LANDFILL WASTEWATER TREATMENT PLANTS (WTP) PERMEATES VS EU DIRECTIVE 2000/60/CE: CASE STUDY OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs)

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EXECUTIVE SUMMARY

Background

Directive 2000/60/EC requires that permeates discharged in surface waters by landfill leachate treatment plants, respect limit values for priority substances emission. This study aims at assessing adequacy between Directive requirements and quality of WTP permeates discharged by 12 Municipal Solid Waste (MSW) landfills. It focuses on naphthalene, anthracene and 6-Borneff group, which are the most concerned PAHs as they belong to EU list of priority substances.

Methodology

The study includes every sanitary landfill currently exploited, or recently closed, in Wallonia and established all around its territory. In order to compare MSW landfills emissions with others sectors, 3 industrial WTP belonging respectively to steel, paper and PVC plants, have been included in the study. These three installations purify process waters, industrial solid waste (ISW) leachates or both together. Various processes are installed in these WTP, such as decantation, bioreactor treatment, physico-chemical coagulation, flocculation, reverse osmosis, ultra filtration, ozonation and activated carbon adsorption. PAHs concentrations have been measured every 6 months during four years at entry and exit of every WTP. Analyses results are related to semi-annual cumulated outflows of permeates. It gives rough assessment of PAHs emission flow-rates.

Results

PAHs concentrations in wallonian MSW leachates are much weaker than those indicated in Andreottola et al. (1992). MSW leachates are more concentrated than ISW, but PAHs distributions are similar: the same molecules (naphthalene, phenanthrene and fluoranthene) are predominant. Industrial process waters present a much less distributed composition: naphthalene reach 97 % of the total PAHs amount. Regarding EU and wallonian limit values, about 50 percents of the leachates are conform for Naphthalene (46% of sample < 1 µg/l) and anthracene (46 % of sample < 0,1 µg/l) without any treatment. The 0.1 µg/l limit value, fixed by Wallonian legislation for 6-Borneff PAHs sum, is the more restrictive: 80 percents of leachates exceed this limit. All studied WTP processes present very good reduction rates, especially when activated carbon adsorption is used as completion stage. Reverse osmosis system, with old technology, is the only with a poor purification rate. MSW permeates shows extremely low PAHs amounts. Only very few exceedances of the limit value (fixed for receptor river and not for emitted permeate before dilution) were observed. On average, PAHs are less frequently detected in MSW than in ISW permeates. As in leachates, naphthalene is the prevailing PAH in permeates, in which the heaviest molecules are almost absent. Cumulated PAHs emission flow-rate from MSW landfill reaches only 43 g/year. As a comparison, a single paper plant rejects about 20 kg of PAHs in a year; decantation basins in a PVC manufacture discharge 10 kg per year.

Conclusions

All together, wallonian landfills have a negligible contribution to the global PAHs emission in surface waters. EU Directive 2000/60/CE objectives of “progressive reduction” for pollutants and “cessation or phasing-out of discharges, emissions and losses” for priority hazardous substances can be considered as "already reached" by the waste landfilling sector in Wallonia. Other industrial sectors emit extremely larger amounts of PAHs. The study allows selecting, among the 15 studied PAHs, naphthalene as the most relevant tracer for landfill PAHs emissions.

1 INTRODUCTION

1.1 Legal background

The European Directive 2000/60/EC establishes a framework for Community action in the field of water policy. The action settles strategies to prevent pollutions of water. Landfilling waste management yields large quantities of leachate, which contains hazardous contaminants. Some of them appear in the list of the 33 priority substances established by the EU parliament decision N° 2455/2001/EC. According to the Directive, produced leachates have to be purified in WTP before being discharged to surface waters. The rejected permeates shall cope with the "progressive reduction" for pollutants and "cessation or phasing-out of discharges, emissions and losses" for priority hazardous substances.

Polycyclic Aromatic Hydrocarbons (PAHs) hold an important place in the list of priority substances: as a group, they are identified as "priority hazardous substances". As single compounds, naphthalene and anthracene belong to a group of 15 priority substances subjected to a review for identification as possible "priority hazardous substance". Every PAH of the 6-Borneff group is in the "priority substance" list without the “hazardous” label. Among these 6 compounds, fluoranthene is in the list "as an indicator of other, more dangerous Polyaromatic Hydrocarbons".
The Walloon Decree 12/09/2002 transposes the Directive in Wallonian law. It establishes a monitoring network for water quality and plans to reduce pollution caused by dangerous substances, among which PAHs. It gives the following limit values for PAHs in surface water: naphthalene - 1 µg/l, anthracene - 0,1 µg/l, sum of 6-Borneff group - 0,1 µg/l.

1.2 Technical background

PAHs are persistent organic pollutants compounds showing 2 to 7 benzenic rings. Their molecular weight vary between 128 and 300 g/mol, they stay solid under 100 °C. Some PAH have proven carcinogen effects (IARC, 1983). PAHs are generated during incomplete organic matter combustion, or synthesized during the formation of fossil matters like oil and coal. They are spread in environment by fume diffusion or by using derivative oil products. PAHs are rarely found as single compounds. Most of the time, industrial processes produce mixtures of various PAHs species, discharging them together in environment (Rosik-Dulewska et al., 2008). Most of the rejections are first atmospheric, before being settled on the grounds or in surface waters. They accumulate in various organisms with bioconcentration factors from 4 to 7800 observed in unicellular algae (LCPE 1994). Principal human exposure pathways are alimentation and inhalation of particles.

A lot of specific papers and reviewing articles on MSW landfill composition are available. However, most of these works give no or few information about PAHs case study. For naphthalene only, Kjeldsen et al. (2002) gives mean value (34 µg/l) and range (0.1-280 µg/l). These values come from a few of old studies. Haarstad & Maehlum (1999), focused on Norvegian landfills, cites a range of 0.2-21µg/l for the sum of PAHs without giving neither the source of these data or the list of analysed PAHs. Further in the past, Andreottola et al. (1992), gave some statistics on others PAHs (see column “ref.” in table 2) coming from Shridharan & Didier (1988) or even older publications. Up-to-date works on PAHs in leachates (Rosik-Dulewska et al., 2008) do not cite more recent references.

2 METHODOLOGY

2.1 WTP processes of selected landfills

Studied MSW sanitary landfills are equipped with various water treatment processes (see table 1): Coagulation-Neutralisation-Clarification-Flocculation systems; membrane bioreactor (MBR) processes; reverse osmosis; activated carbon adsorption (ACA); and sometimes some combination of these elements.

The work includes every MSW sanitary landfills currently exploited in Wallonia, or recently closed : Froidchapelle (FRO), Hallembaye-1 (HAL1), and Hallembaye-2 (HAL2), Belderbusch (BEL), Beaumont (BEA), Cour-au-Bois (CAB), Happe-Chapois (CHA), Hapay-la-Neuve (HAB), Tenneville (TEN), Morialmé (MOR), Malvoisin (MAL) and Mont-Saint-Guibert (MSG). The last three send their waste waters to urban treatment plants; they do not treat the leachates in their own WTP.

In order to compare the emissions of MSW sector with others industrial water treatment plants, 3 other sites have been included in the study:

- Seraing (SER) WTP treats leachates from the solid waste landfill of a steel-making plant (Arcelor)
- Virton (VIR) is the WTP of a big paper-making plant which purifies its process water mixed with leachate from the associated industrial solid waste landfill (Burgo).
- Jemeppe (JEM) wastewater outflow is rejected by large decantation basins which collect process water from a PVC-making plant (Solvay).

TABLE 1 **Characteristics of the studied landfills and leachate treatment plants in 2007**

	Area (ha)	**Waste volume (10^3 m^3)**	**Type of plant – Working stage – Type of water**	**WTP process**	**leachate flow m^3/month**
FRO	13	951	MSWL(1) - In exploitation - Leachates	MBR/ACA	700
HAL1	10	1.640	MSWL - Aftercare - Leachates	Reverse osmosis	930
HAL2	20	2.700	MSWL - In exploitation - Leachates	Reverse osmosis	2.200
BEL	14	500	MSWL - Aftercare - Leachates	MBR/ACA	1.480
BEA	18	660	MSWL - in exploitation - Leachates	MBR/ACA	1.700
CAB	36	5.200	MSWL - in exploitation (phase 1 closed) - Leachates	MBR/ACA	2.240
CHA	8	1.190	MSWL - in exploitation (phase 1 closed) - Leachates	MBR/O_3, then MBR/ACA	2.560
HAB	15	950	MSWL - in exploitation (phase 1 closed) - Leachates	CNFF/BR/ACA	5.770
TEN	14	1.160	MSWL - in exploitation (phase 1 closed) - Leachates	CNFF/BF/ACA	6.180
MOR	2	270	MSWL - Closed in 2006 - Leachates	none (Urban WTP)	775
MAL	1,2	63	MSWL - in exploitation (closure in 2008) - Leachates	none (Urban WTP)	580
MSG	26	4.000	MSWL - in exploitation (phase 1 closed) - Leachates	none (Urban WTP)	6.200
SER	37	1.500	Steel ISW(2) - in exploitation - leachates + PW(3)	GR(4) + decantation	13.520
VIR	6	175	Paper mill + ISW - in exploitation – leachates + PW	GR + aeration, PCP(5)	1.964.000
JEM	2,4	40	PVC mill – in exploitation – PW	Décantation basins	307.700

(1) municipal solid waste landfill (2) industrial solid waste (3) process water (4) Greases removal
(5) Physico-Chemical Purification

Table 1 presents the technical data of each landfill, including water purification processes. The abbreviations used in this table are developed as follow.

- "MBR" is a combined system including biological degradation and microfiltration stage.
- Reverse osmosis process consists in four times concentrating leachates (within two successive membranes systems) and evacuating the concentrated leachate out of the site for further treatment (thermal destruction).
- "O_3" means "ozonation".
- "CNFF" is the abbreviation for the following sequence: physico-chemical **C**oagulation (Al^{3+}) + soda **N**eutralisation + biological **F**locculation (cationic polymer) + **F**lottation ;
- "BR" indicates the aerobical denitrification in **B**iological **R**eactor
- "ACA" means "activated carbon adsorption", it is mainly used as completion stage of other processes.

2.2 Pluviometry and leachate flow-rate

Especially on MSW landfills, there is a strong relation between pluviometry and WTP outflow. Furthermore, the total volume of leachate to be purified is influenced by landfill area, capping nature, and eventual use of recirculation process. Figure 1 shows the link between pluviometry inflow and WTP outflow. A time gap appears in MSW between inflow and outflow peaks. This retardation coefficient is variable (some days to some weeks), from one site to the others, depending on local specificities.

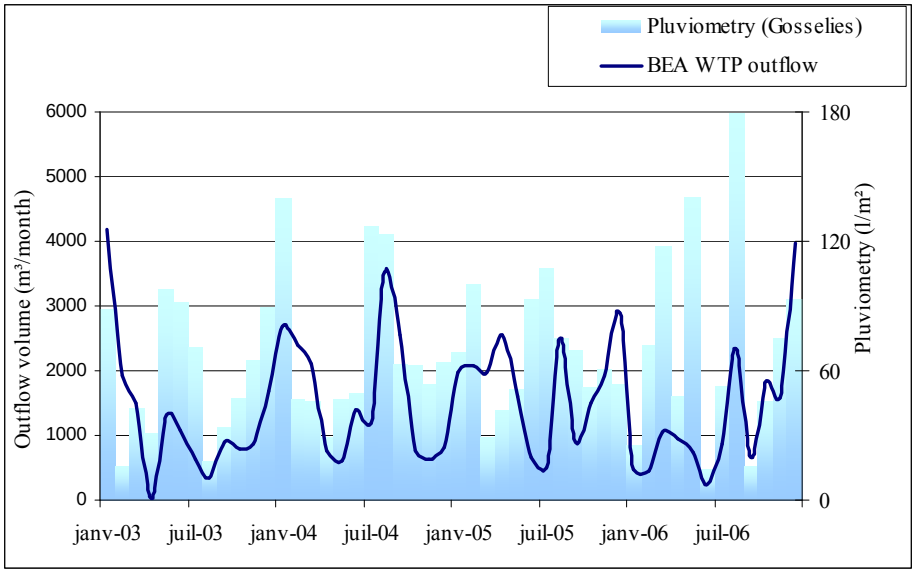

FIGURE 1 **Relationship between pluviometry and landfill WTP outflow case study of BEA landfill, 2006**

2.3 Sampling and chemical analysis methods

Leachate and permeate sampling events occurred systematically in March and September, from September 2003 to March 2007. Each survey take typically place during 4 consecutive open days for covering each of the 15 sites. Samplings were performed at entrance (leachate) and exit (permeate) of each WTP. In the three landfills without WTP, only leachates were collected. For permeates only, automatic samplers were used for constituting 24h average samples, from 24 sub-samples (200ml each) collected hourly. Automatic samplers are generally installed at the official WTP outfall of sewage. Leachates were sampled manually (instantaneous sample), directly in their retention basins. Samples were handled and stored in freeze boxes before chemical analysis (achieved maximum 3 days after sampling).

Analyses were carried out in accordance with standard NBN EN ISO 17993 which includes the following steps: pre-concentration, liquid-liquid extraction and fluorimetry detection. This standard method allows analysing every PAHs of the 16 EPA group except acenaphthylene, as it is not detected by fluorimetry method. By facility, the 15 others PAHs are named “15 EPA” hereafter.

2.4 Rough assessment of emission flow-rates

Leachate volumes from the three MSW without WTP (MAL, MOR, MSG), have been measured during pumping to urban WTP or to collecting trucks. Most of the MSW and industrial - WTP are equipped with automatic flowmeters at the waterfall of sewage. Failing this, regular manual measurements were performed by owners with portable flowmeters. Landfill owners installed these measurement tools themselves, in order to follow their taxation obligation. As each operator may select himself which system to use, differences in precision from one site to another can be high. From these instantaneous flow-rates, it is easy to obtain the total volume of wastewater that is discharged during semesters preceding each sampling event. These semi-annual volumes, multiplied by the PAHs concentrations measured once, in the middle of semesters, give rough assessments of semi-annual PAHs emission rates in surface waters. The biggest approximation in such an assessment is linked to the variability of wastewater composition with time. As it is well known, leachate composition shows high temporal variations. But WTPs induce a smoothing effect on these variations. The composition of WTP purified water is then much more stable than the one of incoming leachates.

3 RESULTS AND DISCUSSIONS

The 4 years monitoring, including 8 complete surveys, allow collecting 111 leachate samples, among which 95 from MSW and 16 from industrial solid waste (ISW) landfills. In addition, 20 industrial untreated process waters were sampled. Finally, 85 samples of permeates (purified waters) complete the samples set.

Table 2 presents an overview of analytical results obtained from these samples. In VIR and JEM WTP, ISW leachate and process wastewater are treated together, after mixing. For this reason, the analytical results obtained on industrial permeates are not dissociated. In the same table, "D" is the detection limit of the analytical methods, "MED" are the median values computed for the different types of samples.

When more than 50 percents of the results are below the detection limit, the computation of the median value is not relevant anymore. In permeates of MSW landfills, this is the case for every PAHs compounds. The only relevant parameter value is the proportion of samples with concentration higher than detection limit. For permeates of industrial treatment plants, only some of the median values are computable, the other PAHs are also detected in less than 50 percents of samples. Due to lack of more complete, more representative and more recent reviewing work, the median concentrations published in Andreottola et al. (1992) are given as reference values.

TABLE 2 **Analytical results (in µg/l unless specified) obtained on the different types of samples**

		Leachates					**IPW** (industrial process waters)		**Permeates**		
		MSW		**ISW**		**R**[(1)]			**MSW**	**ISW+IPW**	
Number of samples		95		16			20		61	24	
		% > D[(2)]	MED [(3)]	% > D	MED	MED	% > D	MED	% > D	% > D	MED
Naphthalene	0,05	97%	1,05	69%	0,08	33,7	95%	1,30	49%	83%	0,26
Acenaphthene	0,03	96%	0,28	50%	0,01	17,6	60%	0,02	33%	67%	0,03
Fluorene	0,01	99%	0,22	63%	0,01	26,8	60%	0,02	41%	75%	0,01
Phenanthrene	0,015	98%	0,43	69%	0,03	50,7	75%	0,03	30%	67%	0,02
Anthracene	0,002	100%	0,11	81%	0,01	-	65%	0,002	36%	79%	0,001
Fluoranthene[(6b)]	0,005	100%	0,45	94%	0,06	39,1	70%	0,01	33%	92%	0,02
Pyrene	0,005	100%	0,32	94%	0,06	-	70%	0,01	41%	83%	0,01
Benzoanthracene	0,005	100%	0,10	81%	0,02	-	45%	-	23%	54%	0,002
Chrysene	0,005	100%	0,11	88%	0,02	-	65%	0,01	31%	67%	0,002
Benzo(b)fluoranthene[(6b)]	0,005	99%	0,06	81%	0,02	-	45%	-	15%	46%	-
Benzo(k)fluoranthene[(6b)]	0,005	98%	0,02	69%	0,01	-	25%	-	8%	17%	-
Benzo(a)pyrene	0,003	98%	0,04	81%	0,01	-	40%	-	20%	46%	-
Dibenzoanthracene	0,005	46%	-	25%	-	-	5%	-	0%	4%	-
Benzo(g,h,i)perylene[(6b)]	0,005	92%	0,03	63%	0,01	-	20%	-	5%	13%	-
Indeno(1,2,3-c,d)pyrene[(6b)]	0,005	81%	0,02	63%	0,01	-	25%	-	2%	8%	-
6-Borneff (Sum)	-	100%	0,62	94%	0,14	-	75%	0,05	48%	92%	0,02
15-EPA[(4)] (Sum)	-	100%	4,15	100%	0,43	-	95%	1,86	67%	100%	0,51

(1) reference value (Andreottola et al., 1992) (2) Highest detection limit = 0.05 µg/l (3) Median value
(4) the 15 analysed compounds of the 16-EPA list, less the acenaphtylene (6b) belong to the 6-Borneff group

In the following sections, results are discussed in three stages. The first one focuses on concentrations measured in the leachates, proportions of each compound and total amount of pollutants to be reduced by WTPs. The second stage presents PAHs reduction rates of every WTP and compares their relative efficiencies. The third stage assesses quantities of PAHs rejected in permeates, which finally give total emission rates to the surface waters. In order to make this discussion easier, results are systematically presented by isolating the two "EU-priority" parameters (naphthalene and anthracene) and the 6-Borneff group, which have a maximal value in Walloon legislation. Other substances are discussed together ("7 other PAH"). Finally, 15 PAHs total amount is presented in order to give an idea of global emission rates.

3.1 Composition of leachates

As shown in table 2, in every 95 MSW leachates, at least five of the 15 PAH are detected simultaneously. Among the 15 molecules, dibenzoanthracene is by far the least frequently detected one.

PAHs concentrations measured in wallonian landfill leachates are surprisingly low. They are significantly lower than those presented in Andreottola (1992). No concrete argument may explain a variation of such an order of magnitude. Of course, the nature of wastes, their age and a multitude of other parameters contribute to create wide range of concentrations; but observed landfills of both studies contain, in first approximation, similar wastes. Except for naphthalene, results of the present study never reach the lower limit of Andreottola's ranges. And even for naphthalene, only the highest measured concentrations fall in the lower part of the range. On the contrary, the values published by Kjeldsen et al. (2002) and Haarstad & Maehlum (1999), are closer to the observed concentrations.

Histograms of EU-priority compounds presented in figure 2 shows that about 50 percents of leachates are below the limit for naphthalene (49 % of sample < 1 µg/l) and anthracene (46 % of sample < 0.1 µg/l) without any treatment. The same graph indicates that 0.1 µg/l value, fixed by Walloon legislation for concentrations of 6-Borneff PAHs sum, is the most restrictive. With it only, this standard value forces to purify 92 percents of MSW leachates. If the same 0.1 µg/l maximal value was imposed to fluoranthene alone, 80 percents of leachates would already exceed it.

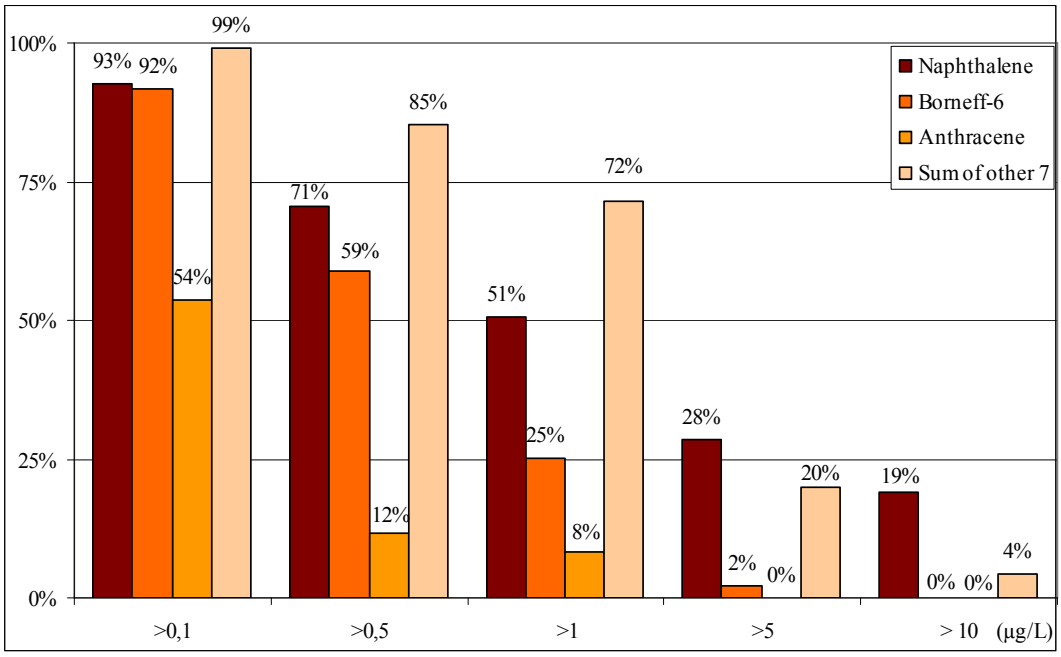

FIGURE 2 **Histograms of EU-priority PAHs in MSW landfill leachates**

Radial graphs in figure 3 present distributions of PAHs median concentrations, in MSW leachates, ISW Leachate and industrial process water respectively. The proportion of each species refers to the sum of median concentrations. Naphthalene prevails in MSW landfill leachates, followed by phenanthrene and fluoranthène, which represent 70 % of the 6-Borneff sum of median concentrations. ISW and MSW leachates partitioning are similar, but median total PAHs concentrations are different: MSW leachates are more concentrated than ISW leachates. Highest ratios are observed for light molecules like naphthalene, phenanthrene and fluoranthene. On the contrary, median composition of industrial process waters is not comparable. Naphthalene predominance becomes extreme (93% of median composition). Other compounds are not detectable or found in negligible proportions.

Another interesting observation concerning leachate composition is the absence of correlation between total amounts of PAHs measured leachates and their volume produced during the month before sampling. It would have been expected that a higher monthly volume would correspond to a lower concentration, but such relation is not observable in the data collected during this study.

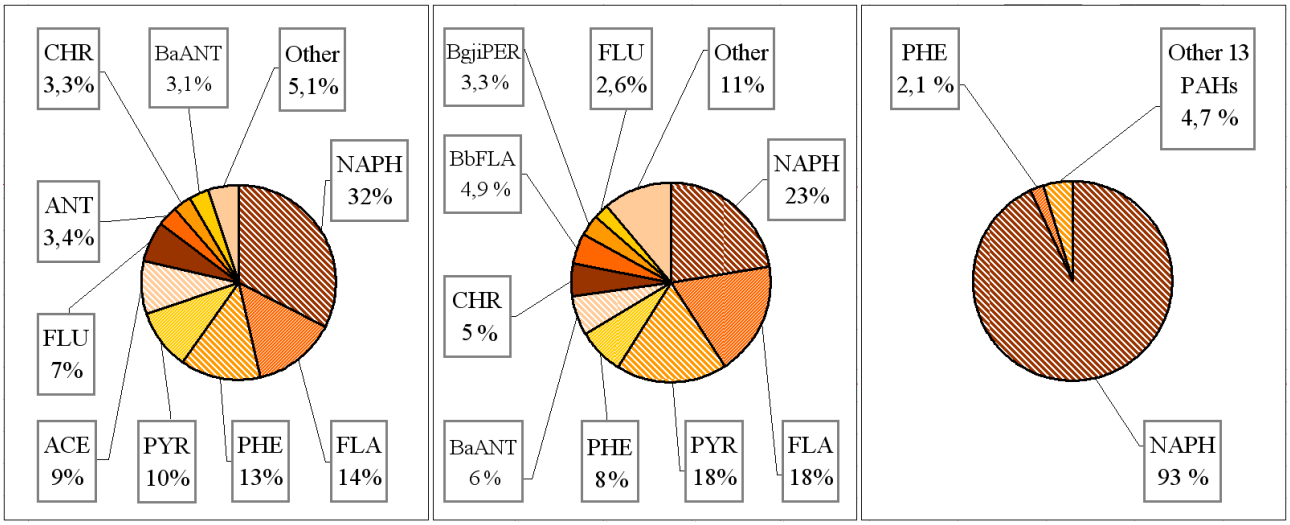

FIGURE 3 **Median PAHs distribution in MSW landfill leachates**

Abbreviations : Naphthalene - NAPH, Acenaphtene - ACE, Fluorene - FLU, Phenanthrene - PHE, Anthracene - ANT, Benzo(a)anthracene - BaANT, Fluoranthene - FLA, Benzo(b)fluoranthene - BbFLA, Benzo(g,h,i)pérylène - BghiPER, Pyrene - PYR, Chrysene - CHR,

3.2 Purification rates of leachate treatment plants

Table 3 presents mean reduction rates of every studied WTP systems. It focuses on the EU-priority substances, giving additional values for "other PAHs" and "total PAHs". These rates are assumed to be the ratios between concentrations measured in leachates collecting basins and the ones measured, almost at the same time, at WTP exit. This is an approximation as it does not take into account retention times in leachates storage basins and purification durations. However homogenisation occurring in retention basins and 24-H sampling method for permeates contribute to minimize these errors.

TABLE 3 **Mean PAHs purification rates in leachate treatment plants**

MSW	WTP process	Naphthalene	Anthracene	Borneff-6	7 other PAH	15 EPA PAH
CAB	MBR/ACA	100%	100%	100%	100%	100%
FRO		100%	100%	100%	100%	100%
BEL		100%	100%	100%	100%	100%
CHA(1)		98%	97%	99%	98%	98%
BEA		98%	98%	96%	94%	97%
HAB	CNFF/BR/ACA	89%	100%	100%	99%	98%
TEN		100%	100%	100%	100%	100%
CHA(2)	MBR/O_3	96%	99%	96%	98%	97%
HAL1	Reverse Osmosis	46%	90%	97%	84%	62%
HAL2		90%	97%	96%	92%	92%
	Mean	**91%**	**98%**	**98%**	**96%**	**94%**

(1)3 of the 8 surveys (2) 5 of the 8 surveys

Purification rates of all systems are very good independently of HAPs species, except for HAL1 whose naphthalene purification rate is poor. The best PAHs reduction rates (> 98 %) are reached by systems including an ACA completion stage. CHA case study gives an illustration of ACA performances. Changing from O_3 system to ACA improved the total reduction rate by 1 percent. Reverse osmosis appears to be the least efficient for HAP purification. However, it gives the best results for mineral contaminations, which compensate its higher permeability to some organic compounds. Reduction rate of reverse osmosis decreases with the number of benzene rings, giving the worst ratio for naphthalene. The gap between HAL1 and HAL 2 is only due to the age of the plants. HAL2 has been installed recently. It profits from more recent techniques regarding osmotic membranes.

3.3 Composition of permeates

The graph in figure 4 shows that naphthalene is the only PAH reaching, sometimes, concentrations higher than 1 µg/l, which is its limits value fixed by EU-Directive. All these exceedances were observed at the same landfill (HAL1). Exceedances are not huge: concentration higher than 5 µg/l is never observed. The same graph indicates that 5 % of collected samples, from three different landfills (HAL2, BEL and CAB), show 6-Borneff concentrations higher than wallonian maximal value.

In order to avoid any confusion, an important commentary has to be given concerning the limit values fixed by the directive and wallonian law. These concentrations are quality targets for surface waters. In other words, it means that they have to be satisfied by the rivers in which permeates are rejected. Concentrations measured directly on discharged waters are diluted by mixing with surface water. Comparing permeates concentrations with in situ target values is a too strict strategy. Other measurements, achieved by ISSeP in the rivers, allowed verifying that any PAH concentration has never reached the limit value in rivers even just downstream WTP's points of disposal.

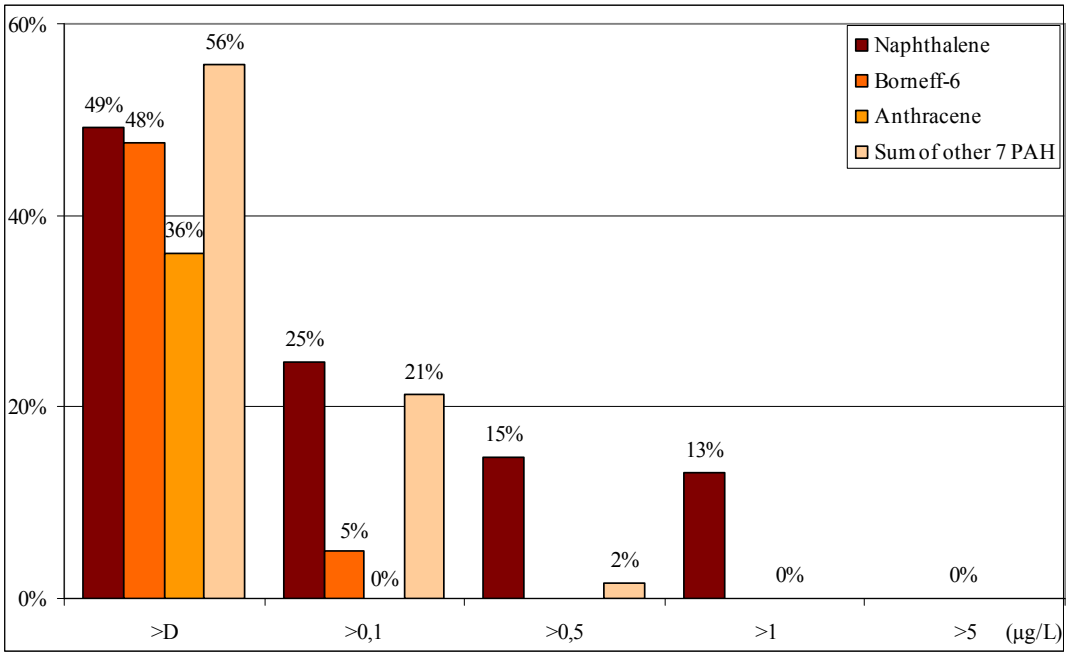

FIGURE 4 **Histograms of EU-priority PAHs in MSW landfill leachates**

In figure 5, relative frequencies of PAHs concentrations higher than 0,05 µg/l has been carried in graphs. It is a way to compare the proportion of each molecule rejected in the environment. The graphs compare the two types of liquid emissions: MSW and ISW+IPW permeates. The y axis of the graphs show, in percent, the proportion of values higher than 0.05 µg/l compared to the total number of measured value (number of samples*15 measured concentrations). Ratios indicated to the right of each histogram give, for each molecule, the number of event higher than 0.05, compared to the number of analysed samples. Finally, the distribution of events for each individual molecule is graphically represented by the proportion of each corresponding step in the total height of histograms. The following observations can be drawn from these histograms.

- The proportion of permeates containing PAHs traces is low.
- Industrial permeates are more frequently contaminated by PAHs traces than MSW permeates.
- As in leachates, naphthalene is the prevailing species in permeates.
- Heavy molecule are almost absent in permeates.
- Pyrene and fluoranthene are more frequent in industrial permeates than in MSW permeates, for fluorene, it is the contrary.

Of course there is a lot of samples where more than one PAH is present. These multiple detection events are not distinguishable in figure 5. A good way to apprehend this other point of view is to compare numbers of "totally clean" samples (the ones without any concentration higher than detection thresholds). This value is also better for MSW permeates with 47.5 percents of "clean" permeates compared to none percent in industrial permeates.

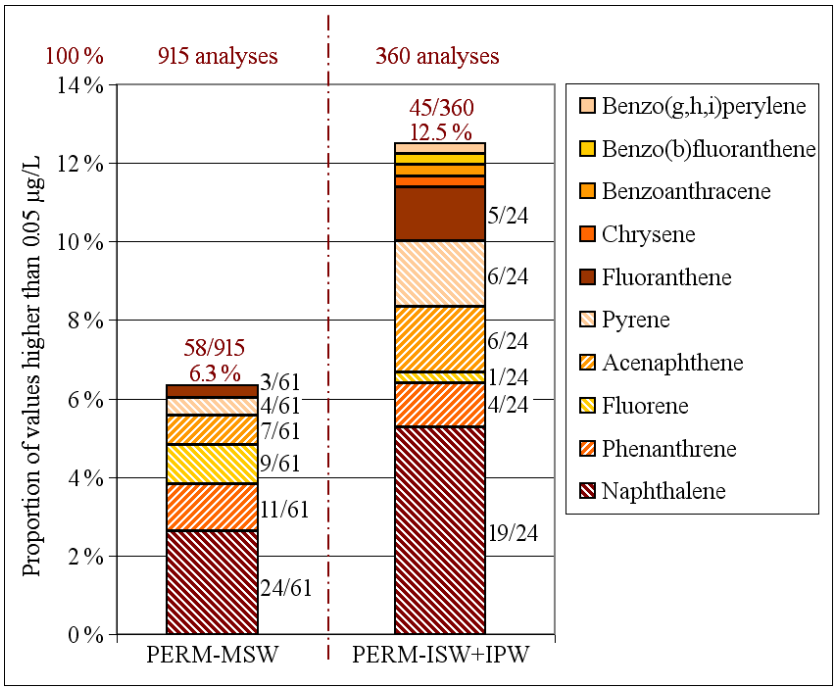

FIGURE 5 **PAHs distributions in MSW landfills and industrial permeates**

3.4 PAHs emission flow-rates

Table 4 presents total PAHs emission flow-rates of each individual industrial site and, as a comparison, cumulated flow-rates of all MSW landfill together. These values have been assessed roughly, as explain in section 2.6.

TABLE 4 **Flow-rates**

	Incoming flow-rate (leachates)		**Emission flow-rate (permeates)**	
	total amount (g/y)	proportion	total amount	proportion
VIR	36901	51,3 %	18755	68,4 %
JEM	32478	45,2 %	8532	31,1 %
SER	217	0,3 %	99	0,36 %
9 MSW landfills own - WTP	1788	2,5 %	43	0,16 %
3 MSW landfills - urban WTP	506	0,7 %	-	-

It shows that the global amount of PAHs emitted in surface water by all MSW landfills together (40 g) is negligible in comparison with the flow discharged by only one paper industry (20 kg) or a classical PVC plant (10 kg). The reason of this is double.

- First, the quantity of PAHs-rich wastewater produced by the industry is huge in comparison with landfill leachates production. There is no possibility for plants to reduce these incoming flow rates, as the processes themselves require water. In the contrary, landfill progressive covering tends to reduce leachates volumes in time.
- Secondly, industrial WTP do no include any treatment unit specifically dedicated to persistent organic compounds such as PAHs. In comparison with industrial WTP, the relative proportion of PAHs emitted by MSW WTP is even ten times lower than at the entrance of the purification plants.

It has to be noted that the importance of PAHs emissions coming from industrial WTP is not due to high concentrations but to high volumes, which is much more difficult to solve by purifying systems.
The last important observation, made on MSW WTP only, during the 4 years survey is a progressive improvement of WTP efficiency, which results in a reduction in time of PAHs emission flow-rates.

4 CONCLUSIONS

As a conclusion, this 4 years survey of PAHs in MSW landfills leachates and permeates all around the wallonian territory allows concluding that all landfills together have a negligible contribution to the global PAHs emission in surface waters. EU Directive 2000/60/CE objectives of "progressive reduction" for pollutants and "cessation or phasing-out of discharges, emissions and losses" for priority hazardous substances can be considered as "already reached" by the waste landfilling sector in Wallonia. Any further improvement of purification rates, without substantial effort in other industries would be, of course positive, but nevertheless ineffective in term of global quality of surface waters. Another big interest of this study is to show that naphthalene could be a sufficient tracer for every other PAHs contamination coming from landfill WTP. If a second molecule had to be monitored, fluoranthene (in place of borneff-6 sum) could be the best one. Finally, anthracene is totally useless in wallonian context: it is never detected in significant amount.

5 ACKNOWLEDGEMENTS

The authors would like to thank, for financial support, the Division de la Police de l'Environnement (DPE) - Ministère de la Région Wallonne. In particular, Mr. S. Godefroid, director of the DPE, must be thanked for giving to ISSeP the time and the opportunity for developing scientific work in parallel to routine surveying tasks. The authors wish to thank the whole teams of ISSeP's Labs for their valuable help in the sampling and analyses task.

6 REFERENCES

Andreottola, G., & Cannas, P., (1992): Chemical and biological characteristics of landfill leachate. In Landfilling of waste : leachate. pp 65-88. Edited by Christensen T.H., Cossu R., Stegmann R. (1992) Chapman & Hall, London, 520 p.

Awono, S., Kheffi, A., Hennebert, P., & GODFROID, S., (2005): Landfills waste water treatment plants permeates and European Union Directive 2000/60/EC : a case study of PAHs. Proceedings Sardinia 2005, Tenth International Waste Management and Landfill Symposium, S. Marg. di Pula, Cagliari, Italy.

European Commission (2000) : Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy. Official Journal of European Commission (O.J.E.C.). L 327 of 12.22.2000.

European Commission (2001) : Decision N° 2455/2001/EC of the European Parliament and of the Council of 20 November 2001 establishing the list of priority substances in the field of water policy and amending Directive 2000/60/EC. Official Journal of European Commission (O.J.E.C.) L 331 of 12.15.

Haarstad, K. & Maehlum, T. (1999) : Important aspects of long-term production and treatment of municipal solid waste leachate. Waste management and research, 17, pp 470-477 © ISWA, 1999.

IARC - International Agency for Reseach on Cancer (1983) : Monographs on the evaluation of the carcinogenic risk of chemicals to humans. Polynuclear Aromatic Compounds. Lyon. Part 1, volume 32.

LCPE - Loi Canadienne sur la Protection de l'Environnement (1994) : Liste des substances d'intérêt prioritaire-Rapport d'évaluation-Hydrocarbures Aromatiques Polycycliques, 69p.

Kjeldsen, P., Barlaz, M.A., Rooker, A.P., Baun, A., Ledin, A. & Christensen. TH. (2002) : Present and long-term composition of MSW landfill leachate: a review. Critical Reviews in Environmental Science and Technology, 32(4):297-336.

Rosik-Dulewska, C., Ciesielczuk, T. & Ramus, K. (2008) : Change in the polycyclic Aromatic Hydrocarbons (PAHs) content in an urban waste composting process. From : Management of Pollutant Emission from landfills and sludge. Pawlwska & Pawlowski (eds), pp 85-89, © Taylor & Francis Group, 2008.

Shridharan, L. & Didier, P. (1988) : Leachate quality from containment landfills in Winsconsin. ISWA 88 Proceedings, ed. L. Andersen & J. Moller, Vol. 2. Academic Press, London.