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Odour and VOC emissions reduction on coil-coating lines by using waterborne paints - part. II: full waterborne system application

S. Vigneron[a], P. Deprelle[b], J. Hermia[c]

[a] Research Engineer, Head of the Laboratory for Waste Effluent Treatment and Olfactometry (LTEG), Société Belge de Filtration/UCL, Louvain-La-Neuve, Belgium.

[b] Engineer in charge of the analytical support, LTEG, UCL

[c] Professor at the Université Catholique de Louvain, SBF Chairman

Adress: Laboratoire de Traitement des Effluents Gazeux et d'Olfactométrie, UCL, Unité des Procédés, Voie Minckelers, 1 - B-1348 LOUVAIN-LA-NEUVE (Belgium), tel. +32 10 47 23 19 - Fax +32 10 47 24 69 - email lteg@ucl.ac.be

1. Foreword

It is more and more admitted that the use of abatement techniques in order to reduce Volatile Organic Compounds emissions will not allow to achieve in the future the planned rejection objectives. This fact is especially admitted; indeed the abatement techniques located at "end of line" will always have a limited efficiency while their investment costs, possibly operating costs, can penalize products competitiveness.

Therefore, chemical industry agreed to make particular efforts for the elaboration of substitution products in order to eliminate as much as possible the organic phase. By suppressing the emission source, nuisances, which can affect workers as well as the environment, are eliminated.

In particular, in the paints field, new types of systems have appeared : paints formulations, coating application mode, etc...

Unfortunately, the originality of the proposed systems usually requires a deep process readjustement since the required application or curing mode is totally different. Finally, these systems are not yet adapted to all materials, for instance the application processes based on U-V or electron beam curing for steel [1]. For the chosen example, i.e. coil-coating where paint is applied on a steel sheet running continuously through roll coaters, homogeneity and layer thicknesses are typical characteristics of the delivered product. In this industry, only a few paints, which can be considered as high solids, are used (e.g. PVC), as well as waterborne paints chosen as primer or backer.

The present paper presents the advancement of an E.C. research [2] studying the feasibility of waterborne systems for coil-coating applications. Nowadays in Europe almost coil-coaters are using exclusively Organic Based Solvent (OBS-) paints. The SOLLAC steelmaker group, especially on its industrial plant localized at Onnaing (France), is an exception: great interest is accorded to Waterborne (W-) systems because their use allows to increase the rate of production without explosive problems. So, W-paints are used as backer.

After some generalities giving informations about the coil-coating process and the analytical methods which were used, the paper will be divided into three parts:

- comparison of two epoxies (OBS- and W-) used in a semi-classic paint system (OBS-primer and OBS-top coat + W-backer);
- presentation of the test results from a full waterborne system at industrial scale;
- comparison of the W-system with a full OBS-system.

All results are presented on the basis of equivalent ESP's*.

2. Generalities

2.1. *Description of the coil-coating line*

The concept of a coil-coating line is shown in figure 1.
Steel and galvanized steel coils 600 to 1500 mm wide and 1,25 to 1,5 mm thick are alternatively unrolled on two drive machines; a stitching machine realizes the junction between rolls and ensures a continuous steel sheet at the inlet of the installation. The accumulator allows to feed the "process" section during these stitching operations.
A surface treatment is carried out in order to provide, in addition to the anti-corrosion properties, an excellent adhesion of paints on the metal.
The sheet arrives then in the paint cabin (more or less 300 m^3 volume). A first machine, equipped with rolls, allows to apply:

- on the upper side: a 5 µm thickness dry primer;
- on the lower side: a 7 µm thickness dry backer.

After passing in the primer oven (20 s to 1 min according to the speed), a second layer is deposited by a second painting machine (10-19 µm top coat giving the color and the organic coating characteristics) before entering in a second oven (finish oven) and undergoing a cooling under water before rewinding of the coil after the checking post.

Each oven is followed by a dedicated thermal incinerator. The incinerators are supplied by extractors which keep the ovens depressurized.
Thanks to a gas input, the combustion of organic solvents contained in the paints takes place at about 700°C and allows for cracking these solvents.

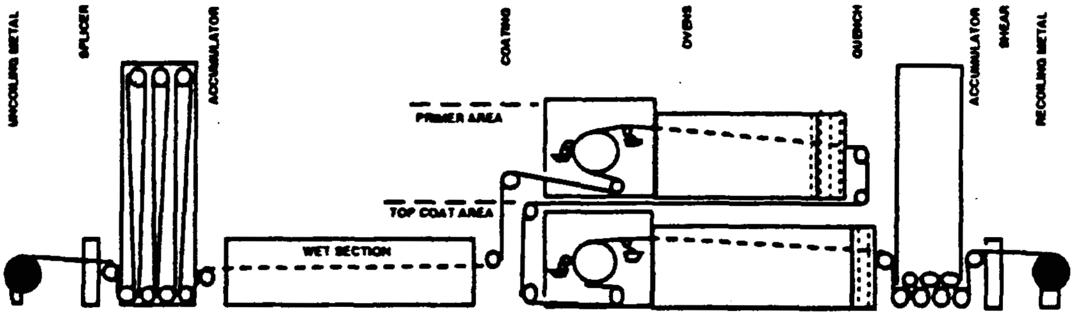

Fig. 1 - Diagram of a coil-coating line

2.2. *Analytical methods*

The given results in this paper concern odour levels measurements coupled with high sensitive chemical analysis; the odorous power (pOU) is a tool used as intermediate result to link chemical compounds concentration with odours concentration.

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* ESP: dry content of the paint, % volume

Odours are measured by means of an dynamic olfactometer (ODILE from Société Belge de Filtration) in reference to the latest works of the European Committee of Standardisation (CEN) [3]. Z is the dilution factor expressed as $\frac{Q+Qod}{Q}$ and r/p is the ratio of perception positive responses of the panel (r) on the full number of presentations of a dilution (p) (number of panellist x number of individual presentation) (see figure 2). The odour concentration (Z_{50}) is determined as occuring to the more probable dilution by the PROBIT [4] statistic at the level (1- α) matching 0,95. Q and Qod are respectively the inodor air and the odorous sampling flow-rates delivered by the olfactometer. This measure is independent of the odour quality.

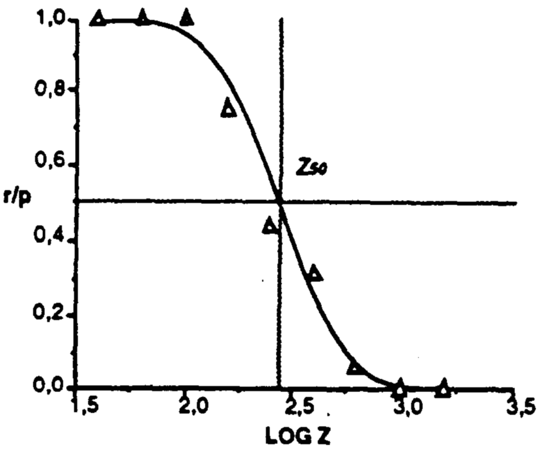


Fig. 2 - Example of an odour curve plotting and odour concentration determination: (△) Experimental response of the panel (positive perception); (–) Statistic fitting for Z_{50} determination (PROBIT): Z_{50} = 274 u.o./m^3

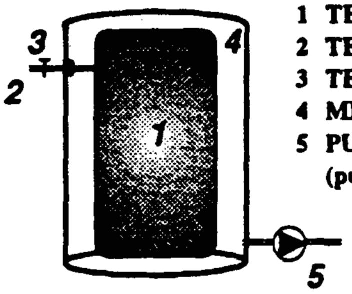
The above procedure is useful to determine the odours annoyance and allows for comparing levels of odours concentrations. It is important to determine which compound or group of compounds is responsible for the odour annoyance. This information can only be obtained by means of high sensitive chemical analysis like chromatography coupled with mass spectrometry (GC-MS).
Correlations can be made in such a way between the air sample analyzed and the applyed paint. So, paint analysis are made in order to determine which compounds are emitted during application by means of dynamic headspaces through the paint.

The pOU illustrates the individual contribution of each volatile compounds identified in the perceived odour. Following its definition it is expressed in term of odour decibel (dBo) as

$$\Sigma\, pOU_i = \Sigma \log(\frac{C_i}{d_i}) \qquad [dBo]$$

where C_i and d_i are respectively the detected concentration of the volatile compound and its individual odour perception threshold (pOU = 0 when $C \leq d$).

Sampling is made via TEDLAR bags which give, with a minimum of precautions, accurate results. Bags are filled as much as possible by a direct vacuum system shown at figure 3.



1 TEDLAR BAG
2 TEFLON TUBE
3 TEFLON VALVE
4 METAL DRUM
5 PUMPING SYSTEM
(pump, battery, flexible tube, flowmeter)

Fig. 3 - Vacuum sampler

Both samples for olfactometry or for chemical analysis are made this way. For chemical analysis, several milliliters of gas are passed through an adsorbent cartridge before to be put into a Thermal Cryogenic Trap (TCT) system as injection device into the GC-MS. Quantification is made in reference to standards. Detection limit of the system is in the range of 0,01 - 0,1 ppb (or µg/m^3).

2.3. Paint composition

Solvent phase compositions for equivalent organic/water paint systems are given in the table I (from PPG [5]).

Table I - Comparison of paint compositions for equivalent organic/water systems

Paint	ESP %	Coverage µ.m²/kg	SOLVENT COMPOSITION (% Vol)				
			aromatics	alcohols	esters	ketones	water
PRIMERS							
OBS-Epoxy	51	310	64	30	-	6	-
W-A-Epoxy	43	310	-	29	7	-	64
TOP COATS							
OBS-P E	63	350	59	24	17	-	-
W-Acrylic	60	350	-	27	-	-	73
BACKERS							
OBS-Epoxy	50	320	52	29	10	9	-
W-A-Epoxy	49	310	-	32	-	9	59

	Compounds	Bulk %
1	Isobutanol	0,36
2	n-Butanol	2,67
3	Benzene	8,55E-04
4	2-Butanone	0,4
5	Ethyl glycol acetate	0,01
6	1-Methoxy 2-Propanol	3,86
7	Toluene	7,65
8	3-Hexene-2-one	0,4
9	Cyclohexanone	0,76
10	4-Methyl 2-Pentanone	0,12
11	Σ C7 alkanes (Heptane incl.)	1,75E-03
12	Ethylbenzene	3,67
13	m+p-Xylenes	5,97
14	o-Xylene	2,37
15	5-Methyl 2-Hexanone	1,19
16	Σ C8 alkanes	2,18E-03
17	Indene	0,02
18	4-Methyl 4-Hydroxy 2-Pentanone	4,7
19	Isobutyl acetate	0,03
20	Butyl acetate	0,5
21	Methyl styrene	0,67
22	2-Butoxy ethanol	0,1
23	Methyl Cellosolve acetate	2,12
24	Σ C3 Alkylbenzenes (=)	0,48
25	Σ C3 Alkylbenzenes	31,11
26	Naphtalene	0,3
27	Σ C9 alkanes	0,09
28	1-Methyl 2,3-Dihydroindene	4,51E-03
29	Cellosolve acetate	-
30	Σ C4 Alkylbenzenes (=)	0,54
31	Σ C4 Alkylbenzenes	21,36
32	Isophorone	0,73
33	Σ C10 alkanes (Decane incl.)	0,39
34	Ethylene Glycol diacetate	0,47
35	Σ C5 Alkylbenzenes	1,46
36	Σ C11 alkanes	0,07
37	2-(2-Butoxyethoxy) ethanol	0,09
38	Butyl diglycol acetate	0,3
39	Ring ether	3,8
	SOLVESSO/SHELLSOL	55,27

As illustrated by tables II and III, the composition of OBS-paints can be complex because of the great use of petroleum cuts (Solvesso/Shellsol) as solvent. The choice of the organic solvents is more functional for W-paint.

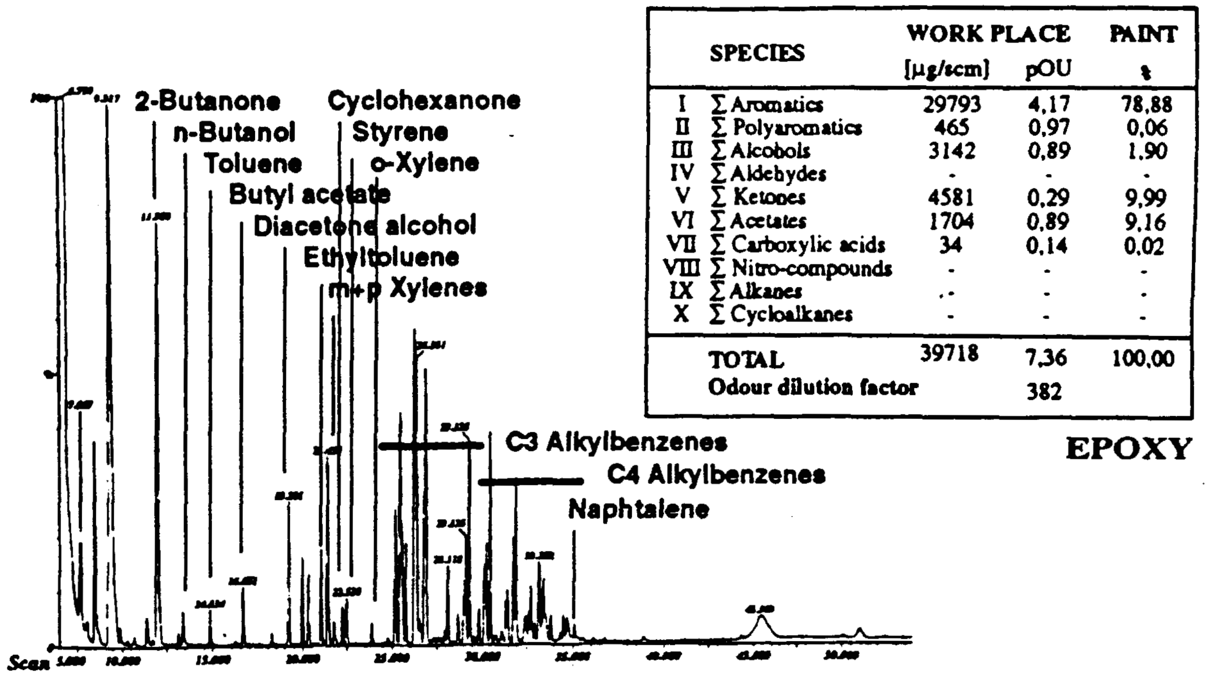
COMPOUNDS	Bulk (%)
Butyl Glycol	25
n-Butanol	23
Toluene	16
Methyl Pirrolidinone	14
Butoxyethoxy ethanol	14
Amines and surfactants	9

Table II - Typical organic solvent content of a W-paint

Table III - Compilation of solvent phase content for based organic solvent paints [6]

3. Comparison of paint cabin ambiences

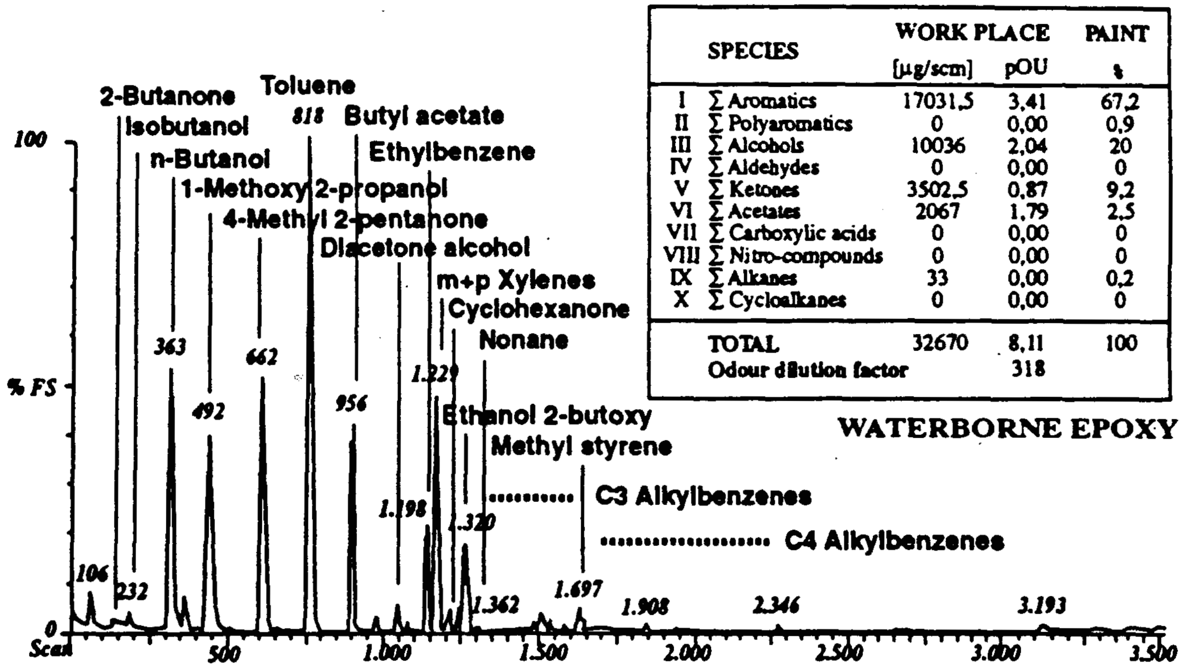
This section compares the emissions occuring during the primer (or backer) coat application of an OBS-epoxy and a waterborne one. Analysis for the W-epoxy was made during the application of a "semi-classic" paint system (primer and top coats being OBS and backer being a W-paint).



	SPECIES	WORK PLACE [µg/scm]	WORK PLACE pOU	PAINT %
I	Σ Aromatics	29793	4,17	78,88
II	Σ Polyaromatics	465	0,97	0,06
III	Σ Alcohols	3142	0,89	1,90
IV	Σ Aldehydes	-	-	-
V	Σ Ketones	4581	0,29	9,99
VI	Σ Acetates	1704	0,89	9,16
VII	Σ Carboxylic acids	34	0,14	0,02
VIII	Σ Nitro-compounds	-	-	-
IX	Σ Alkanes	-	-	-
X	Σ Cycloalkanes	-	-	-
	TOTAL	39718	7,36	100,00
	Odour dilution factor		382	

EPOXY

Fig. 4 - Chromatogram of a work ambience during the application of a OBS-EPOXY as primer coat (recapitulative table of quantitative results in locket)



	SPECIES	WORK PLACE [µg/scm]	WORK PLACE pOU	PAINT %
I	Σ Aromatics	17031,5	3,41	67,2
II	Σ Polyaromatics	0	0,00	0,9
III	Σ Alcohols	10036	2,04	20
IV	Σ Aldehydes	0	0,00	0
V	Σ Ketones	3502,5	0,87	9,2
VI	Σ Acetates	2067	1,79	2,5
VII	Σ Carboxylic acids	0	0,00	0
VIII	Σ Nitro-compounds	0	0,00	0
IX	Σ Alkanes	33	0,00	0,2
X	Σ Cycloalkanes	0	0,00	0
	TOTAL	32670	8,11	100
	Odour dilution factor		318	

WATERBORNE EPOXY

Fig. 5 - Chromatogram of a work ambience during the application of a W-EPOXY as primer coat (recapitulative table of quantitative results in locket)

Measurements were lead into two different coating plants; the air dilution flow-rate around the paint machine corresponding to the figure 1 (OBS-epoxy) was exactly the double of the flow-rate value corresponding to figure 2 (W-epoxy). So it can be seen

that the same levels of concentration and odours are reached when using the waterborne paint although the air dilution flow-rate was reduced by a factor 2.
The chromatograms reflect very well the differences between the two systems. It can be pointed out that the overall composition of W-paints is in accordance with the formulation given by the manufacturer only from a qualitative point of view.

On the basis of those results it was decided to begin an ECSC research to study [2] the application of a full waterbone system.

4. Full waterborne system

4.1. *Results of measurements*

Trials were led on the site of Onnaing (France) with a system constituted of three coats (primer, backer and top coat). Primer and backer are waterborne epoxies whereas top coat was an acrylic one. Table IV summarizes the obtained results on the three work place areas whereas the Table V summarizes the obtained results on both sides of the two incinerators (inlet and outlet).

The chromatogram at figure 6 shows the GC-MS analysis on the inlet effluent of the incinerator treating emissions from ovens drying and curing backer and top coats (respectively, chromatograms shown at fig. 7 and 8).
The chromatogram (figure 8) obtained for the analysis of the top coat paint (waterborne acrylic) can be compared with the chromatogram (figure 9) obtained for the analysis of a based organic solvent paint (polyester).

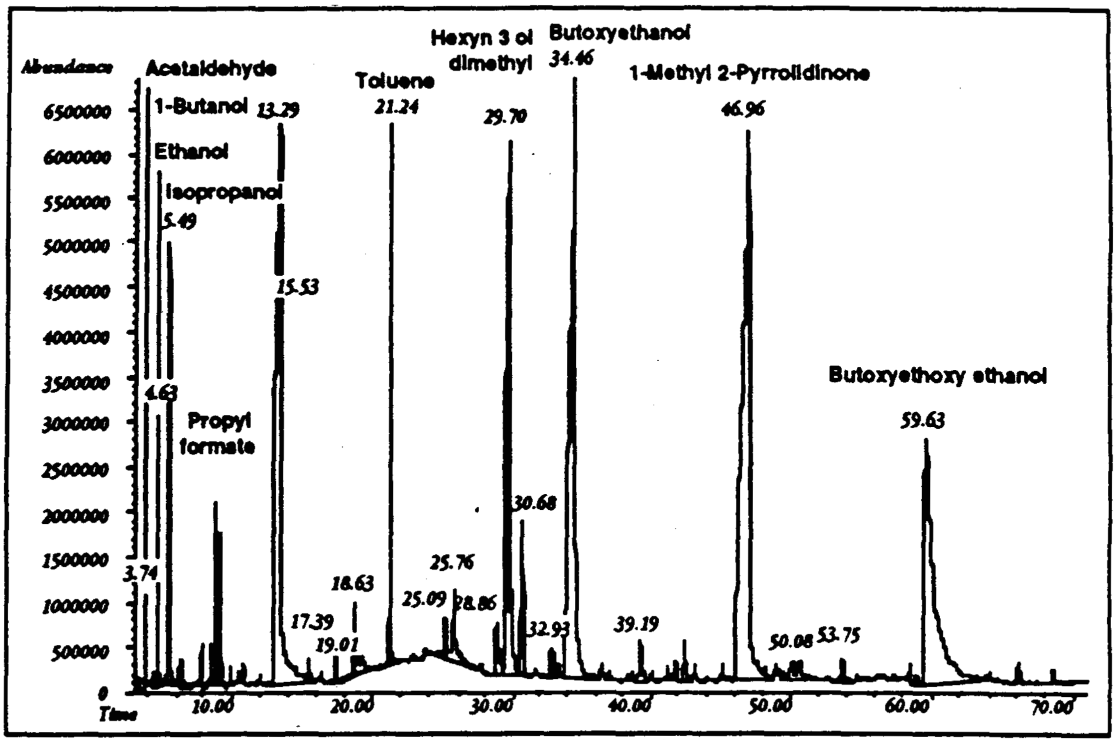


Fig. 6 - Chromatogram of the incinerator inlet: loaded gas effluent by backer and top coats solvents from curing oven

SPECIES	BACKER				TOP COAT				PRIMER			
	Work place			Paint	Work place			Paint	Work place			Paint
	$[\mu g/m^3]$	Σ pOU	%	%	$[\mu g/m^3]$	Σ pOU	%	%	$[\mu g/m^3]$	Σ pOU	%	%
Σ Aromatics	4017,4	2,36	86,6	10,2	17854,6	5,7	45,8	5,8	7280,6	1,8	66,8	11,1
Σ Alcohols	64,0	0,00	1,4	57,5	14869,6	1,7	38,2	68,2	3200,4	1,2	29,4	82,5
Σ Aldehydes	0,0	0,00	0,0	0,0	21,1	1,4	0,1	0,0	2,1	0,4	0,0	0,0
Σ Ketones	312,7	0,00	6,7	0,4	4559,7	0,2	11,7	2,4	334,4	0,0	3,1	0,8
Σ Acetates	163,9	0,00	3,5	0,2	1068,1	3,4	2,7	23,6	59,6	0,8	0,5	5,5
Σ Nitro compounds	0,0	0,00	0,0	30,3	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0
Σ Alkanes	78,4	0,00	1,7	0,0	532,0	0	1,4	0,0	16,0	0	0,1	0,0
Σ Ethers	0,0	0,00	0,0	1,5	54,9	0,0	0,1	0,0	7,5	0,0	0,1	0,0
Sums	4636,3	2,36	1,7	1,5	38960,0	12,5	1,5	0,0	10900,7	4,3	0,2	0,0
u.o./m 3	70				293,0				379			

Table IV - Ambiences analyses at work places compared with headspace paint analyses

SPECIES	PRIMER INCINERATOR							TOP AND BACKER INCINERATOR						
	INLET			Paint	OUTLET		ε	INLET			Paint	OUTLET		ε
	$[\mu g/m^3]$	Σ pOU	%	%	$[\mu g/m^3]$	Σ pOU	%	$[\mu g/m^3]$	Σ pOU	%	%	$[\mu g/m^3]$	Σ pOU	%
Σ Aromatics	49527,9	7,86	13,4	11,1	1068,6	0,05	97,9	70894,3	8,21	11,3	7,9	2026,6	0,44	99,2
Σ Alcohols	248522,7	6,37	67,4	82,5	0,0	0,00	100,0	333599,0	7,05	52,9	63,1	296,9	0,00	99,9
Σ Aldehydes	22169,6	6,23	6,0	0,0	240,8	0,54	98,9	14390,0	8,65	2,3	0,0	120,3	1,40	99,2
Σ Ketones	21580,0	2,53	5,9	0,8	1113,7	0,00	94,8	13338,9	1,91	2,1	1,4	187,5	0,00	98,6
Σ Acetates	20754,6	5,07	6,6	5,5	0,0	0,00	100,0	8777,5	4,90	1,4	12,5	21,7	0,00	99,8
Σ Nitro compounds	2830,2	0,00	0,8	0,0	782,6	0,00	-	177580,0	0,00	28,2	14,4	150,1	0,00	80,5
Σ Alkanes	294,8	0,00	0,1	0,0	5508,5	0,00	-	11515,1	0,00	1,8	0,0	31,3	0,00	99,7
Σ Ethers	2948,1	0,00	0,8	0,0	0,0	0,00	100,0	0,0	0,00	0,0	0,7	0,0	0,00	-
Sums	368627,9	28,06	0,9	0,0	8714,3	0,60	97,6	630094,7	30,72	1,8	0,7	2834,4	1,84	99,6
u.o./m 3	8444				884		89,5	19612,5				1252		93,6

Table V - Incinerators effluents analyses + abatement efficiencies compared with headspace paint analyses

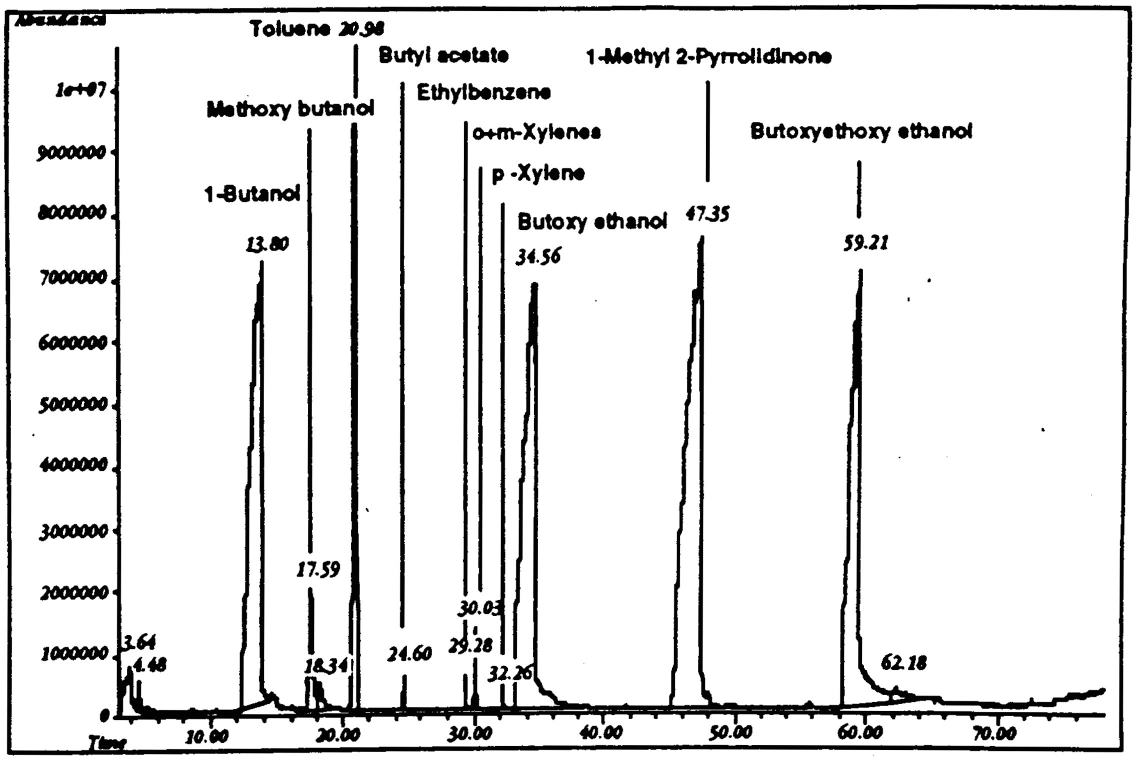


Fig. 7 - Chromatogram of back coat analysis (W-epoxy)

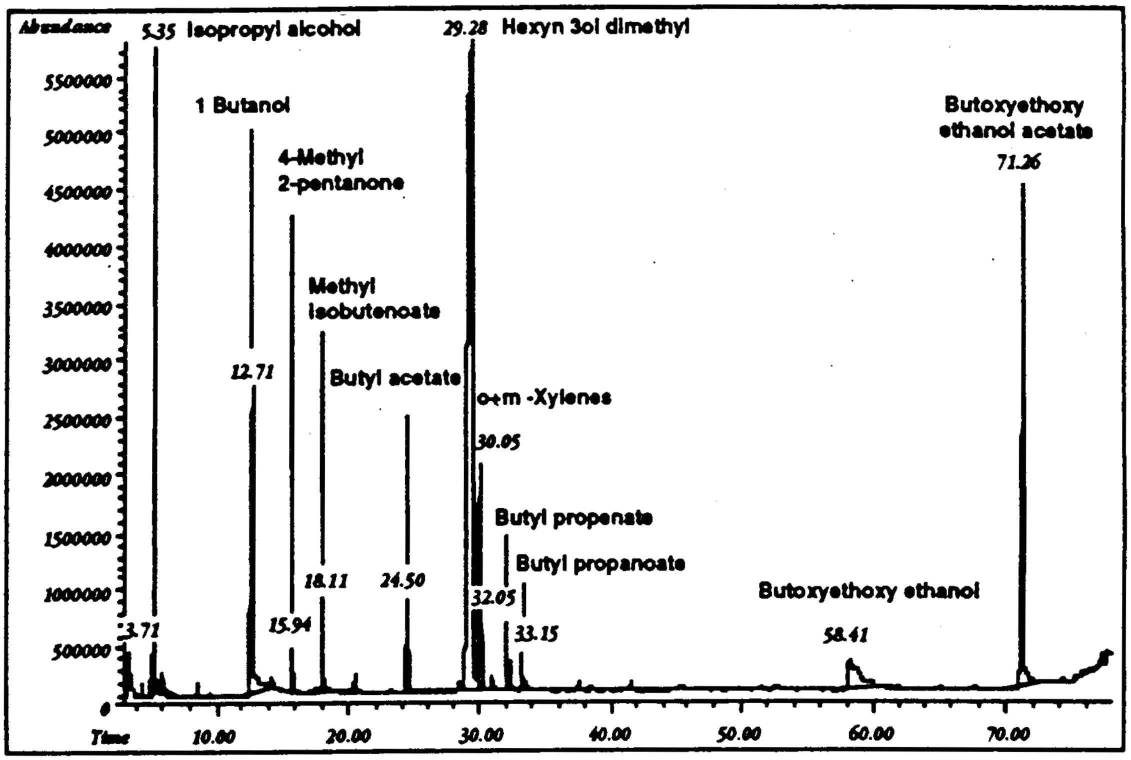
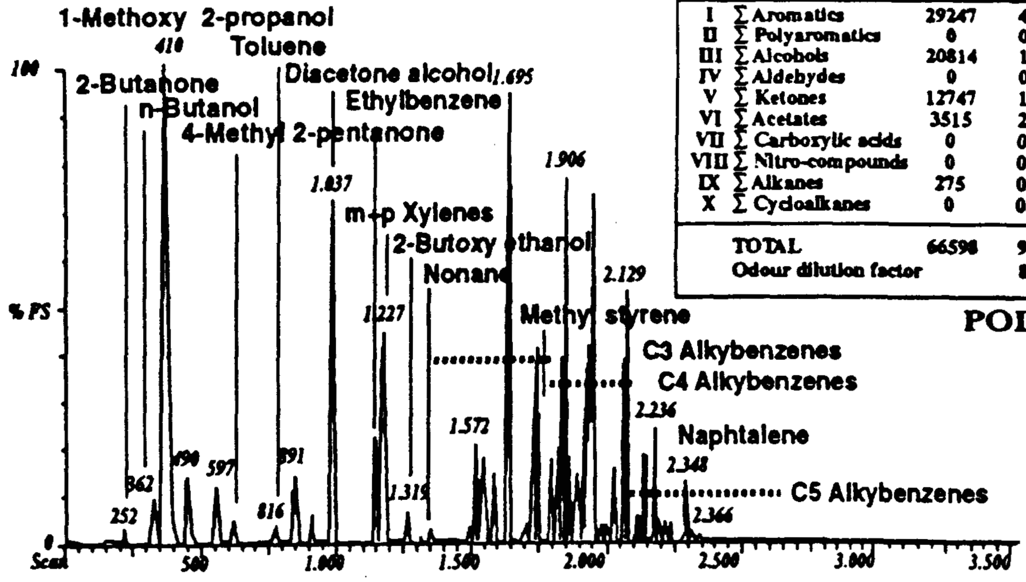


Fig. 8 - Chromatogram of top coat analysis (W-acrylic)



	SPECIES	WORK PLACE [µg/Nm3]	WORK PLACE pOU	PAINT %
I	∑ Aromatics	29247	4,07	58,8
II	∑ Polyaromatics	0	0,00	0
III	∑ Alcohols	20814	1,56	39
IV	∑ Aldehydes	0	0,00	0
V	∑ Ketones	12747	1,77	0,9
VI	∑ Acetates	3515	2,01	1,2
VII	∑ Carboxylic acids	0	0,00	0
VIII	∑ Nitro-compounds	0	0,00	0
IX	∑ Alkanes	275	0,00	0,1
X	∑ Cycloalkanes	0	0,00	0
	TOTAL	66598	9,42	100,00
	Odour dilution factor		864	

POLYESTER

Fig. 9 - Chromatogram of top coat analysis (OBS)

4.2. Discussion

The table VI compares the levels of emissions occuring at work places during the application of a full OBS-system and a full waterborne one.

Sampling	[µg/m3]	∑ pOU (dBo)	u.o./m3
	OBS System		
Primer (EPOXY)	39718	7,36	382
Top Coat (P.E.)	30024	7,04	280
INCINERATOR inlet	637757	12,28	13083
outlet	19000	4,71	671
	W-System		
Primer (EPOXY)	7768,5	4,3*	224,5
Top Coat (P.E.)	38960	12,5	293
INCINERATOR inlet	499361,5	30,72*	14028,25
outlet	5774,5	1,84*	1068

*Maximum value

Table VI - Summary of the comparison between emissions generated by a OBS-system [7] and a W-system.

The value for the ventilation flow-rate of paint cabin corresponding to the OBS-system is the double of the flow-rate corresponding to the cabins where W-system was applied.

It can be pointed out that noticeable reductions of emissions are obtained when using W-system: the level of concentrations are equivalent even if the ventilation flow-rate is double for the OBS-system.

However major differences can be seen from work places and incinerators effluents analysis.

At work places the participation of VOC's species at the global concentration is not in accordance with the VOC's species percentage given by the headspace analysis of the paint, at the opposite of OBS-system analysis. Although alcohols are the main organic solvents in W-paints, aromatics predominates in the ambience composition, probably because the components of this latest species are not soluble in water.

So if W-system is used to reduce the ventilation air flow-rate at the work place ambiences by keeping the same level of concentration than for a OBS system, the two systems are equivalent from a toxicity point of view. To a same level of concentration as measured in this study, W-system does not allow to reduce significantly odours concentrations.

At incinerator inlet (corresponding to the ovens outlet) and outlet, the difference consists in the presence of a greatest quantity of by-products (by partial oxidation or recombination) than for using a OBS-system. Furthermore, some by-products like nitro-compounds are not found with OBS-systems.

The first category of identified by-products are aldehydes. Aldehydes are mainly the product of the partial oxidation of the alcohols and their content into ovens and incinerators effluent outlet is obviously increased since the greatest percentage of alcohols involved in the W-paint formulation: from traces detected for OBS-system (bulk percentage ≤ 0,1 %), aldehydes concentration can reach several %. They are 100 time more odorous than alcohols and it implies greater values in dBo (odour power) but the VOC's blending effect limits very well the increasing of the odour concentration.

The second category of by-products are nitro-compounds: 2-Methyl Pyrrolidinone can be a major component of solvent phase of a W-paint. This compound is used as a reaction susceptor during curing and fully participates as a linking agent. Some quantities of this compound can however be retrieved at the incinerator inlet (0,1 % of the bulk concentration to be compared with the 30 % of the bulk composition of the organic solvents of the backer W-paint, see tables IV-VI). The thermal degradation of this component is easy but susceptible to give by-products like alkyl nitrates, nitriles and nitroalkanes. Formation of HCN is not impossible and precautions must be especially observed to maintain the incinerator temperature at a sufficient level (≥ 700 °C). No traces of nitro-compounds were found at work place ambiences.

5. Implication on the abatement techniques

They are no major disturbances to use W-system with conventional thermal incinerators but because of the presence of greater quantity of by-products precautions must be taken to keep a sufficient high temperature. Investigations led on a pilot catalytic incinerator show that no problem occurs when W-system is used.

Use of W-paints allows a deep reorientation of abatement technologies because a lot of parameters is significantly modified. Techniques such as biofiltration, adsorption, scrubbing and condensation, membrane separation can be investigated. As a matter of fact, the major identified compounds at the inlet of incinerators are ethanol (1,5-3%), n-butanal (1,3-5,1%), 1-Butanol (14,1-22,8%), toluene (6-8%) hexyn-ol-dimethyl (11 %), 2-butoxyethanol (22-29%) and 2-Pyrrolidinone 1-Methyl (0,6-28,1%). From one paint to another, the ranges of percentage (except for the latest compounds) are close. Global percentage in aromatics is included in the range 11,3-13,4%. The limited number of VOC compounds allows for recovery of solvent by adsorption or condensation. The more xenobiotic compounds for a biofiltration system are constituted by aromatics (10%); otherwise the level of concentration is not too high and a higher water content is present.

The main subsisting problem to allow such techniques is the oven temperature level. Almost all the recovery techniques needs an inlet effluent temperature reaching no more than 40 °C to be efficient. Condensation and membrane separation are the two opposite exceptions: condensation needs cryogenic temperature whereas membrane technology is promising to be used at temperature in the range of 200-250 °C.

The response at this problem is coming from new development of electrical heated ovens using magnetic induction properties [8] from which outlet effluent temperatures are liwer than 80 °C.

6. Availability

Waterborne paints are used as primer, backer and topcoat but only for building end uses, i.e. cladding, siding and roofing of industrial, commercial and residential buildings. These coatings are available only in low or semi-gloss finishes on various substrata : aluminium, hot dip galvanized steel, ...

The durability of waterborne topcoats is excellent; waterborne finishes can be classified as very high durable coatings second only to fluorocarbons.

Building market represents about the third of the coil-coating activity so W-systems can be used in large quantities.

For the full scale test done at Sollac plant, some problems about viscosity and pigments decantation into the paint barrels, emulsion at rolls application (craters formation on the surface of the coating) and vaporization rate must be solved.

7. Conclusions

The advantages and the limitations of a waterborne system in comparison with a classical based organic solvent one are well shown in this study. Full W-system allows for reducing by two the needed flow-rate both for paint cabin or area ventilation and for air dilution through the ovens and subsequently through the incinerators. Indeed, the level of odours and VOC's concentrations is lower. In such conditions it was demonstrated in a previous paper [5] that energy consumption can be kept constant or only lightly increased.

Waterborne systems are waiting for the industrial request which can spread their availability for a large range of coating. They offer new possibilities for a more environment-friendly coating operation by example by using recovery techniques for solvent reuse.

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