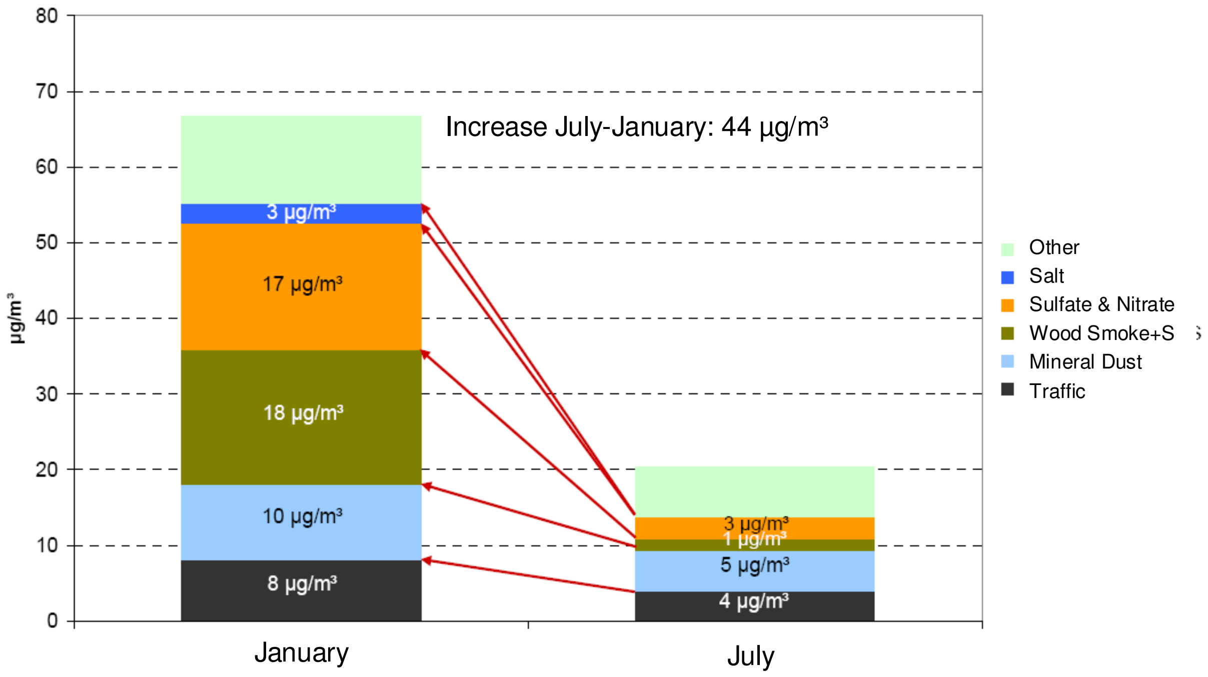


Figure 10: Summer - Winter increase in PM10 levels and source contributions at the site Graz Süd - 2004 (from Bauer et al., 2006)

Wood smoke+S: includes also secondary organic material from wood combustion

At the Hungarian background station K-Puszta the biannual average of wood smoke was 3.7 µg/m³ (Table 10), which is remarkably high compared to other data from Austrian semirural and rural sites (Table 10). The atmospheric levels of the wood smoke concentration have been derived by the tracer compound levoglucosan which is emitted during incomplete combustion of wood (Puxbaum et al., 2007; Caseiro et al., 2009).

The emission of levoglucosan from stoves, fireplaces and boilers is strongly dependent on wood type, wood quality, combustor type, load, and the way how the combustion procedure is employed. The atmospheric concentration is resulting from the variable emissions from stoves and other combustors, however the higher emitters are dominating the wood smoke concentration in the atmosphere, as well as the products of incomplete combustion such as Benzo(a)pyrene. It is therefore no wonder that Benzo(a)pyrene exhibits a similar seasonality as observed for wood smoke.

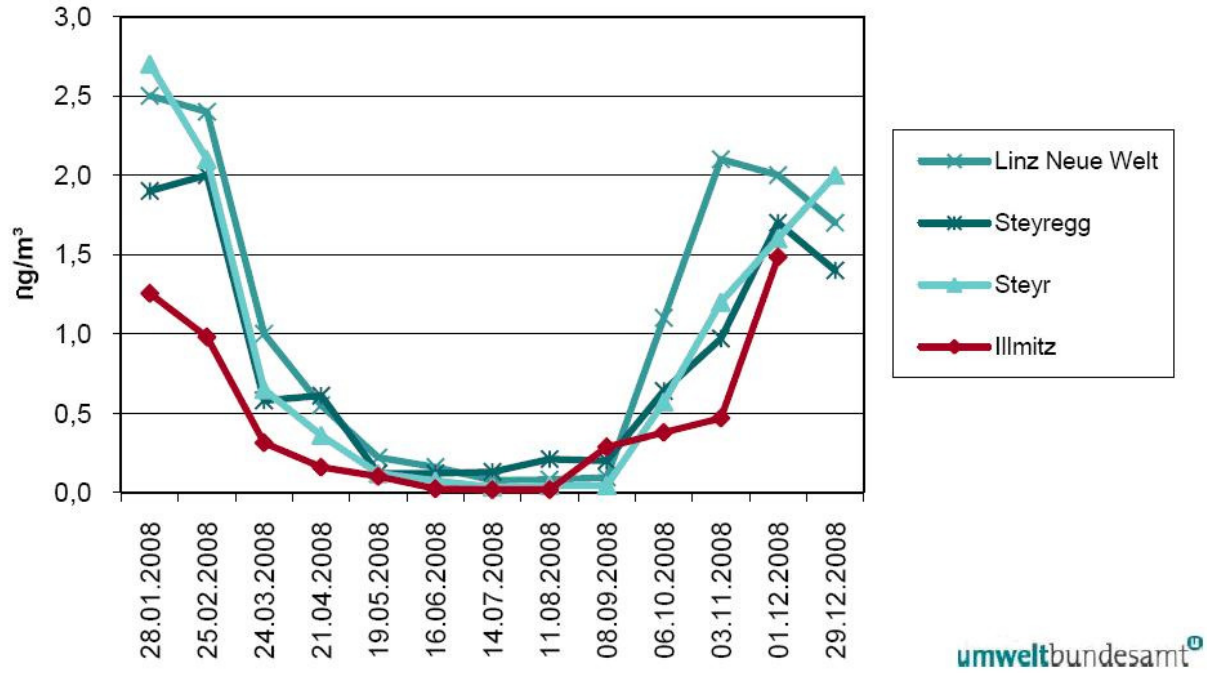


Figure 11: Seasonal variation of the Benza(a)pyrene concentration at various Austrian sites 2008 (from Umweltbundesamt 2009)

The common strong seasonality of the wood smoke and Benzo(a)pyrene concentrations point to wood smoke as the major source of Benzo(a)pyrene levels in the atmosphere.

5 RELATIONSHIP BETWEEN BaP AND PM EMISSIONS FROM WOOD SMOKE

The impact of BaP from wood smoke on ambient air levels has been investigated in a range of studies. We can assume that the majority of the BaP in the atmosphere originates from the higher emitting combustion devices, namely logwood stoves with manual firing. For our consideration, how much of BaP is associated with given fine PM emission from stoves, the relationship between BaP and PM10 or PM2.5 is of interest. The available data from studies reporting BaP and PM levels are listed in Table 10.

We express the BaP/PM ratio as %o (which is equivalent to ng BaP / µg PM). This unit has been chosen, as it allows to derive the atmospheric wood smoke levels which would lead to 1 ng/m^3 wood smoke derived BaP which is the EU standard for the annual average. Since both BaP and particles emissions from wood smoke are predominantly in the fine range, the ratio is not very sensitive when comparing data for PM size fractions PM2.5 or PM10. The BaP/PM ratios for wood combustion in smaller units (as determined from a larger number of studies) exhibit a range from 0.04 – 1.1 for "normal combustion" (Table 10). Higher values were observed for smouldering combustion and for combustion of garden waste (needles, leaves). Interestingly, the lower numbers (BaP/PM < 0.1) are from US combustion tests from stoves and fire places. The combustion tests at European sites yielded typically higher relationships with a grand average of 0.38 and a median of 0.27 ng BaP/µgPM.

It is difficult to judge why the US studies consistently tend to the lower ratios. Also data from other US studies not included in Table 10 (Oros et al., 2001a, b; Fine et al., 2002; Hays et al., 2003) gave comparable results to the Rogge et al., 1998; Schauer et al., 2001 and Gullett et al., 2003 numbers, but are based on small number of combustion tests per wood type. The type and condition of the wood fuel, combustion unit type and construction, temperature at the starting phase, handling of the incensing procedure, combustion duration, air supply during the test burn, etc. influence the emissions of PM and PAH. Differences to the European studies might occur from construction differences of wood burning units, wood quality, air supply and possibly also the relatively long duration of the tests. The average BaP/PM ratio for the three US studies in Table 10 is 0.06 ng BaP/µgPM. This would mean that for an annual mean of 1 ng/m^3 BaP originating from wood smoke, PM levels from wood smoke would reach a level of approximately 17 µg/m^3 annual mean, which is not likely when comparing with ambient data from Libby Montana or other US sites with elevated BaP levels (Ward et al., 2006). It appears that the tests underestimate the BaP emissions.

One study from the European tests gave results similar to the US-studies: The investigation of emissions from a test burn of hard wood (mainly beech) on an open fire test setting (Lee et al., 2005), comparable rather to a fire place than a stove. The tests yielded a BaP/PM ratio of 0.08 ng/µg. The typical difference to stoves is the wide firing cell, where walls are not close to the burning fuel, which reduces quenching of the hot flue gases.

There is a general trend for lower PAH and BaP emissions with increasing output power of wood stoves and boilers, however within a category the range of the emission rate spans up to two orders of magnitude (based on data from Larnesjoe et al., 2000; discussed in Hedberg et al., 2002) presented in Figure 12. The span within a category is obviously due to the factors discussed above (e.g. wood type and condition, stove design, handling of the fuel in the combustor).

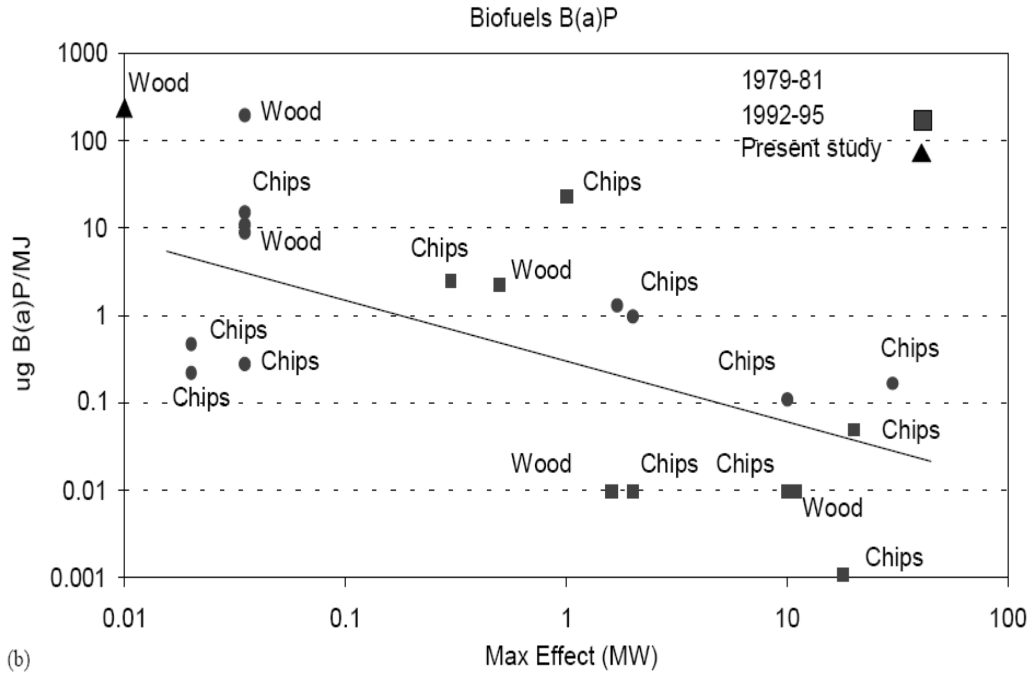


Fig. 12: Emissions of B(a)P from power plants and household devices of output power, using biofuels (Larnesjoe et al., 2000, Hedberg et al., 2002- "present study")

The emission rates of BaP show a certain tendency to increase with increasing PM levels (Fig. 13 for data from US studies).

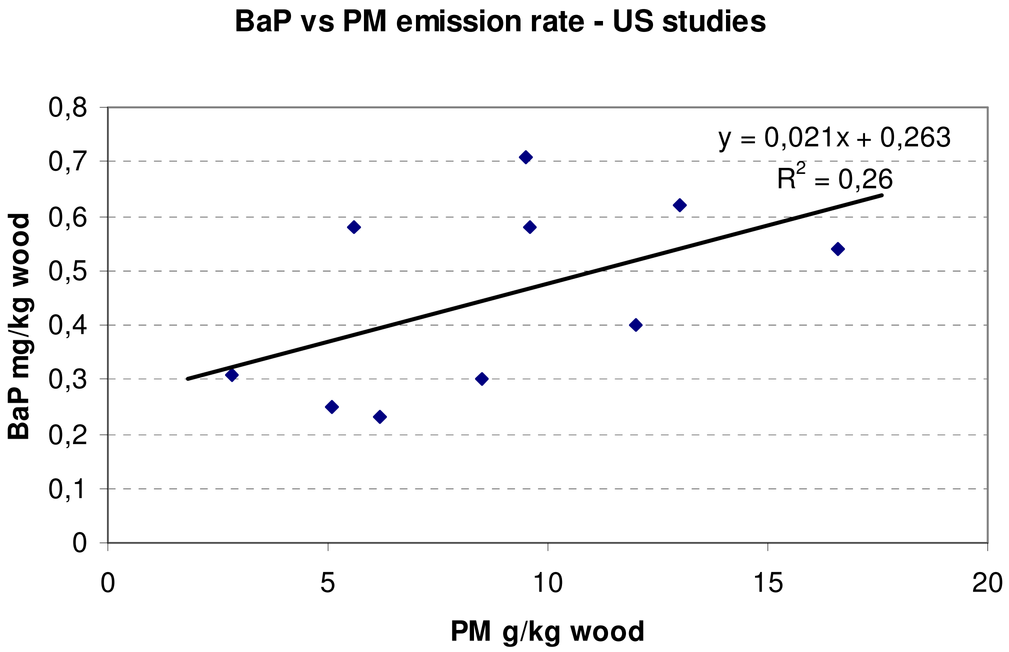


Fig. 13: Emission relation of Benzo(a)pyrene and fine particles (PM) from wood combustion – data from US studies (Rogge et al., 1998; Schauer et al., 2001 and Gullett et al., 2003)

The wood smoke impacted at ambient levels is derived from a large range of combustion units fired with different fuels under different operating conditions. The observed wood smoke concentration thus comprises an averaged sample. To ascribe the BaP/PM emission ratio based on measurements, it is here considered that the best approach is to use the median of the available studies. The reason for choosing the median rather than the average is given by the lesser weighing of extreme values. It is likely that neither the lowest nor the highest values will represent the majority of the emitting appliances; instead, the most probable number will be representative. Thus, for European conditions we consider a BaP/PM ratio of 0.27 ng/µg applicable, with a lower quartile of 0.15 and an upper quartile of 0.53 ng/µg. For Austria we consider the available data sets as representative with a BaP/PM ratio of 0.24 ng/µg applicable, with a lower quartile of 0.16 and an upper quartile of 0.32 ng/µg (Table 11).

The data from European studies are from Sweden, Finland and Austria. The Swedish and Finnish data are obtained from birch wood logs fired in a “soapstone stove” of US origin (Sweden) and a “masonry heater” from Finland. In both studies BaP/PM ratios were relatively high, with a median of 0.67 ng/µg in the Swedish, and an average of 1.11 ng/µg in the Finnish study (Table 11). The data from Austria originate from three different studies, the “Biocomb” study (Luisser et al., 2008), the AQUELLIS FB study (Schmidl et al., 2008) and the “Central Europe wood combustion study” (Rzaca 2010, personal communication). The data are from small wood stoves and pellet stoves and from one boiler from a Hungarian site. The resulting BaP/PM ratios are relatively homogenous, with an average of 0.29 ng/µg. If the high value for the Hungarian boiler is excluded, the average reduces to 0.22 ng/µg. The Hungarian boiler was fired with oak wood also including smaller woody parts such as branches.

Country of study	Source	Stove	Load	PM size	%Water	Fuel		PM	BaP	BaP/PM	BaP/PM
								g/kg wood	mg/kg wood	ng/µg	Ave
USA	W Rogge et al., 1998	fire place			n.r.	Pine, oak, eucalyptus	Ave	9.05	0.42	0.05	0.05
	J Schauer et al., 2001	fire place									
USA	B Gullett et al., 2003	stove, fire place	11-32 kg		17,9,1	Pine, oak, artificial log	Ave	8.65	0.50	0.08	0.08
UK	RGM Lee et al., 2005	open fire test setting	15-16 kg	PM10	11	hardwood/mainly beech		7.9	0.6	0.08	0.08
Sweden	E Hedberg et al., 2002	soapstone stove	1/2/2kg	PM2.5	15	Birch logs	Median	1.6	1	0.67	0.67
Finland	K Hytönen et al., 2009	masonry heater	9 kg	PM1		birch "normal combustion"		1.8	2	1.11	1.11
	(+ J Tissari et al., 2008)	masonry heater	9 kg	PM1		birch "smoldering combustion"		11.1	18.2	1.64	
Austria	Luisser et al., 2008	Log wood stove J		PM10		Beech, briquetts				0.27	0.27
	Biocomb Report	Log wood stove F				Oak, briquetts, spruce				0.20	0.20
(Hungary)		Wood log boiler S				Oak and slash				0.74	0.74
		Pellet stove P				Pellets, chips /spruce				0.10	0.10
Austria	Schmidl et al., 2008	Log wood stove 6kW		PM10		Hornbeam, beech, asch, oak				0.14	0.14
	Aquelis FB Report	Log wood stove 10kW				Briquetts, beech, alder, pine				0.16	0.16
Austria	Rzaca (pers.comm. 2010)	Log wood stove 6kW		PM10		Softwood/Conifers				0.30	0.30
	Centr.Europe wood comb.	Log wood stove 6kW		PM10		Hardwood				0.39	0.39

Ave all	0.33	Median all	0.20
SD	0.32	1. Qu	0.1
RSD%	97.3	3. Qu	0.39
Ave Europa	0.38	Median E	0.27
SD	0.33	1. Qu	0.15
RSD%	86.6	3. Qu	0.53
Ave Austria	0.29	Median A	0.24
SD	0.21	1. Qu	0.16
RSD%	71.6	3. Qu	0.32

Table 11: Compilation of emission results for BaP and PM from various studies

	BaP/PM ng/µg	**PM/BaP** µg/ng
Median all	0,20	**5**
1. Qu	0,10	10
3. Qu	0,39	2,6
Median Europe	0,27	**3,7**
1. Qu	0,15	6,7
3. Qu	0,53	1,9
Median Austria	0,24	**4,3**
1. Qu	0,16	6,5
3. Qu	0,32	3,1

*Table 12: Overview of results for BaP/PM relationships from "all", "European" and "Austrian" studies***.** Data are medians, 1st and 3rd quartiles. The PM/BaP ratio indicates the PM mass concentration in µg/m³ expected for occurrence of 1 ng/m³ BaP from wood combustion. The ranges of the 1st and 3rd quartiles are considered as the possible range of the ratio in ambient air masses for different wood types and equipment in different regions of the study area.

An overview of the results for Bap/PM relationships from "all", "European" and "Austrian" studies are given in Table 12. Data are medians, 1st and 3rd quartiles. The PM/BaP ratio indicates the PM mass concentration in µg/m³ expected for occurrence of 1 ng/m³ BaP from wood combustion. The ranges of the 1st and 3rd quartiles are considered as the possible range of the ratio in ambient air masses for different wood types and equipment in different regions of the study.

The reciprocal of the BaP/PM ratio, the PM/BaP ratio (µg/ng) indicates the PM mass concentration from wood combustion with an associated BaP level of 1 ng/m³. Since 1 ng/m³ is the EU ambient air quality target value for BaP (annual mean), exceeding a certain PM level (annual mean) from wood smoke, as expressed by the PM/BaP ratio would likely lead to a violation of the BaP target value. The median PM/BaP ratio of the European studies is 3.7 µg/ng, with a range (1st-3rd quartile) of 1.9 – 6.7 µg/ng (Table 12). Taking the data from all studies this means that for PM10 levels (annual means) from wood smoke of around 5 µg/m³ and above a violation of the BaP target value is likely. Similar results are obtained for the European and Austrian data set, where about 4 µg/m³ PM is the likely threshold for a BaP violation. Regional deviations might occur when conditions of wood log combustion are different (e.g. other stove/boiler types, other wood types).

6 DERIVING BaP LEVELS FROM WOOD SMOKE AT SELECTED EUROPEAN SITES

The BaP/PM ratios in Table 12 can now be used to derive a BaP concentration for sites where “wood smoke” PM levels were determined. The wood smoke PM levels were derived by using the levoglucosan tracer technique (Simoneit 2002; see discussion in Puxbaum et al., 2007).

Site	**Sample**	**PM10**	**“wood**	**BaP**	**BaP**	**BaP**
		µg/m³	smoke”	**ng/m³**	**ng/m³**	**%** from
			µg/m³		wood smoke	wood smoke
		observed	derived	observed	contribution	contribution
Austria urban sites					f=0,24	
Vienna RBS	PM10	33	1,7	1	0,4	41
Vienna KEN	PM10		1,5		0,4	
Graz Süd	PM10	38	5,2	2,3	**1,2**	54
Graz DB	PM10		4,4		**1,1**	
Salzburg RU	PM10	32	2,3	0,9	0,6	61
Salzburg LE	PM10		2,2		0,5	
Linz RBG	PM10	45	3,1	0,9	0,7	83

Austria suburban and rural sites						
Vienna Lobau	PM10	20	1,3		0,3	
Styria Bockberg	PM10	21	2,7		0,6	
Salzburg AN	PM10	16	1,7		0,4	
Steyregg	PM10	33	2,6	0,8	0,6	78
Other sites					f=0,27	
Aveiro PT	PM10		5		**1,4**	
K-Puszta HU	PM10		3,7		**1,0**	

Table 13: Compilation of determined ambient PM10 and BaP levels (annual averages); derived “wood smoke” from literature data and derived BaP levels from wood smoke (using BaP/PM ratios “f” from Table 12). The derived BaP from wood smoke contribution is expressed in ng/m^3 and % of observed BaP levels.

The levoglucosan tracer technique has been introduced by US groups (e.g. Schauer et al., 1996) and is now used by many research teams. Levoglucosan is a cellulose pyrolysis product; since cellulose is the dominant natural biopolymer, levoglucosan is emitted during the combustion of most biomaterials. By analysing the levoglucosan content of ambient PM samples it is possible to derive the wood smoke contribution to ambient PM levels. A compilation of winter-time levoglucosan levels at various sites is given in Table 14. The estimate of the corresponding PM levels from wood smoke are obtained by multiplying the levoglucosan levels roughly by 10 (Puxbaum et al., 2008; Bauer et al., 2010).

Location	**Measurement period**	**Mean**	**Reference**
		ng/m³	
Dettenhausen, Germ	November 2005-March 2006	806	Bari et al., 2010
Lycksele, Sweden	Winter 2002	897	Hedberg et al., 2006
Graz, Austria	Winter 2004	860	Caseiro et al., 2009
Elverum, Norway	January 30-March 15, 2002	407	Yttri et al., 2005
Oslo, Norway	November 4-December 14, 2001	166	Yttri et al., 2005
Gent, Belgium	January 12-March 11, 1998	477	Zdráhal et al., 2002
Salzburg, Austria	Winter 2004	330	Caseiro et al., 2009
K-Puszta, Hungary	Winter 2002/03	576	Puxbaum et al., 2007
Aveiro, Portugal	Winter 2002/03	957	Puxbaum et al., 2007
Fresno, California	Winter 2003/04	600	Gorin et al., 2006
Bakersfield, California	December 1995-January 1996	2390	Nolte et al., 2001
Fresno, California	December 1995-January 1996	2980	Nolte et al., 2001
Libby, Montana	November 2004-February 2005	3040	Bergauff et al., 2008
Rondônia, Brazil	September 3-October 31, 1999	2006	Zdráhal et al., 2002
Southeast Asia	1994 and 1997	1400-40200	Abas et al., 2004

Table 14: Levoglusosan levels (winter period) from various sites (from Bari et al., 2010)

The derived wood smoke PM levels where annual averages were available are displayed in Table 13. By using the BaP/PM ratio derived in Table 12 (median for Austria and median for Europe), the potential BaP levels from wood combustion have been derived. These levels are also expressed as percentages of the observed BaP concentrations (Table 13). The derived BaP levels from wood smoke are for the Austrian cities in the range of 41-83%, and for a background site 78%.

This result appears very likely when comparing with the emission estimate for BaP in Austria: According to the “Emission trends” of the Umweltbundesamt (2008) the average share of wood smoke of total PAH emissions in Austria is 75%, the share of traffic emissions 18%. The urban sites where BaP data were available are situated at roads with heavy traffic, where the traffic influence is higher than the average from the emission inventory. For the background site, the derived BaP contribution from wood smoke is close to the emission inventory number.

From Table 13 we see also, that for sites where the average wood smoke contribution to PM is around 4 and more, the 1 ng/m³ target value for BaP is likely to become exceeded, which is the case in Graz and is predicted by our method for Aveiro, Portugal and for K-Puszta, Hungary.

Exceedings of the 1 ng/m³ BaP level are now reported for many sites, e.g. in USA, Canada, Australia (Environment Australia 2002), Germany, Austria (Umweltbundesamt 2009) and in particular for Asian sites, e.g. India. In most cases biomass combustion is the main source of the elevated BaP levels. A mitigation of the BaP source strength would require improvement of the wood combustion technology by using filters or applying more efficient combustors.

The Libby, Montana study (Ward et al., 2010) allows to derive BaP/PM ratios from wood smoke data from ambient measurements. For the 2004/2005 heating period, an average BaP of 6.3 ng/m³ and a PM 2.5 level of 28.3 was observed. The wood smoke contribution to PM2.5 was estimated at 70% (Ward et al., 2006). From these data a BaP/PM2.5 ratio of 0.32 ng/µg is obtained. This is slightly above the median for the European studies of 0.27 ng/µg.

7 SYNTHESIS

Wood and other cellulosic biomass is traditionally used for residential space heating namely in countries with sufficient supply from forests. In many countries the use of woody biomass is recommended to mitigate the emission of CO_2 from fossil fuels. Further drivers for the increasing use of fuel wood were the increasing costs of fossil fuels resulting in a favourable competitive situation for wood, and comforts of the visible wood fire in a fire place simulating chimney-stove or the pleasant radiation from tiled stoves. To fulfil the goals of the EU climate and energy policy a doubling of the use of woody material from 2005-2020 for energy purposes is projected (UN/ECE/FAO/Univ Hamburg, 2007; Table 15).

Wood million m³	**Material use (EFSOS calculation)**	**Energy goals (policy objectives)**	**Total use**
2005	478*	343*	821*
2010	495	481	976
2020	536	738	1274
2020 "75% scenario"	536	620	1156
2005*actual figure			

Table 15: EU/EFTA future wood required to fulfil EFSOS scenario and renewable policy objectives (from UN/ECE/FAO/Univ Hamburg, 2007)

The combustion of wood particularly in small scale stoves and boilers is accompanied by smoke emissions. Biomass combustion smoke consists of gases and fine particles. It contains toxic substances such as carbon monoxide, oxides of nitrogen, hydrocyanic acid, benzene, PAH, phenols and soot. Namely two constituents of wood smoke – fine particles and benzo(a)pyrene – are emitted in quantities which have reportedly lead to violations of air quality standards of PM2.5 and PM10, as well as violations of the target value of BaP in wood burning communities. An interesting case study in this respect is reported for Libby, Montana, with a population of 2700, and an estimated 1500 homes heated by wood combustion (Ward et al., 2009). There, a heating season average for BaP of 6 ng/m³ (2000/2005) and a violation of the PM2.5 primary National Ambient Air Quality standard was observed. Average PM emission rates for log fired wood stoves and for boilers are comparable with emissions of coal fired stoves and boilers (Table 16). The data in Table 16 show that conventional oil (extra light) and gas stoves and boilers exhibit far lower PM10 emissions, than wood fired combustion units.

Fuel	**Single stoves**	**Boilers**
	PM10 emission kg/TJ	
wood	148	90
coal	153	94
oil	<0.5	<0.5
gas	0	0
Energiebericht 2003		

Table 16: Emission rates PM10 for stoves and boilers in Austrian homes (from Energiebericht 2003)

Elevated ambient levels for PM2.5 or PM10, as well as for BaP have been observed at many sites in Europe, USA, Australia and Asia – in most cases as a result of biomass based fuels (e.g. Ramdahl et al., 1984; Moeller et al., 1985; Sexton et al., 1984; Freeman and Cattell, 1990; Schauer et al., 1996; Zheng et al., 2002; Environment Australia, 2002; Leithead et al., 2006; Puxbaum et al., 2007; Szidat et al., 2007; Wang et al., 2008; Glasius et al., 2009; Caseiro et al., 2009; Bari et al., 2010; Schnelle-Kreis et al., 2010).

In this thesis the relationships of BaP and PM from available emission and ambient data was examined. The BaP/PM ratio can be used as diagnostic tool for estimating ambient BaP levels from wood smoke data. A further application of the BaP/PM ratio is seen in modelling tasks. The over all results of the evaluation for “all”, “European” and “Austrian” studies is shown in Table zzz. The median BaP/PM ratios obtained are 0.20 for “all” studies, 0.27 for the “European” studies and 0.24 for the Austrian studies.

	BaP/PM ng/µg
Median all	0.20
Median Europe	0.27
Median Austria	0.24

Table 17: Overview of results for BaP/PM relationships from “all”, “European” and “Austrian” studies. The PM/BaP ratio indicates the PM mass concentration in $\mu g/m^3$ expected for occurrence of 1 ng/m^3 BaP from wood combustion.

The diagnostic BaP/PM ratios have been used to estimate wood smoke BaP contributions for a set of sites where simultaneous wood smoke PM10 and BaP data were available. For the Austrian urban sites Vienna, Graz and Salzburg, the estimated contribution of BaP from wood smoke to the observed BaP levels ranged from 41-61%, and for Linz the wood smoke BaP contribution yielded 83%. For a semi rural site near Linz (Steyregg) the wood smoke BaP contribution was 78% (Table 19). According to the “Emission trends” of the Umweltbundesamt (2008b), the average share of wood smoke of total PAH emissions in Austria is 75% and the share of traffic emissions 18%. The urban sites where BaP data were available are situated at roads with heavy traffic, where the traffic influence is higher than the average from the emission inventory. For the background site, the derived BaP contribution from wood smoke is close to the emission inventory number.

Site	**Sample**	**PM10**	**“wood**	**BaP**	**BaP**	**BaP**
Austria		**µg/m^3**	smoke” **µg/m^3**	**ng/m^3**	**ng/m^3** wood smoke	**%** from wood smoke
urban sites		observed	derived	observed	contribution	contribution
Vienna RBS	PM10	33	1,7	1	0,4	41
Graz Süd	PM10	38	5,2	2,3	**1,2**	54
Salzburg RU	PM10	32	2,3	0,9	0,6	61
Linz RBG	PM10	45	3,1	0,9	0,7	83
suburban site						
Steyregg	PM10	33	2,6	0,8	0,6	78

Table 18: Compilation of determined ambient PM10 and BaP levels (annual averages). Derived “wood smoke” from literature data and derived BaP levels from wood smoke (using BaP/PM ratios from Table 13). The derived BaP from wood smoke contribution is expressed in ng/m^3 and % of observed BaP levels.

The diagnostic BaP/PM ratios have been further used to estimate violations of the EU target value for BaP due to wood smoke impact. From the available data exceedings of the BaP target value are predicted for the Austrian city of Graz, for the coastal town Aveiro in Portugal and for the region of the Hungarian background site K-Puszta (Table 20). The sites are considerably impacted by wood smoke as determined by the Levoglucosan tracer technique. From this result we can derive, that there will be many other sites in Europe, where violations of the BaP target take place. However, due to the limited availability of wood smoke or BaP data from background sites the information about the regional distribution of high wood smoke impacts is still sparse.

Site	**Sample**	**“wood smoke”** **µg/m^3** derived	**BaP** **ng/m^3** wood smoke contribution
Graz Süd, Austria	PM10	5,2	**1,2**
Graz DB, Austria	PM10	4,4	**1,1**
Aveiro, Portugal	PM10	5	**1,4**
K-Puszta, Hungary	PM10	3,7	**1,0**

Table 19: Sites with predicted violations of the EU target value for BaP due to wood smoke impact – derived from “wood smoke” data from literature data using BaP/PM diagnostic ratios. The derived BaP from the wood smoke contribution is expressed in ng/m^3.

The results of the thesis indicate that elevated PM and BaP levels from wood smoke are to be expected for densely populated rural and semi rural environments, where wood combustion is a major source of residential heat generation. The respective data for Europe until now are relatively sparse, as monitoring programs for BaP started only recently and were predominantly focused on traffic impacted urban sites. It appears that larger urban centres are not typical wood smoke combustion areas, while settlements in suburbs and rural areas seem to use fuel wood more frequently, in particular in areas with frequent temperature inversions. It can be assumed that in densely forested countries more fuel wood is used than in countries with sparse wood areas.

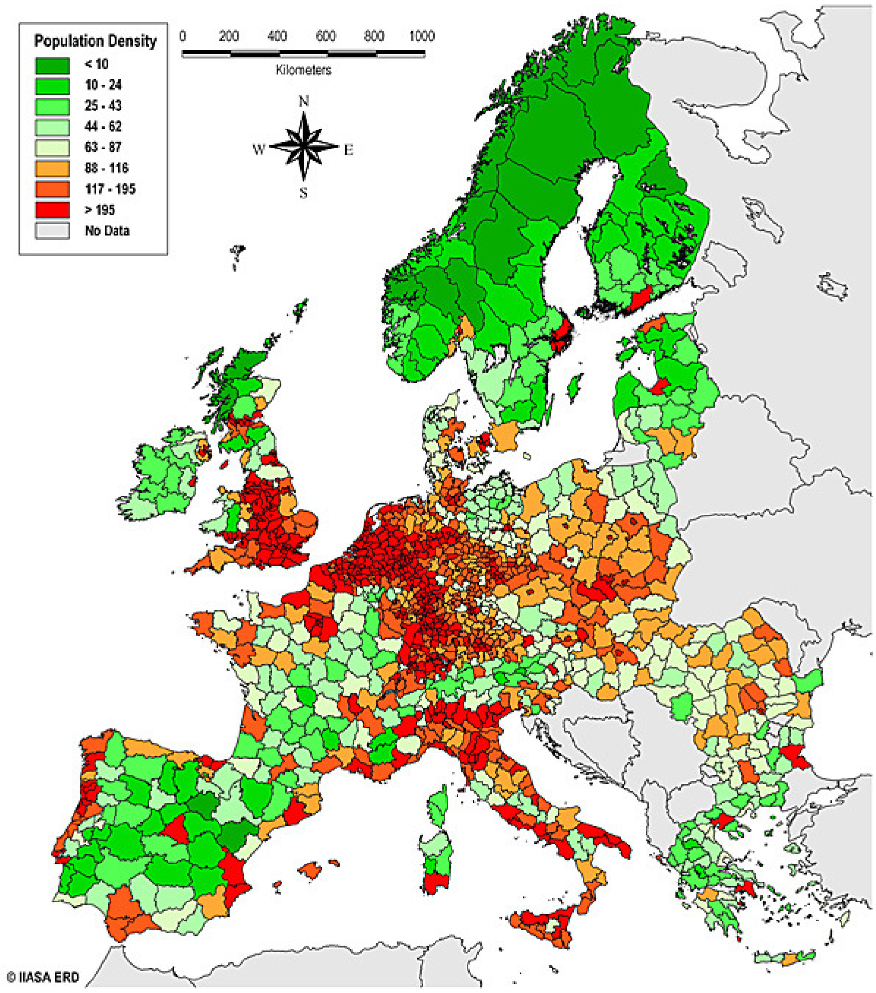


Figure 14: Population density in Europe (Source: European Rural Development (ERD) Project. Copyright © 2002 IIASA

8 CONCLUSION

Considering the rising fuel costs and concerns with global warming, it is apparent that wood combustion for energy use will increase: wood is not related to international energy market changes, and is CO_2 neutral (Reitze et al., 2008). Reitze et al. suggest that even with unchanged energy policies, wood use will probably double in Austria, Germany and Switzerland. A more recent study forecasts a 50% growth for Europe's biomass capacity until 2013, pushed by CO_2-reducing policies; the study estimates that between 2008 and 2013, the number of biomass plants will double to a total of 1050, while generating ability will also increase by 50% (reaching approximately 10,000MW). In fact, in 2009, biomass capacity increased by 40% in Europe, largely due to new biomass plants in Germany, Austria, and Scandinavia. Germany has a powerful feed-in tariff program that was an important contributor to small scale biomass efforts (Mackinnon, 2010). However, more work is needed to improve the knowledge basis on forest biomass and on biomass emissions.

With a predicted biomass capacity growth, there is the concern of biomass emissions as discussed in this paper. Decreasing biomass combustion polluting emissions will provide social and economic advantages: less infrastructural and financial strain on the healthcare system (due to reduced cases of illnesses attributed to smoke inhalation) and fewer lives lost to illnesses caused by air pollution due to biomass combustion. Thus it will be required to regulate decreasing emissions from residential heating, in particular woodstove combustion, in particular for new units.

In order to find appropriate solutions to biomass emission controls, there needs to be a shift in some current approaches which deem biomass a relatively straightforward fuel, where the important factors are moisture content and stage of processing (logs, chips, pellets). Emission concentrations are then based on the fuel, technology and combustion conditions. Grand-scale movements can be efficient, but in order to foster interest and cooperation and replace old stoves, financial incentives should be instated, in parallel with education campaigns that inform people about the health dangers of wood smoke inhalation. The focus is now to achieve low emissions, and a balance between appliance efficiency, dependability, and price.

There must be a separation between old and new biomass combustion appliances, especially since new biomass combustion technologies emit less organic aerosols (due to “almost complete burn-out”) compared to their older counterparts. Therefore, replacing old systems with new ones can result in a large reduction of aerosol emissions (Biomass Austria, 2005), although lowest particles emissions are taking place in gas and extra light oil burning appliances. Also, emission records for biomass combustion need to be investigated in more detail according to fuel types and technology available.

Based on the experiences of this study it is recommended to perform a retrospective screening study of PM2.5 or PM10 filter samples from urban fringe and densely populated rural sites for the wood smoke tracer levoglucosan and for BaP, which would allow an insight about the geographical extension of potential violation areas of the BaP target value from wood smoke impacts. Figure 14 shows the population density in Europe. It is obvious from this chart that a large fraction of the population lives outside the major cities. In those areas, namely in the forest rich countries, a screening for wood smoke impacts is recommended.

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