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INVESTIGATION OF AIR POLLUTANTS FROM RESIDENTIAL HEATING

DÓRA MENTES[1]–HELGA KOVÁCS[2]–
GÁBOR NAGY[3]–CSABA PÓLISKA[4]

Beside of industry, agriculture, and transport, primarily the solid combustion plants used by households are responsible for the emission of gaseous and solid pollutants in ambient air. Thus, the actual reduction of air pollutant concentration also can be achieved by regulating of residential heating. New, solid-fired, individual room heating systems will have to meet stricter emission requirements from 1 January 2022. People in poorer regions are not able to buy new equipment, so in most households old equipments in poor condition remain in use. In addition, the population – albeit illegal – in most cases also burn the combustible waste generated in the households in their stove or boiler, which further increases the amount of air pollutant emissions. So far the authorities cannot be to clearly demonstrate the combustion of waste from the flue gas and residual ash, because these methods are expensive and hardly unworkable, as the authorities find it difficult to get into private homes.

Keywords: the limit value of air pollutants, emissions factors of combustion equipment, residential heating

INTRODUCTION

The harmful effects of air pollution on health and the environment are well known. In Europe, greenhouse gas emissions have declined significantly over the past decades, resulting in improved air quality in the region, but still the concentrations of air pollutants are high [1]. According to World Health Organization (WHO) and European Economic Area (EEA), this reduction in total emissions of air pollutants does not automatically lead to a similar reduction in air pollutant concentrations [2].

Most of European population live in a place – mainly in cities – where the air pollution exceeds the air quality limits at certain regular intervals: the NO_2 and particulate matter pollution pose a serious health risk [1]. The laws do not place any

[1] Department of Combustion Technology and Thermal Energy, University of Miskolc
H-3515 Miskolc-Egyetemváros, Hungary
tuzdora@uni-miskolc.hu

[2] Department of Combustion Technology and Thermal Energy, University of Miskolc
H-3515 Miskolc-Egyetemváros, Hungary
kovacs.helga@uni-miskolc.hu

[3] Department of Combustion Technology and Thermal Energy, University of Miskolc
H-3515 Miskolc-Egyetemváros, Hungary
nagy.gabi@uni-miskolc.hu

[4] Department of Combustion Technology and Thermal Energy, University of Miskolc
H-3515 Miskolc-Egyetemváros, Hungary
tuzcsaba@uni-miskolc.hu

emphasis on reducing emissions where people are most affected by air pollution and where the highest concentrations are [2].

The European Union has initiated infringement proceedings against Hungary for persistent exceedances of particulate matter levels and NO_2 levels. NO_2 is mainly presented in emissions from industry, transport, domestic heating, and agriculture. Limit exceedances occur predominantly during the heating season and are mainly resulted of the solid firing by the public [3].

The effect of air pollutant from household solid combustion plants is influenced by three circumstances:

- condition of the combustion plant and chimney,
- quality of fuel used,
- firing method applicable to the equipment.

In the past decade, especially in the poorer sections of society, many households have returned for the use of solid fuel equipment instead of their gas-fired heating systems because of economic reasons [4]. The population has no acceptable information and knowledge of how and what way operate their boilers correctly. Regardless of the age and type of the boilers, in the absence of proper knowledge, combustion is imperfect, which further increases pollution.

In poor households, as they are unable to buy the right quality fuel, besides solid fuels the household wastes are also thrown into the boiler. In the case of incomplete combustion, along with the increase in the amount of harmful flue gas components and flue dust, also the growth the amount of polycyclic aromatic hydrocarbons (PAH) content have to be taken into account.

1. QUANTITY AND LIMITS OF AIR POLLUTANTS IN HUNGARY

Free atmospheric tests are called immission measurements, or air quality tests too, and investigation of air pollutants from point sources are called emission measurements. [5].

Limit values for air pollutants affecting ambient air quality are given in "4/2011. (I. 14.) Decree of the Ministry of Rural Development on limit values for air load level and emission limit values for stationary air pollution point sources" *(Table 1)*.

The concentration of benzo[a]pyrene should be determined as the annual average of the total quantity in the PM10 fraction [6]. In 1984, the United States Environmental Protection Agency (EPA) appraised the risks posed by PAH mixtures with the assumption that all carcinogenic PAHs have the same environmental effects as benzo[a]pyrene. [7]. Several studies disagree with the fact that benzo[a]pyrene is used as an indicator of the carcinogenic risk of atmospheric polycyclic aromatic compounds, as benzo[a]pyrene amount is highly more dependent on the sunlight, temperature, and the amount of NOx, SO_2, O_3 than the amount of other polycyclic aromatic hydrocarbons [8], [9].

The CO and NO_2 concentration in the atmosphere showed a slight decrease between 2013 and 2016 *(Figure 1)*. PM2.5 and PM10 have remained almost constant since 2013, while CO_2 emissions have increased since 2014.

Table 1

Limit values for pollutants emitted to ambient air in Hungary [6]

Pollutant	Limit values [µg/m^3]		
	24 h PM10	**Annual PM10**	**Annual PM2.5**
PM10/PM2.5	50 up to 35 times a calendar year	40	24
	1 h	24 h	Annual
NO_2	100 up to 18 times a calendar year	85	40
	1 h	24 h	Annual
CO	10,000	5,000	3,000
	24 h	Annual	
Benzo[a]pyrene	0.001	0.0012	

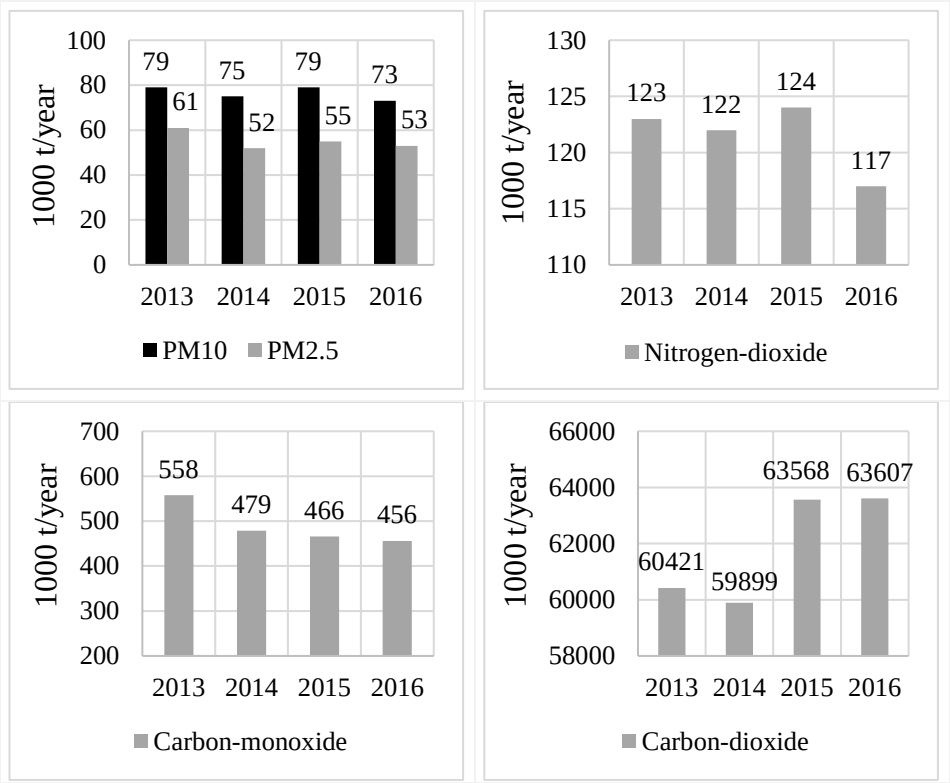

Figure 1

Quantity of pollutants emitted into the air in Hungary [thousand tons/year] [10], [11]

In 2016, households released the largest share of CO_2 (37% of total emissions), followed by the energy sector (23%). Nearly two-thirds of PM10 is emitted into the atmosphere from residential heating *(Table 2)*. More than 85% of the total emissions of CO and PM2.5 come from the households. Households, agriculture, and transport are equally responsible for emissions of NO_2.

Table 2

Percentage distribution of air pollutant emissions from Sectors in 2016 in Hungary [10], [12], [13], [14], [15]

Sectors	**CO_2**	**CO**	**PM10**	**PM2.5**	**NO_2**
	[m/m%]				
Households	36.58	86.99	65.73	87.62	20.91
Energy sector	23.27	1.72	0.35	0.41	8.75
Manufacturing industry	17.56	5.12	2.86	2.92	11.28
Transport	6.94	2.14	1.98	2.11	19.09
Agriculture	3.82	1.07	14.06	2.54	22.38
Building Industry	1.67	0.43	10.02	1.79	3.33

According to the website of the National Air Pollution Network, in Hungary 51 monitoring and 3 background stations operate within the framework of the National Air Pollution Measurement Network. Most environmental authorities have mobile stations, including the B.-A.-Z. County Environmental Authority, which has two mobile stations. Taking into consideration all air pollutants, the number of stations with qualification "appropriate" and "contaminated" ratings increased in 2017 compared to 2016, and the number of "good" qualificated stations decreased. Since 2015, the "heavily polluted" rating has not been given to a monitoring station [16].

The exceedance of average annual limit of PM10 concentration was measured at one station in 2015, while in 2017 at three stations, of which two stations are in the Sajó Valley (Miskolc and Sajószentpéter). In the case of NO_2 concentration, starting from 2013, two stations continuously exceed the annual limits (Pécs, Szabadság Road; Budapest, Széna Square) [16]. *Figure 2* shows the annual average values of air pollutants in Miskolc.

In addition to the annual limits, many daily and hourly limits are determined. As stated in the regulation, the PM10 concentration in a calendar year should not exceed the daily limit more than 35 occasions. Between 2014 and 2018, air pollution values measured at Miskolc stations exceeded the daily concentration limit more than 35 occasions *(Table 3).* In the most cases there are limit exceedances during the heating season in winter, which can be explained by the emission of solid-fired equipment [20].

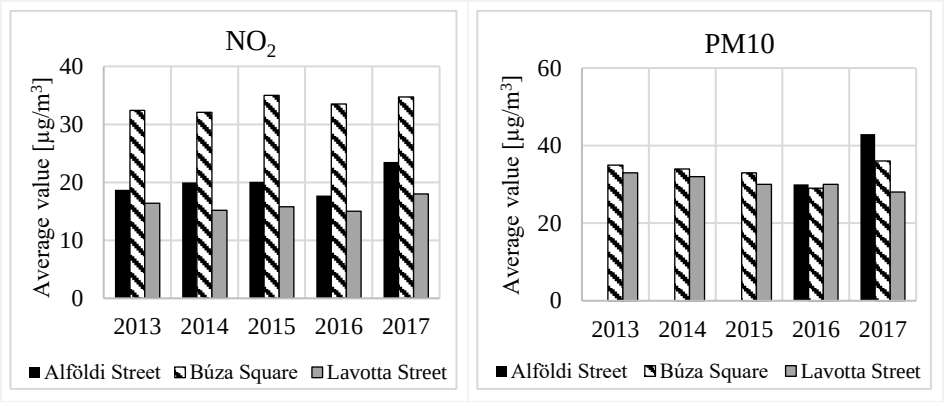

Figure 2

The annual average values of air pollutants in Miskolc [µg/m³] [17], [18], [19]

Table 3

Number of daily limit exceedances in Miskolc 2014–2018 [21]

Pollutant	Location	Number of daily limit exceedances [db]				
		2014	2015	2016	2017	2018
PM10	Miskolc, Alföldi Street	10	19	55	68	63
	Miskolc, Búza Square	61	55	40	68	84
	Miskolc, Lavotta Street	59	40	40	42	32

2. THE RELATIONSHIP BETWEEN RESIDENTIAL SOLID COMBUSTION AND THE REGIONS

In Hungary in 2011, slightly more than 45% of households used boilers, while 37% of households have been heated by convectors or stoves *(Figure 3)*. Heating types can be further grouped according to their fuel.

In the case of boiler and convector/stove heating type, if we rank the fuels according to the frequency of their use, the gas heating is in the first place, a second most common type of heating is mixed with gas and wood, and the only wood-firing is in the third place [22].

There is a strong correlation between residential solid fuel heating and regional development levels. Out of 174 districts, more than three-quarters of households in one of the district and more than half of households in 19 districts are exclusively heated with wood and in 22 districts more than three-quarters of households are heated (at least partially) with wood *(Figure 4)* [23].

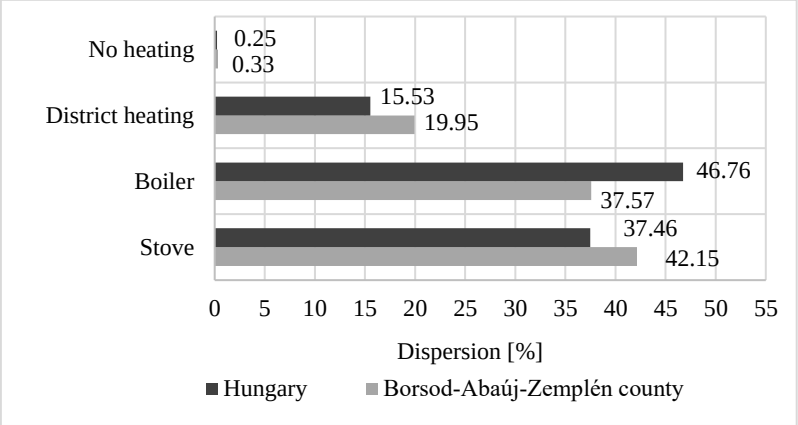

Figure 3
Distribution of households by type of heating in 2011 [22]

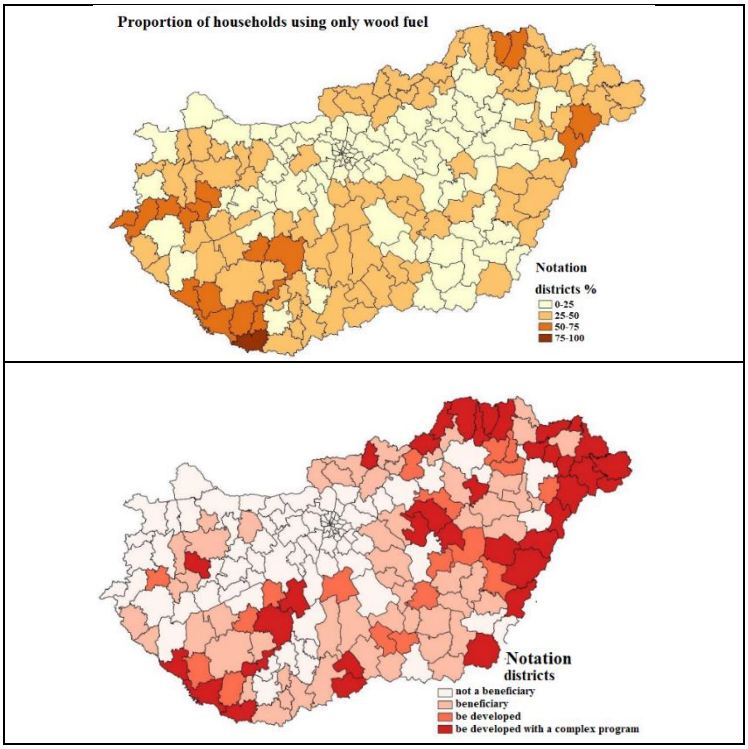

Figure 4
In Hungary, the ratio of households using solid fuel (upper), beneficiary regions classified by the 290/2014. (XI. 26.) Government Decree (lower) [23]

The complex indicator measuring the level of development of districts takes into consideration both socioeconomic and infrastructure development. Comparing the map showing the concentration of households which are heating with wood, with a map of complex indicators measuring the level of infrastructure development, it can be seen that among the 19 districts where more than 50 percent of the households are heated with wood, it is among the 11 most disadvantaged districts. Thus, the proportion of households heating with wood in disadvantaged districts is higher [23].

3. EMISSIONS FROM RESIDENTIAL HEATING, THE NEW RESTRICTION

While emissions from industry can be measured and limited, the control of citizens mixed-fired equipment is difficult. Authorities are not able to act against illegal burning, arguing that there is no device in their hands to deal with the situation. As a result, the European Commission has issued the implementation of Commission Regulation (EU) 2015/1188 implementing Directive 2009/125/EC of the European Parliament and of the Council with regard to ecodesign requirements for local space heaters. According to the expectations with this limitation, the number of emissions from households will be decreased and it will become much more controllable. However, households that have old types of firing equipment are typically financially incapable of being buying a new one. Separate room heating systems operating with solid fuel from 1 January 2022 must meet the requirements of the following table *(Table 4)*, which include limit values for seasonal room heating efficiency and pollutant emissions.

Table 4

Limit values for the environmentally conscious design of solid fuel installations [24]

	Efficiency (min) [%]	**Emission of pollutants (max)**			
		PM [mg/m^3]	**Gaseous organic compounds [mg/m^3]**	**CO [mg/m^3]**	**NO_x [mg/m^3]**
open combustion solid fired individual room heating equipment	30	50	120	2,000	200
working with solid fuel (which different from pellet) individual room heating equipment with a closed combustion chamber	65	40	120	1,500	200
working with pellet individual room heating equipment with a closed combustion chamber	79	20	60	300	200

The European Environment Agency's 2016 guidebook summarized the emission values in case of different fuel *(Table 5)* and the amount of pollutants emitted by different combustion plants in case of wood firing *(Table 6)*. The values in the table are converted from GJ/g to mg/m^3. The air pollutants in the tables are emitted into the atmosphere during optimum operation of the combustion plants.

Table 5

The emission factor of stove [16]

Fuel	Pollutant				
	PM [mg/m^3]	NOx [mg/m^3]	Benzo[a]pyrene [µg/m^3]	CO [mg/m^3]	SO_x [mg/m^3]
Firewood	73.87	2.46	5.96	196.99	0.54
Lignite	37.51	4.17	10.42	208.39	37.51
Coal	18.52	6.04	6.04	80.53	18.12

One of the biggest advantage of wood against coal is that it contains just a little amount of sulfur or not at all. Therefore, the resulting SO_x emissions will also be significantly lower. In terms of unit energy, the emission of the particulate matters is the highest during wood burning.

Table 6

Emission factors of different combustion plants in case of wood firing [16]

Equipment	Pollutant				
	PM [mg/m^3]	NOx [mg/m^3]	Benzo[a]pyrene [µg/m^3]	CO [mg/m^3]	SO_x [mg/m^3]
Open fireplace	81.75	2.46	5.96	196.99	0.54
Traditional stove	73.87	2.46	5.96	196.99	0.54
Traditional boiler	46.79	3.94	5.96	196.99	0.54
Energy saving stove	36.94	3.94	5.96	196.99	0.54
Modern/Eco-labeled boiler	9.26	4.68	0.49	98.50	0.54
Pellet boiler	5.91	3.94	0.49	14.77	0.54

In *Table 6*, the fuel is the same in every case and the types of combustion equipments are varied. *Table 6* shows that the use of more modern technologies can significantly reduce emissions of both particulate matter and benzo[a]pyrene.

In the literature there are a lot of information about which parameters in the firing process can cause to the formation of air pollutants *(Table 7)*. Increased emissions of CO were found at high excess air ratios. This is probably a result of the cooling of the combustion process. The enhanced emissions of CO were accompanied by emissions of other unoxidized components: total organic carbon, and polycyclic aromatic hydrocarbons [25], [27]. To maximize combustion efficiency and minimize PAH emissions, the fuel moisture content should be low [29].

The mass concentration of particles was larger in the worst old-type case. From the number and mass size distributions, it can be concluded that the emission of sub-micron particles is enhanced by poorer combustion conditions [25]. The high ash content of the fuel leads to the formation of more particulate matter [30].

It was also observed, that during the firing phase and after fuel adding PM size distribution changed slightly to bigger size fraction. This is probably caused by an insufficient supply of air and insufficient mixing of air and fuel [26].

CO and particulate emissions composition are mainly influenced by fuel such as the households waste and the wood type, i.e. structure, elemental composition, water or ash content[26], [28].

Table 7

Effect of firing parameters on emission factors

Characteristics of the equipment	**Fuel**	**Moisture content [m/m %]**	**PM [mg/m³]**	**NOx [mg/m³]**	**PAH [mg/m³]**	**CO [mg/m³]**
Old type boiler [25]	firewood	15	17.24	3.55	n/a	256.09
	firewood	15	108.35	1.38	3.15	807.67
Modern type boiler [25]	firewood	15	1.33	6.16	0.01	34.82
	firewood	26	1.21	5.37	0.01	24.53
	firewood	38	4.18	4.75	0.14	177.77
Old type tile stove [26]	Alder and MSW	14–18	160.34	386.20	0.87 *µg/m³	4,640
11 kW Pellet stove [27]	firewood	7.9	n/a	6.83	0.02	1,288.80
	firewood-household waste	8.5	n/a	14.73	0.08	1,098.26
	demolition and construc-tion wood waste-house-hold waste	4.6	n/a	90.36	0.02	339.85
Logwood stove 6 kW (primary and secondary air) [28]	briquette standard test	7.7	150.9	176	n/a	1,331
	beech standard test	6.5	111.4	155	n/a	2,779
	oak standard test	9.7	107.3	166	n/a	2,948
	spruce standard test	8.5	156.6	96	n/a	2,240
	spruce standard test	8.5	124.2	87	n/a	2,161
	spruce low air	8.5	312	86	n/a	2,841
	spruce high fuel	8.5	118	80	n/a	1,989

CONCLUSION

Most of European population live in a place – mainly in cities – where the air pollution exceeds the air quality limits at certain regular intervals: the NO_2 and particulate matter pollution pose a serious health risk.

The biggest emitters of air pollutants are the industry, the agriculture, the transport and the households. Regardless of the quality of the combustion process, solid powder and polycyclic aromatic compounds are formed during solid firing in the household. The quality of combustion is strongly influenced by the quality of the fuel used, the type and age of the equipment, and the user's knowledge of the use of the equipment. As soon as the quality of the firing deteriorates due to any parameter, the number of air pollutants emitted into the atmosphere also increases.

This is exacerbated by the fact that in most cases, households with solid combustion plants (albeit illegal, though) are also burning household waste. There is not a proper way for authorities to clearly demonstrate the combustion of waste from flue gas and residual ash yet, therefore authorities are not able to act effectively against it. As a result, the European Commission has issued the implementation of Commission Regulation (EU) 2015/1188 implementing Directive 2009/125/EC of the European Parliament and of the Council with regard to ecodesign requirements for local space heaters.

Emissions would be reduced not only by modernizing the equipment but also by calling for the correct use of combustion equipment and by developing the flue gas cleaning equipment for residential use.

A further direction of the research is to find a connection between the amount of gas and solid air pollutants in the flue gas, the components remaining in the ash and the fuel used. Hopefully, with the help of the connection found, it would be easier to detect illegal waste combustion.

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