# AN ASSESSMENT OF POP RELEASES FROM BIOMASS COMBUSTION IN FRENCH UTILITY BOILERS

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## Introduction

Greenhouse gas (GHG) emissions, including $CO_2$ emissions, from renewable energy sources are either low or zero. In July 2005, a French law has set ambitious objectives for renewable sources of energy through the implementation of an Energy Policy Orientation Programme (loi de programmation et d'orientation sur l'énergie - loi POPE). This law requires a significant increase in biomass uses, since it aims at increasing by 50% the share of renewable thermal energy between 2005 and 2010. In addition, since this French law was voted, the European Union has published its renewable energy roadmap which sets out a long term vision for renewable energy sources in the EU. It proposes that the EU establish a mandatory target of 20% for renewable energy's share of energy consumption in the EU by 2020. Among the three energy sectors covered by this EU communication, the heating and cooling sector is the most challenged one by this overall target. Reaching this target means that the contribution from renewable energies in this sector should more than double, compared with the current share of 9%[1]. Most of the growth could come from biomass. Besides GHG emissions reduction, biomass brings some co-benefits such as employment, local development, energy independence and forest upkeep. For instance, in France, there is an unexploited forest resource for biomass energy uses of around 50 million $m^3$/year (12 million toe/year). Since 2000, the French Agency for Environment and Energy Management has coordinated a programme dedicated to the development of biomass-fuelled boilers. Between 2000 and 2006, 1400 utility boilers and 480 industrial boilers have been implemented thanks to this programme.
However, it is well known that the main trade-offs are air emissions and solid residue releases. POPs are particular pollutants of concern in the case of biomass combustion. Firing installations for wood (< 50 MW) are considered a major stationary source of PCDD/F emissions by the Aarhus Protocol. Annex V of the Protocol proposes Best Available Techniques (BAT)[2]. Since December 2007, it has been decided to revise the current Protocol. In addition, firing installations for wood and other biomass fuels is a source category subject to the requirement of the article 5 of the Stockholm Convention. In 2007, the Stockholm Convention Conference of the Parties has adopted the revised draft guidelines on BAT and Best Environmental Practices (BEP)[3] to help Parties take the measures listed in the aforementioned article 5. France is a Party to both legally binding instruments.
Unfortunately, emission factor data on POP releases from utility boilers are still very limited. For instance, in the standardized toolkit for identification and quantification of PCDD/F releases developed by UNEP Chemicals, emission factors for clean wood fired power boilers (subcategory 3.b, class of process n°2) are derived from 3 references only. Emission factors (EF) for air emissions and solid residues are 50 µg TEQ/TJ and 15 µg TEQ/TJ respectively[4]. Broadly speaking, the scientific literature is mainly focused on combustion of treated wood for example with CCA, on residential combustion, on industrial boilers or on assessments at laboratory and pilot scales. Lavric et al[5] carried out an extensive review of data which was available in the literature. Among the 80 references quoted in this paper, only a few dealt with data derived from utility boilers burning clean wood.
A comprehensive assessment of energy and environmental performances of biomass boilers in real operation was initiated by ADEME in 2007 within the framework of a new French nation-wide programme entitled « Biomasse Energie 2007-2010 ». This paper presents first results regarding POPs releases from the utility boiler category. The main objective of this work is to derive EFs which are representative of technologies and biomass fuels used on the French territory.

## Material and methods

During the 2007-08 wintertime period, a measurement campaign was achieved on 8 French utility boilers fed with different kinds of biomass fuels: wood, straw, bark, sawdust, crushed pallets, non hazardous industrial wastes, related products from sawmills, particle board wastes, including some fuel mixtures. Table 1 shows the characteristics of each installation and each fuel used to feed these installations. In all 8 facilities, grate-type furnaces are used. Thermal inputs are ranging from 320 kW to 6 MW. Depending on the fuel, the moisture content varies considerably (from 8.6% to 49.5%). All installations are equipped with multicyclones (MC) as air pollution control (APC) devices. Installations n°1 and n°2 only have implemented additional dust control devices such as fabric filters (FF) or electrostatic precipitators (ESP). No dedicated dioxin removal technique such as activated carbon injection was implemented. Boilers n°1 to n°7 were well operated and well managed. On the other hand, a simple visual check indicated that boiler n°8 seemed to be badly operated: the whole installation faced significant problems of airtightness and the air supply system seemed to be defective.

**Table 1. Characteristics of the investigated boilers and the fuel used.**

| Boiler n° | Commis-sioned in | Operating conditions | Thermal input (kW) | Backup fuel | Biomass fuel | Biomass GCV (MJ/kg) | Biomass moisture content (%) | APC |
|---|---|---|---|---|---|---|---|---|
| **1** | 2005 | continuous | 6000 | oil | wood | 14.7 | 23.4 | FF |
| | | | | | straw | 18.7 | 8.60 | |
| **2** | 2005 | continuous | 4000 | gas | bark + sawdust | 11.6 | 45.5 | ESP |
| **3** | 1993 | continuous | 3500 | oil | related products from sawmills | 10.6 | 49.5 | MC |
| **4** | 2006 | continuous | 2500 | gas | related products from sawmills + non hazardous industrial wastes + crushed | 15.2 | 34.1 | MC |
| **5** | 2006 | continuous | 320 | gas | crushed palets | 16.0 | 14.3 | MC |
| **6** | 2002 | continuous | 2 x 750 | gas | palets | 13.2 | 45.6 | MC |
| **7** | 2004 | batch (20h/d) | 2 x 450 | gas | non hazardous industrial wastes + crushed wood | 16.1 | 24.0 | MC |
| **8** | 1992 | continuous | 400 | oil | particle board | 18.2 | 23.8 | MC |

PCDD/Fs, indicator PCBs (i-PCBs), dioxin-like PCBs (DL-PCBs), HCB and PAHs were measured at the stack through a 3-hour sampling. Standardized reference methods were used for dioxins (NF EN 1948) and for PAHs (NF X 43 329). The 16 EPA PAHs were taken into consideration. NF EN 1948 was used to measure DL-PCBs. NF X 43 329 was used to measure HCB and i-PCBs. Analysis quantification limits in air samples were 5 pg I-TEQ/sample, 20 ng/sample, 0.7 pg/sample, 50 ng/sample and 20 ng/sample for PCDD/Fs, i-PCBs, DL-PCBs, HCB and PAHs respectively. Other combustion pollutants were measured at the stack : dust, $CO_2$, NOx, non methane volatile organic compounds (VOCs), CO, Hg and other heavy metals. These substances were measured using standardized reference methods, respectively: NF EN 13284, NFX 20301, NF EN 14792, NF EN 13526, NF 43012, NF EN 13211 and NF EN 14385. The dust size distribution lower than 1 µm (PM1) was also assessed by using an impactor as described in the prISO 20301 standard. PCDD/Fs were also measured in bottom ashes. It is worth highlighting that, in all installations, fly ashes and bottom ashes are collected separately, except in the case of installations n°3 and 6 where both ashes are mixed within the process. The analysis quantification limit in ash samples was 5 pg I-TEQ/sample for PCDD/Fs. These POPs were not measured in the fuels. Nevertheless, 77 organic compounds and halogenated organic coumpounds were measured in the fuels feeding the 8 installations. Ash and fuel sample analyses were carried out using GC/MS. The analysis quantification limit in fuel samples was 10 µg/kg.

**Results and discussion**

Table 2 provides organic compounds concentrations in fuels, PCDD/Fs concentrations in ashes and PCDD/Fs, PCBs, PAHs and HCB concentrations in air emissions. In all installations, less than 10 organic compounds are present in the fuels at concentrations higher than the detection limit values. The most detected organic compound was toluene, followed by dichloromethane. Toluene concentrations were ranging from 10 to 325 ng/g, the highest value being related to straw. In almost all installations, POPs air emissions could be considered very low, especially in the case of HCB since HCB concentrations were generally lower than the quantification limit values. No i-PCBs emissions were detected. Only installation n°8 showed significant higher PCDD/Fs, PCBs and PAHs concentrations. If installations n°1 to n°7 are considered, PCDD/Fs concentrations are lower or slightly higher than 0.1 ng I-TEQ/$m^3$. Apart from installations n°5 and n°8, PAHs concentrations were lower than 200 µg/$m^3$. In the case of installation n°5, PAH high concentrations were solely due to naphthalene. BaP emissions however were very homogeneous amid the 7 first boilers. For these 7 first boilers, PCDD/F releases through solid residues were ranging from 1.79 to 117 pg I-TEQ/g. Installation n°8 again showed significantly higher PCDD/F concentration in residues. Table 3 gives concentrations of other air pollutants. Despite the absence of fabric filters and ESPs for 6 boilers, dust emissions can be viewed as quite low, as concentrations are ranging from 16 to 169 mg/$m^3$. For boilers n°1 to n°7, good combustion conditions can be regarded as fulfilled, since dust, NOx and CO concentrations are consistent with the emission levels associated with European BAT for large combustion plants[6]. As opposed to these results, VOCs and CO emissions from boiler n°8 are very high, which confirms the suspected bad operating management of this boiler. Thus, bad operating conditions in this boiler can be seen as responsible for the significantly higher POPs emissions at the stack and through solid residues.

**Table 2. Organic compounds in fuels and POPs releases through air emissions and solid residues.**

| Boiler n° | Fuel | | Solid residues | Air emissions | | | |
|---|---|---|---|---|---|---|---|
| | Organic compounds | | PCDD/Fs | PCDD/Fs | DL-PCBs | PAHs | HCB |
| | ng/g | | pg/g I-TEQ | ng/$m_0^3$ I-TEQ | ng/$m_0^3$ | µg/$m_0^3$ | ng/$m_0^3$ |
| **1 - wood** | toluene | 167 | - | 0.017 | 0.0005 | 124 | <38 |
| **1 - straw** | toluene | 325 | - | 0.015 | 0.0003 | 167 | <42 |
| **2** | dichloromethane | 21 | 3.48 | 0.0003 | 0.0003 | 4 | <21 |
| | toluene | 10 | | | | | |
| **3** | p-isopropiltoluene | 56 | 9.49 | 0.031 | 0.0010 | 23 | <29 |
| | toluene | 37 | | | | | |
| **4** | n.d. | | 62.0 | 0.132 | 0.0031 | 41 | <28 |
| **5** | dichloromethane | 46 | 10.6 | 0.044 | 0.0015 | 11762 | <62 |
| | toluene | 128 | | | | | |
| **6** | dichloromethane | 60 | 117 | 0.060 | 0.0004 | 14 | <41 |
| **7** | n.d. | | 1.79 | 0.055 | 0.0035 | 145 | <101 |
| **8** | Toluene | 16 | 6924 | 2.88 | 0.0931 | 90969 | 889 |

n.d. : not detetected

EFs expressed per energy input (g/GJ) were derived from concentrations listed in table 2, fuel GCVs, fuel consumptions and operating times, on a site by site basis. EFs for PCDD/F, PCB and BaP for installation n°2 to n°7 are gathered in table 4. Due to lack of data at the time of writing, EFs for installation n°1 could not be assessed properly. Given its particular case, installation n°8 was considered separately. Table 4 shows that the mean values for PCDD/F, PCB and BaP are respectively 49 ng I-TEQ/GJ, 1.6 ng/GJ and 0.14 mg/GJ. These EFs are slightly different but still consistent with the ones used currently for the French official air emission inventory. The PCDD/F emission factor is very close to the one proposed in the Stockholm Convention toolkit. For the specific case of installation n°8 where POPs emissions are the highest because of bad operating conditions, EFs are 3000 ng I-TEQ/GJ, 97 ng/GJ and 228 mg/GJ for PCDD/Fs, PCBs and BaP respectively.

To conclude with, for utility boilers using clean wood as a fuel and that are properly managed and operated, the following conclusions can be drawn from the previous results derived from measurements on real installations:

- POPs emissions can be regarded as quite low. More particularly, air emissions of PCDD/Fs are lower (or slightly higher) than 0.1 ng I-TEQ/$m^3$.
- The key parameter influencing POPs releases is the way the boiler is operated. POPs releases are significantly minimized where good combustion operating conditions are implemented.
- It should be emphasized that these air emissions can be obtained with simple dedusters such as multicyclones and without any specific PCDD/F emission control such as activated carbon adsorption, provided good combustion operating conditions are implemented.
- Apart from combustion operating conditions, POPs releases could not be correlated with any other parameter such as thermal inputs or fuel characteristics (source of biomass, moisture content or organic content).

**Table 3. Air emissions for other pollutants.**

| Boiler n° | Dust | PM 1 | NOx | VOCs | CO | Heavy metals G1 | Heavy metals G2 | Pb | Heavy metals G4 |
|---|---|---|---|---|---|---|---|---|---|
| | $mg/m_0^3$ | % | $mg/m_0^3$ | $mg/m_0^3$ | $mg/m_0^3$ | $mg/m_0^3$ | $mg/m_0^3$ | $mg/m_0^3$ | $mg/m_0^3$ |
| **1 – wood** | 16 | 51 | 229 | 1.6 | 240 | 0.001 | 0.000 | 0.021 | 0.114 |
| **1 - straw** | 7 | 60 | 284 | 0.9 | 98 | 0.002 | 0.000 | 0.025 | 0.318 |
| **2** | 18 | 66 | 269 | 57 | 63 | 0.000 | 0.000 | 0.006 | 0.147 |
| **3** | 162 | 61 | 145 | 90 | 424 | 0.001 | 0.002 | 0.005 | 0.202 |
| **4** | 127 | 77 | 359 | 13 | 218 | 0.002 | 0.005 | 0.082 | 0.682 |
| **5** | 99 | 86 | 124 | 4.4 | 43 | 0.007 | 0.046 | 0.032 | 0.556 |
| **6** | 71 | 56 | 190 | 3.1 | 555 | 0.018 | 0.007 | 0.032 | 1.83 |
| **7** | 169 | 89 | 323 | 3.5 | 1520 | 0.001 | 0.001 | 0.009 | 0.333 |
| **8** | 55 | 74 | 184 | 2192 | 9269 | 0.003 | 0.004 | 0.043 | 0.421 |

G1 : Hg+Cd+Tl G2 : As+Se+Te G4 : V+Cr+Mn+Co+Ni+Cu+Zn+Sn+Sb

**Table 4. POP emission factors for air emissions**

| POP | Mean value | Minimum value | Maximum value |
|---|---|---|---|
| **PCDD/F** (ng I-TEQ/GJ) | 49 | 0.2 | 132 |
| **PCB** (ng/GJ) | 1.6 | 0.2 | 3.1 |
| **BaP** (mg/GJ) | 0.14 | 0.006 | 0.4 |

**References**


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