## Toxic Emissions from Solid Fuel Combustion in Small Residential Appliances

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### Introduction

Combustion of solid fuels is a source of pollutant emission to the atmosphere. Low combustion efficiency (usually not exceeding 50% average per year), poor fuel quality and no or little cleaning of exhaust gasses result in higher emissions. Especially the combustion of coal either in residential heating appliances (i.e. stoves, furnaces) or boilers of low power is widely considered to be of the most pollutant emitting sources.

Research carried out in the past few years proves that besides significant emission of CO,  $SO_2$ ,  $NO_x$  and particles, these sources are also responsible for significant amount of organic pollutants (TOC), including polycyclic aromatic hydrocarbons (PAH), dioxins and furans – PCDD/F, and VOC such as aliphatic hydrocarbons, benzene and its derivatives (BTX), aldehydes and ketones, but also phenol and its alkyl derivatives, heterocompounds of nitrogen and sulphur etc., and heavy metals <sup>1, 2, 3</sup>.

The negative impact from the use of low efficiency heating devices is multiplied by the combustion of fuels of poor quality with significant sulphur and ash contents, low calorific value and coal sludge. This is especially true for the Central and Eastern European Countries (CEEC) where the cold climate, ease of access to coal and poor economic conditions make the heating of residences by coal an attractive option. At the same time biomass becomes a more and more popular fuel used in residential sector, due to the strategy to achieve reduction of  $CO_2$  in same countries. But also installations burning biomass are often characterized by higher emissions of particulates and related pollutants.

### Use of Solid Fuel Combustion for Residential Heating

Solid fuel use in the residential sector is much higher in the CEEC than in the EU - 15 both on per-capita basis and also in absolute terms. On per capita basis the average consumption of wood is three times higher and that of coal almost ten times. The use of solid fuels in Poland is the most common, where per capita twenty times more coal is used than in EU. On the other hand in Latvia the per capita wood consumption is still ten times higher than EU-15 average (Table 1).

Also the kind of heating appliances is different. On average 38 % percent of the flats in Candidate Countries and New Member States are not equipped with central heating systems varying from 71 % in Bulgaria to a mere 3 % in Czech Republic (1996 data). In the EU-15 the share of flats without central heating is 23 % (1995 data). Most of the non-central heating flats in the EU-15 could be found in countries with warmer climate and lower energy demand for heating like Greece, Spain and Portugal. In EU–15 countries with similar climate as the CEEC the share of central heating is significantly higher.

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Data on solid fuel consumption and non-central heating share presented in the table 1 cover a period of almost a decade ago <sup>4</sup>. Their advantage is that they were obtained by a harmonised methodological approach. Moreover it has been shown that the consumption of solid fuels, especially firewood, was usually not adequately addressed by the statistical report based on the supply side data collection, since especially the self and non-commercial supply of wood was not taken into account in many cases.

	BG	CZ	EE	HU	LT	LV	PL	RO	SI	SK	CC*	EU°
coal [GJ / capita]	4.3	5.1	1.9	3.7	2.4	1.0	20.3	0.5	1.4	4.6	9.1	0.9
fuel-wood [GJ / capita]	4.2	5.6	18.3	5.1	5.9	15.4	3.3	8.6	7.6	4.1	5.6	1.9
share of flats without central heating [%]	81	3	34	44	8	30	33	60	14	8	38	23

\*weighted average for Central and Eastern European Countries, 1996 data

weighted average for EU-15 without Italy, 1995 data

Besides the combustion of licensed fuels also uncontrolled burning of waste might be an important issue especially in the CEEC and in societies with poor economies either as cocombustion in solid fuel-fired appliances or the so-called open burning. Co-combustion of waste might be motivated by savings in partly substituting licensed fuels and/or resulting from lower level of awareness on the consequences of such a bad practice. The open burning of waste occurs mainly in the rural areas with less developed waste collection systems.

Sound data on these activities are difficult to obtain, however some structural differences point out that these activities might be of much greater relevance in the CEEC than in EU-15 due to:

- higher share of solid fuel-fired appliances which offers more possibilities for waste cocombustion,
- higher share of low-income households which might consider energy use of the waste,
- less developed waste collection systems, as for instance in Latvia only about 50-60% of household waste is collected and land filed<sup>6</sup>.

The above-mentioned differences as well as the wide-spread use of older technologies in the industrial sector led the Council to note the need to further investigate the situation in regards to dioxins. This was reflected in the Community Strategy on Dioxins, Furans and PCBs<sup>7</sup> and was already addressed in the communication on its implementation<sup>8</sup> as well as on a separate EUR report <sup>4</sup>.

## **Emission Inventories from Small Combustion Installations**

The contribution of emissions from small combustion installations to the total emissions varies and depends on pollutants type and given country. A very important role is played by the emissions from small residential installations which are typically responsible for more than a third of the **total particulate matter** emissions of stationary combustion <sup>9</sup>,<sup>10</sup>,<sup>11</sup> but in some countries this sector may even dominate. For example in Austria in 1995 more than 70% of PM emissions from stationary combustion are thought to have originated from this source<sup>12</sup>.

The contribution of fuel combustion in commercial, residential and other small capacity installations to the total **heavy metals** emission in Europe in 1990 was for As 12.4%, for Cd 15.9% and for Hg 27.8% <sup>13</sup>.

Also emissions of **PAH and PCDD/F** from those activities are significant. For instance, residential use of solid fuels and biomass accounts for about half of the emissions of polycyclic aromatic hydrocarbons <sup>14</sup> and one third of dioxin emissions in the EU <sup>15</sup>. Those are characterized by seasonal variations, as it was reported that emission of B[a]P in winter is 10% higher than in summer <sup>16</sup>.

Many countries using coal (but also biomass) as a major part of domestic and commercial heating requirements have serious air pollution problems, such as example Austria and Poland. In Austria, the residential combustion is already the most significant source of dioxins <sup>15</sup>. In Poland the TSP emissions from small combustion sources is 35% of the national total emissions, and up to 90% of the total TSP emissions from combustion activities <sup>11</sup>. It was further reported that in Poland the main source of PCDD/F (47% of national total) and PAH emission (87% of national total) are non-industrial combustion plants (residential, district heating, agriculture, forestry). The share of heavy metals emissions such as Cd, As, Cr, Cu, Ni, Zn due to high emissions of TSP is also higher (respectively: 55%, 36%, 27%, 25%, 50%, 30%). In general those sources have a more important contribution to the above-mentioned pollutants where a higher share of solid fuels exists in the fuel mix of the residential sector.

Furthermore, due to the particularly low height of the stacks used in such installations (usually not exceeding 10 m), the emissions from such residential heating appliances, especially in highly urbanised areas, result in high concentration of pollutants at ground level and thus more dangerous for the human health, both by direct inhalation, as well as by ingestion with contaminated food. This particularity makes this sector responsible for the "low-height emissions" in urban centres where such appliances are used.

The effect of environment pollution caused by coal combustion for residential heating is particularly obvious in large urban centres where about 60% of annual pollutants emission derives from municipal sources, while it can reach 90% during the winter time. This relates especially to such harmful pollutants as CO, particulate matter (TSP, PM) as well as the above-mentioned organic pollutants, including PCDD/F and PAH.

## Influence of combustion technology and type of fuel on emissions

It is clear that the kind of technology employed significantly influences the resulting emissions. This can be easily seen in the figures 1 and 2, where the emission factors from stoves, boilers manually fed and boilers, which are mechanically fed, are compared for both coal and wood combustion. Stoves, which have the lowest efficiency and poorly organised combustion technology, have the higher emission factors, not only for CO, but also for VOCs, PM, PAHs and PCDD/Fs, followed by manually fed boilers. In fact, the kind of uncontrolled process, represented by those devices, is the most emitter producing. In comparison, mechanically fed boilers have relatively low emission factors.

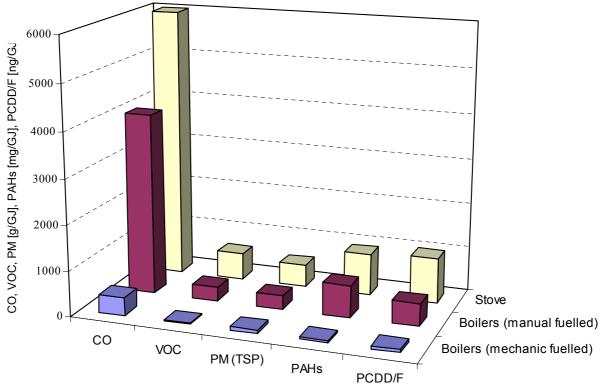
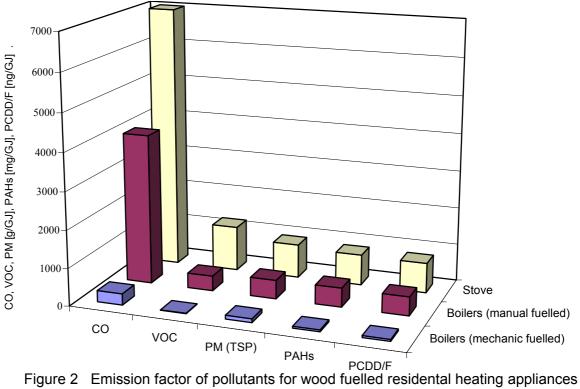


Figure 1: Emission factor of pollutants for coal fulled residental heating appliances



## Profiles of toxic emissions

The technologies of solid fuels (coal, biomass, refused derived fuels, and so on) combustion by means of fixed bed are carried on under conditions far from optimum (by means of stoves, furnaces and old design devices) which results in incomplete combustion and in consequence they are the source of not only classic gaseous combustion pollutants (NO<sub>x</sub>, SO<sub>2</sub>, CO, CO<sub>2</sub>, TSP, PM<sub>10</sub>, PM<sub>2.5</sub>, HM) but also of the most hazardous pollutants – organic compounds such as the persistent organic pollutants (POP – PCDD/F and PAH) and other toxic atmosphere organic pollutants (TOAC as sum of VOC, SVOC; POP); volatile organic compounds – VOC, and semi-volatile organic compounds SVOC and high volatile organic compounds (HVOC – are comprised of more than 4-rings PAH). The VOC responsible for generation of troposphere ozone (POCP), are aliphatic hydrocarbons  $C_1$ - $C_{10}$ , BTX, other aliphatic derivatives of benzene and ox-compounds. The group of SVOC includes aromatic hydrocarbons, together with 16 PAH by EPA, among which are listed benzo(a)pyrene and hydrocarbons of 3-, 4-, 5- rings with carcinogenic and mutagenic properties. There are important benzene derivatives of anthracene, for instance dibenzo(a,h)anthracene, which toxicity factor is higher than for benzo(a)pyrene.

Such compounds are present in the combustion gases both in the gaseous phase, as well as on particles. They are especially dangerous in this last form because inhaled particles can cause big effects to the human health.

From the speciation of selected NMVOCs for both coal and wood pellet combustion in conventional stoves it is clear that each fuel has a characteristic fingerprint (see Figures 3 and 4 below). Wood pellets for instance, emit higher levels of benzene, toluene and xylene (BTX) to the atmosphere when compared to coal combustion. The difference is also evident on the amount and profiles of PAHs emitted by those fuels as seen in the figure 4 below.

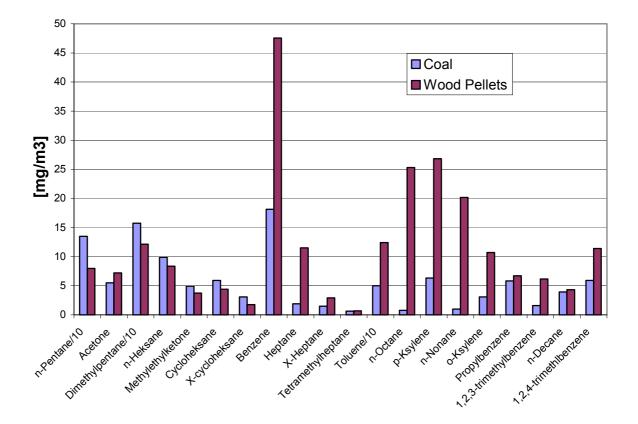


Fig. 3 Profiles of selected NMVOCs from combustion of coal and wood pellet in conventional stove<sup>17</sup>

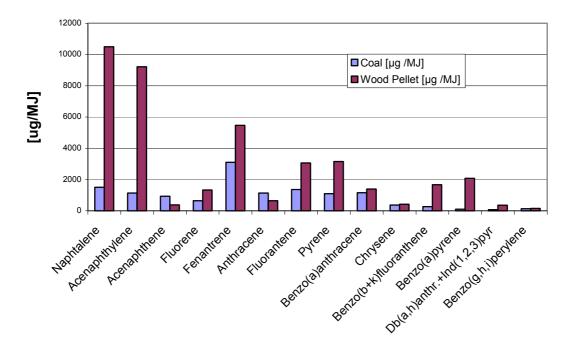


Fig. 4 Profiles of 16 PAHs from combustion of coal and wood pellet in conventional stove <sup>17</sup>

## PCDD/Fs emissions from solid fuels combustion

Little research has been devoted up to now on the measurement or estimation of dioxin emissions from solid fuel combustion in small residential heating. However, there are strong indications that there is a high potentiality for production of dioxins under certain circumstances.

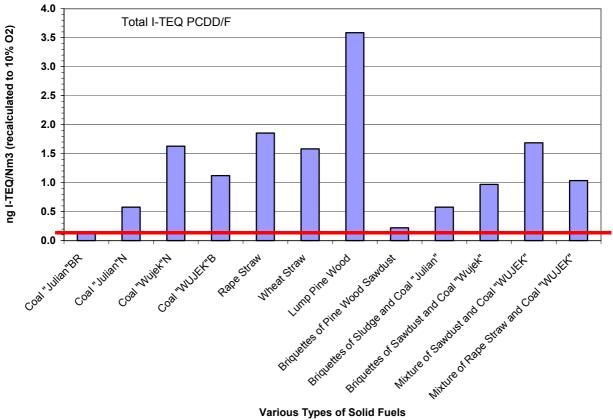
The high emission of PCDDs/Fs from coal combustion, especially polish coal, is discussed in both Austrian and German studies <sup>15</sup>, <sup>18</sup>. The research reveals also much higher dioxins emission from combustion of coal imported from Poland, which ranges from 108,5 up to 663,9 µg I-TEQ/tone of combusted fuel <sup>18</sup>. These amounts are comparable to emissions observed during the tests carried out in Krakow, including co-combustion of coal and wastes <sup>19</sup>. Relatively high dioxins emission from the samples of Polish coal revealed by Austrian and German works can be derived from relatively high chlorine contents in Polish coals, that usually ranges from trace quantities up to 0,4%, or even up to 1,5%.

Between 1999 and 2002, within the scope of research projects budgets funded by EC (INCO-COPERNICUS programme) combustion tests using several solid fuels: two coals with different chlorine contents, different types of wooden biomass and mixtures of coal and biomass, either in briquette form and a two-component mixture were carried out <sup>1</sup>,<sup>20</sup>. These fuels were combusted by means of a chamber water-boiler of 30kW capacity, with combustion at the bottom part of the bed (co-current combustion with distribution of air supplied for the process).

For the purpose of comparison emissions were also measured from the combustion of the same type of coals in a retort boiler. Retort boiler of stoker type with continuous, automatic fuel supply and controlled amount of air supplied to combustion chamber, with applied technique of combustion in the upper part of the bed, co-current combustion, can be listed among the most modern and the most effective low capacity boilers designs, enabling "clean combustion of coal" technology.

The test results are presented on Figure 5. As can be seen from the figure, the PCDD/Fs emission factors, expressed as ng I-TEQ/m<sup>3</sup>, are in every case (except of retort boiler)

significantly higher than allowable value specified by the 2000/76/EC Directive for waste incineration plants <sup>21</sup>, which is equal to 0,1 ng I-TEQ/m<sup>3</sup>.



Various Types of Solid Fuels

Fig. 5: PCDD/Fs Toxic Equivalent values for different types of fuel – technique combinations. R means Retort Heart Boiler, B - coal of bean size, N - coal of nut size. The red line indicates the limit for waste incineration.

The amount of PCDDs/Fs emitted from chamber boilers irregardless of the combusted fuel type was generally at the same level of decimal units of µg I-TEQ/tone. Combusted biomass, either wood or agricultural, emits significant quantities of dioxins (from 2 up to 33 µg l-TEQ/tone for wood biomass, from 12 up to 13 µg I-TEQ/tone of straw). However, it should be noted that the used boiler was not adopted for biomass combustion.

In the case of hard coal, a fuel of lower (2,5 times) chlorine contents (Julian coal) emitted more than 3-times less PCDDs/Fs.

A decrease of grain size also lowered dioxins emission to about 30%. In the case of briquetted fuels dioxins emission has been generally lower as it was also observed by German studies <sup>15</sup>. While the above-mentioned effect has not been observed for the mixtures of coal and straw; in this case an addition of approximately 10% (m/m) of straw or sawdust did not cause significant differences by dioxins emission.

To summarise, the current research indicates that the influence of chlorine contents is observed on individual fuel groups but a significantly higher impact have combustion process organization, chemical composition of fuel and homogenisation of fuel properties and finally chlorine contents.

# Future

In the future, the amount of emissions from this sector are expected to decrease, both due to improving economic conditions and therefore use of better fuel quality and upgraded solid fuels and biomass, as well as due to improvement of combustion techniques. Conversion to central heating fuelled with gas or oil, biomass introduction instead of coal and increase of energetic efficiency of new type boilers and furnaces fuelled with solid fuels especially with coal can all improve the emissions.

As an example, it was already presented that the retort hearth boiler produces significantly lower amounts of PCDD/Fs than other more classical types of boilers. But this is also true for other pollutants. The reduction of more classical pollutants by the replacement of old stoves, by more advanced technology, was estimated to be close to 99%, as can be seen in the Figure 6 below.

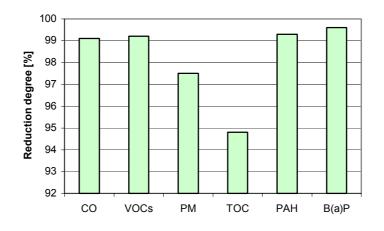


Fig. 6: Reduction of pollutants emission due to replacement of old stove by the advanced technology (retort hearth boiler) <sup>3</sup>.

## Conclusions

Small combustion installations, especially those intended for residential heating, have a potentiality for emitting high amounts of atmospheric pollutants and in particular toxic substance, such as PAHs and PCDD/Fs. The combination of old technologies with low quality fuels is especially bad for the environment and human health, but the advent of new technologies and use of better fuel hold a promise for the future.

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