

**Final Background Document
on the sector**

Speciality organic chemical industry

Prepared in the framework of EGTEI

Prepared by CITEPA, Paris

1. Data from the bibliography (p.3)

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The speciality organic chemical industry covers the production of different types of chemicals produced in campaign basis, in multipurpose and multiproduct plants (pharmaceutical active ingredients, biological products, food additives, photographic chemicals, dyestuffs and intermediates, pesticides and other speciality products...).

The pharmaceutical product manufacturing is covered by SNAP 06 03 06 or NFR 3C Chemical product manufacture and processing.

The production of other types of products is not well represented in the SNAP. Depending on each Party, emissions from this activity could be encountered in SNAP codes like 060314: other chemical products manufacturing or processing as carried in the French emission inventory.

The pharmaceutical product manufacturing covers:

- The production of primary pharmaceutical products: production of bulk pharmaceuticals, drug intermediates and active ingredients by means of synthesis, fermentation, extraction or other processes, in multipurpose and multiproduct plants and on a campaign basis.
- The activities related to formulation of finished drugs and medicines using the active ingredients supplied by the bulk plants (taking place in finishing plants). Active ingredients are converted into products suitable for administration. Physical formulation, filling and packaging are involved.

These two activities are very different in term of production techniques used. For the determination of NMVOC emission reduction costs, only the active ingredient production is considered. No data have been obtained for formulation activities but it can be assumed that costs collected for the production of primary pharmaceutical products can be used for representing these activities.

During the meeting of 22/07/02 between CITEPA and the French Union of Chemical Industry (UIC) [14], it was decided to consider the pharmaceutical product manufacturing as the representative activity for the whole speciality organic chemistry. It is consequently assumed that costs derived for the pharmaceutical product manufacturing are representative of the whole speciality organic chemistry. The document has been prepared on the basis of information provided by the SICOS : French Association of the Organic Chemistry and Biochemistry Industries.

Activity : solvent input (new + recycled solvent)

Pollutant : NMVOC

1 Data from the literature

1.1 Data currently used in the RAINS model

At its present stage of development, the RAINS sector PHARMA represents the pharmaceutical product manufacturing. The activity level is represented by the quantity of solvent consumed [9].

1.1.1 Control options

Two options of reduction are considered: combination of primary measures (good housekeeping and solvent management plans) with add-on techniques such as incineration and adsorption (INC/ACA). The considered applicability factor ranges from 60 to 100 %, according to parties and the technical efficiency ranges from 80 % to 90 % (in tables 1.1 to 1.3, three examples are given. They do not represent the entire range of parameters presented in this paragraph).

1.1.2 Abatement measure costs

Costs of the combination of techniques range from 2500 to 6000 €₁₉₉₀ / t NMVOC non-emitted.

Costs used for some Parties are presented in tables 1.1.2.1 to 1.1.2.4.

Table 1.1.2.1: Costs data used in the RAINS model [9] for France

	Emission factors kg NMVOC/kg solvent	Applicability %	Technical efficiency %	Cost €1990/t NMVOC
France: activity level 1990: 60.6 kt solvent, 2010: 71.45 kt solvent; NMVOC emissions scenario business as usual: 1990: 9.0 kt, 2010: 6.6 kt				
No control	0.148			
HSE +ACA	0.0653	70	80	2 783
HSE + INC	0.0549	70	90	2 851

Table 1.1.2.2: Costs data used in the RAINS model [9] for Germany, old Landers

	Emission factors kg NMVOC/kg solvent	Applicability %	Technical efficiency %	Cost €1990/t NMVOC
Germany: activity level 1990: 233.33 kt solvent, 2010: 466.66 kt solvent NMVOC emissions scenario current regulation: 1990: 35.0 kt, 2010: 35.9 kt				
No control	0.15			
HSE +ACA	0.0587	70	87	2 523
HSE + INC	0.0587	70	87	2 920

Table 1.1.2.3: Costs data used in the RAINS model [9] for Germany, new Landers

	Emission factors kg NMVOC/kg solvent	Applicability %	Technical efficiency %	Cost €1990/t NMVOC
UK: activity level 1990: 35.0 kt solvent, 2010 70.0 kt solvent NMVOC emissions scenario current regulation: 1990: 5.25 kt, 2010: 5.38 kt				
No control	0.15			
HSE +ACA	0.0587	70	87	2 523
HSE + INC	0.0587	70	87	2 920

Table 1.1.2.4: Costs data used in the RAINS model [9] for Hungary

	Emission factors kg NMVOC/kg solvent	Applicability %	Technical efficiency %	Cost €1990/t NMVOC
UK: activity level 1990: 0 kt solvent, 2010: 0 kt solvent NMVOC emissions scenario business as usual: 1990: 0.01 kt, 2010: 0.02 kt				
No control	0.150			
HSE +ACA	0.0587	100	87	2 523
HSE + INC	0.0587	100	87	2 920

HSE : Good house keeping and solvent management plans

ACA : Activated carbon adsorption

INC : Incineration

Cost data are available for other Parties. They can be downloaded on the web site of IIASA [13]. Investments and operating costs are not more detailed in the current documents. According to information received from IIASA, costs have been mainly derived from study [10].

1.2 Remarks on Entec Study [10]

In the Entec study [10], costs of application of the NMVOC directive in England are compared to a reference base line, which integrates the application of the IPPC directive. The IPPC directive is assumed to abate emissions from this industry by 43 %. Application of the NMVOC directive is assumed to abate NMVOC by 56 %. The costs presented in the Entec study are those involved to achieve the additional emission reduction of 13 points of %. This explains the average high costs of this study. An average cost of 4 360 €/t NMVOC is presented in [10]. According to the survey carried out, some industries explained that abatement measures had an economic impact; other ones, on contrary, would be able to meet the total NMVOC emission limits of the directive with a limited economic impact. The costs involved by the reduction of the first tonnes of NMVOC are lower than costs for reducing the last potential tonnes of NMVOC. These first costs have not been considered in study [10].

Costs from the Entec study are not helpful for EGTEI. Transparency on their derivation is lacking.

2 Short technology description

This industry is very heterogeneous: plants manufacture a large range of products, using a large number of production processes and may store and use several hundred raw material substances or intermediate products. Processes are usually operated on a campaign basis and in multipurpose plants. For one active ingredient, several transformation stages are required. The processes typically involve between 1 to 40 transformation stages depending on molecules. Process stages cover the full range of unit operations such as: reactions, liquid/liquid extraction, liquid/liquid or liquid/solid or gas/solid separation, distillation, crystallisation, drying, gas adsorption... Production is carried out in discontinuous processes (or batch processes). Equipment is rarely specific but, most often, multi-application. Processes frequently use solvents. Any reacted raw materials may be either recovered or recycled or ultimately discharged to the environment after appropriate treatment [7].

As said in reference [7], because of the diversity of processes used in this sector, no simple process description can be made. Instead, a brief outline of characteristics of existing pharmaceutical product production plants is provided [2, 3 and 6].

- ***Significant number of NMVOC-emission release points***

Gaseous discharge circuits are complex. For the same equipment, several discharge points do exist, depending on the performed operations. The large number of discharge points is due to:

- Quality constraints required in this sector in order to avoid risks of cross-contamination,
- Security constraints in order, for example, to avoid the contact of incompatible gases.

Plants having an annual solvent consumption ranging from 900 to 1 500 t may have from 10 to 50 NMVOC emission discharges in the atmosphere.

A large number of discharge points are equipped with condensers to trap NMVOC. To trap corrosive or toxic gases, several vents are related to abatement absorption columns. When secondary abatement techniques are applied, collecting the vents proves to be necessary.

- ***High variability of NMVOC discharges with the time***

NMVOC concentrations may vary widely from one discharge point to another. Discharges with high waste gas flow rates and low concentrations do exist; general ventilation of a factory belongs

to this group. Other discharges, such as production equipment vents are characterised by very low waste gas flow rates (some Nm³/h) and NMVOC concentrations that may be high.

NMVOC discharges present a very high variability: high variability with time when there is a discharge and non-permanent discharges.

This situation leads to more significant costs for emission treatment: the gas-cleaning device should be able to accept emission peaks. Abatement technique dimensioning must be based on the peak discharge (the frequency of peaks should be considered as well). Investments are thus higher than for more regular emissions in time.

- ***A large number of solvents used***

In this activity, even though 5 solvents (methanol, toluene, acetone, ethanol, methane dichloride) represent about 70 % of the new solvent consumption, around 40 different solvents are in use. In France for example [6], the consumption of chlorinated solvents represents about 20 % of the total consumption of solvents. This large number of solvents, the presence of chlorinated solvents and security and quality constraints make the use of secondary abatement techniques more difficult and more expensive (treatment of HCl if incineration, limited potential for collection and recycling of solvents).

3 EU regulation : Directive 99/13 of 11 march 1999

Only the production of pharmaceutical products is considered by the EC Directive. The solvent consumption threshold is 50 t / y.

Operators concerned can conform to the Directive in either of the following ways:

- Option 1: by complying with both the NMVOC emission limit values in residual gases and the fugitive emission limit values,
- Option 2: by complying with the total emission limit values.

Emission limits are presented in table 3.1.

Table 3.1: Emission limits implemented by Directive 99/13

	Option 1		Option 2
	NMVOC emission limit in residual gases mg C / n m ³	Fugitive NMVOC emission limit % of solvent input	Total NMVOC emission limit % of solvent input
New installations	20	5	5
Existing installations	20	15	15

The Directive requirements are not all described in this chapter. In this activity, R40 halogenated solvents or R45 (or R61...) solvents can be used. Regulatory constraints for these compounds are more tightened than for common solvent without this risk phrases.

4 Definition of reference installations

In this activity, reference installations cannot be characterized by the active ingredient productions. These data are not available and most often process routes for producing different active ingredients are totally different. The production rate ranges from some kilograms to several thousands tons.

In this sector, a reference installation can be characterized by its annual solvent input. This input corresponds to the quantity of bought solvents (new solvents) plus the quantity of recycled solvent. This definition is in accordance with the expression of total emissions used in the SED directive (appendix III).

Only one reference installation is taken into account since, for the time being, costs expressed in €/ t NMVOC abated are equivalent for all the sizes of installations initially which could have been defined (see chapter 7, explanatory notes).

Table 4.1: Definition of reference installations

Reference Installation Code (RIC)	Description
01	<u>Medium Installation:</u> annual solvent input (new + recycling) : 3 000 t/y representative for a range between 1 000 and 5 000 t/y

5 Emissions abatement techniques and costs

5.1 Definition of primary measures and secondary measures

In order to reduce solvent losses and emissions into the atmosphere, a wide range of best practices and process improvements are possible and have been implemented in plants several years ago. These measures aim at containing NMVOC emissions. Such measures include [2, 3, 6, 8] for example (the list presented is not exhaustive):

- Work in concentrated environment in order to reduce the consumption of solvents,
- Increased use of low volatile solvents and of solvents easier to condense,
- Modification of certain operating conditions for distillation (e. g. distillation under ordinary pressure instead of vacuum distillation),
- Implementation of good housekeeping, increased condenser efficiency (increased exchanger surfaces and increased refrigerating capacities),
- Technology change: dry-sealed vacuum pumps instead of liquid ring vacuum pumps; closed pressure filters or vacuum filters more leak free than open filters; vacuum dryers leading to a better solvent condensation...

According to [2, 3], the above mentioned measures allow a significant NMVOC emission reduction.

Remark:

Good housekeeping includes [1], [6]:

- Better controlling of feed rate, mixing, temperature as well as other reaction parameters (pressure control to minimize nitrogen consumption and associated losses from reactors...),
- Optimisation of process parameters,
- Effective production and maintenance scheduling,
- Improved material handling and storage procedures,
- ...

No unique abatement technique can be implemented in a general way in all plants, due to the diversity of situations. Consequently, secondary abatement techniques which could be applied in pharmaceuticals production plants are not defined separately: since it is difficult to determine the implementation potential of each of these reduction technologies, secondary measure 01 takes into account the use of several techniques: thermal incineration, condensation, activated carbon adsorption, absorption.

Conclusion

According to information received from [2, 3 and 6], three situations may be considered:

- Installations emitting more than 15 % of the solvent input: an average value of 30 % is taken into account. This corresponds to primary measure 00 where no specific primary controls nor secondary measure are used,
- Installations emitting between 5 to 15 % of the solvent input: an average value of 8 % is taken into account. This corresponds to primary measure 01 and secondary measures 01.
- Installations emitting less than 5 % of the solvent input: an average value of 3.5 % is taken into account. This corresponds to primary measure 02 and secondary measures 02 (see below).

In this case, the two options cannot be cumulated.

Table 5.1.1: Measures adopted

Primary Measure Code PMC	Secondary Measure Code SMC	Description
00	00	Conventional primary measures and no secondary measure
01	01	Primary measure program 1 and low use of secondary measures
02	02	Primary measure program 2 and high use of secondary measures (both Incineration, adsorption and / or condensation)

*For a detailed description, please refer to the above section

5.2 Emission factors, cost data and uncertainties of the different combinations

A compromise has been adopted between the requirement of simplicity in the description of the activity and its effective complexity. We have to bear in mind that it is impossible to describe the complexity of this activity. Our aim is to obtain quite good representative costs (All following data are more detailed in chapter 7 : explanatory notes).

Table 5.2.1: Emission factors, investments, operating costs, abatement efficiencies and uncertainties for relevant combinations (€2000)

Combination Code	Emission Factor for NMVOC [kg/t solvent used]	Abatement efficiency [%]	Quality of data	Confidential interval [%]
01 00 00	300		4	50
01 01 01	80	73	4	50
01 02 02	35	88	4	50

Combination Code	Investment [€]	Fixed operating cost [€y]	Variable operating costs [€y]	Quality of cost data	CI %
01 00 00	0		0	0	
01 01 01	2 640 000	132 000	316 800	3	100
01 02 02	6 678 000	333 900	675 650	4	100

Table 5.2.2 presents marginal costs of the two options.

Table 5.2.2: Marginal costs of different options

Combination Code	Total marginal costs [€/tNMVOC]
01 00 00	0
01 01 01	1 173
01 02 02	2 305

6 Data to be provided by national experts for the completion of the data basis for their country

The following tasks are required:

The following tasks are required:

6.1 Validation work

The national expert is invited to comment the methodology defined by the secretariat.

- Validation of the default investment costs provided,
 - Validation of the method of derivation of operating costs,
- Or
- Provide other costs for the same combination of techniques and justify them.

Comments have to be sent to the secretariat in the two weeks after electronic publication of the document

6.2 Provision of specific data

Tables to be filled in with country specific data by national experts

- Determination of country specific data to calculate variable costs (*Specific values for energy, man power... are valid for all NMVOC sectors and have to be entered in the tool only once*)

Table 6.2.1: Country specific data

Parameters	Country specific costs
Electricity net of taxes [€/kWh]	
Natural gas net of taxes [€/kWh]	
Wages [€/h]	

For this activity, operating costs are not country specific

- Total activity level in accordance with units used in the document: solvent input: new solvent + internally recycled solvent (This activity level should be coherent with the activity level used for emission inventories delivered for UNECE).

The solvent input to be considered is the total solvent input for the speciality organic chemistry. Table 6.2.2 must be completed. Some default values for the confidence interval are provided. They can be used by one Party if no data is available.

The methodology used in Rains for estimating the future activity level will be described in the methodology. It can be used or information can be obtained from the industry.

Table 6.2.2 : Activity levels on reference installations - Solvent input t/y

RIC	2000	CI%	2005	CI%	2010	CI%	2015	CI%	2020	CI%
01										
Default values proposed for CI		10		20		50		100		100

For explanation on the confidence interval (Refer to the methodology chapter).

- Respective percentage of combinations of reduction measures in 2000 as well as if possible, the percentage of use in 2005, 2010, 2015 and 2020 due to the NMVOC directive or national regulations and applicability according to the definition used in the RAINS model.

Table 6.2.3 : Application rate and applicability factor for each combination of reduction measures

Reduction measure combination for RIC 01			Percentage of use in 2000 %	Percentage of use in 2005 %	Applicability factor 2005 %	Percentage of use in 2010 %	Applicability factor 2010 %	Percentage of use in 2015 %	Applicability factor 2015 %	Percentage of use in 2020 %	Applicability factor 2020 %
RIC	PMC	SMC									
01	00	00			100		100		100		100
Default value proposed for the applicability factor					100		100		100		100
01	01	01			100		100		100		100
Default value proposed for the applicability factor					100		100		100		100
01	02	02			100		100		100		100
Default value proposed for the applicability factor					100		100		100		100
Total			100	100		100		100		100	

The total of percentages of uses of each combination or reduction technique must be 100 since the three combinations are assumed to represent the different situations in this activity.

The percentage of use of the different techniques depends on the Party's regulation. If the directive is applied, combination 00 00 should be forbidden in 2007, except for installations consuming less than 50 t of solvent per year. If these installations constitute a large part of the activity, it can be assumed that a certain percentage of use of measure 00 00 could remain in 2005 or later.

The two measures 01 01 and 02 02 are in compliance with emission limits implemented for existing plants.

The measure 02 02 with an emission factor of 3,5 % of solvent input, allows to meet the emission limit implemented for new plants.

In the tool the absolute terms are directly entered (% for a given combination multiplied by the total activity)

- Unabated emission factor

The unabated emission factor can be modified in a given range.

Table 6.2.4 : Unabated emission factor [kg VOC / t of solvent used]

Default data mean	CI %	User input (mean)	CI %
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300	50		
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The “default data mean” can be modified in a range of $\pm 10\%$.

7 Explanatory notes

7.1 Definition of the reference installation

Three reference installations were initially defined according to data collected in study [6] and used in study [15].

Reference Installation Code	Description
01	<u>Small Installation</u> : annual solvent input (new + recycling) : 400 t/y representative for a solvent consumption < 1 000 t/y
02	<u>Medium Installation</u> : annual solvent input (new + recycling) : 3 000 t/y representative for a range between 1 000 and 5 000 t/y
03	<u>Large Installation</u> : annual solvent input (new + recycling) : 8 000 t/y representative for a solvent consumption > 5 000 t/y

However, available cost data are not sufficiently detailed to be able to take into account such a difference in size. Only one installation is consequently taken into account. This simplification is in agreement with the requirement of simplicity of working documents.

The medium size installation has been considered in this document.

7.2 Derivation of Emission Factors

For combination of primary and secondary measures, the emission factor results directly of their definitions.

Table 7.1.1: Emission factors for primary / secondary measures

			Description	Emission Factor [kg NMVOC/t solvent input]	Abatement efficiency [%]	Emissions avoided t NMVOC/y
RIC	PMC	SMC				
01	00	00	Installation emitting 30 % of the solvents used	300	0	
01	01	01	Installation emitting 8 % of the solvents used	80	73.3	660
01	02	02	Installation emitting 3.5 % of the solvents used	35	88.3	795

7.3 Derivation of Cost Data

7.3.1 Derivation of costs of combination 01 01

The following costs have been obtained during study [6].

Replacement of highly emitting ventilated dryer by less NMVOC emitting dryer:

Technique	Investment – k€	NMVOC reduced t /y	Investment cost €/t NMVOC non emitted
Fluidised bed dryer for product with 35 % of solvent	1140	56	20
Stirred dryer for product with 10 % of solvent	640	16	40
Rotative dryer for product with 10 % of solvent	1000	1	1000

The investment cost ranges from 20 to 40 k€/ t NMVOC abated but can be higher.

Other available data are as follows [6]:

Technique	Investment cost k€/ t NMVOC abated	Operating cost k€/ t NMVOC abated
Production equipment	20 to 40	
Vent collection	1,4 to 12	
Thermal incineration + acid gas absorption	6	0,6
Cryogenic condensation	1,6 to 29	0,3 to 1,6
NMVOC with liquid and solid waste incineration	0,5	0,1

The following costs have been adopted for this study :

Primary Measure 01 and secondary measure 01

Costs for the combination primary measure 01 and secondary measure 01 given in Table 7.2.1 are average costs derived from information given by [3] and collected by CITEPA for study [6].

Average investment costs: 4000 €/ t NMVOC abated

Fixed operation costs: 5 % of the investment

The operating costs are estimated to represent 12 % of investment

Average annual operating costs: 480 €/ t NMVOC abated

7.3.2 Derivation of costs of combination 02 02

Example 1:

The following data have been obtained from an industry expert [11] :

Installation emitting 224 t NMVOC

Working time : 24/24 h/h; 365 days.

Reduction techniques chosen for reducing NMVOC emissions is a mixture of several techniques as follows :

	Investment costs €
Emissions from vents of two multi purpose workshop	
100 vents to be collected – Concentration of NMVOC between 5 to 80 g/nm ³ for a waste gas flow rate of 3000 nm ³ /h. The collection of vent is performed to avoid any risk of contamination.	350 630
Cryogenic pre-condensation	91 470
Scrubber with water	182 940
Final adsorption on activated carbon (with vapour desorption)	548 820
Waste water treatment plant	
Covering of the waste water treatment for an odour treatment and ventilation	169 220
Biofiltration	56 410
Final treatment with activated carbon adsorption	56 410
Other : erection...	198 180
Total	1 654 070

Operating costs are as follows :

	Annual costs €/ year
Electricity	3 050
Vapour	7 620
Nitrogen	3 810
Maintenance	19 820
Solid waste elimination	182 940
Total	217 240

All this equipment abates **190 t of NMVOC**. The efficiency is 85 %.
Investments are on average **8 700 €/ t NMVOC** and operating costs **1 140 €/ t NMVOC**.

Example 2 :

The following costs are presented in study [12] for a plant emitting 203 t NMVOC or 35 % of solvent input (new solvent + recycled solvent : 580 t). About **180 t NMVOC** are abated. Emissions after treatment represent 4 % of solvent input.

Investments costs €2000	Case 1	Case 2	Case 3
Scrubbers (packages)	1000000	830000	600000
Ducting	90000	150000	300000
Valves, utilities, coolers	280000	250000	230000
Electricity work	30000	40000	90000
Civil work	20000	20000	15000
Engineering	60000	60000	60000
Adaptation of vacuum pumps flow	80000	80000	80000
Total investment	1560000	1430000	1375000

Operating costs are estimated to 127 000 €/year.

On average, investment costs are **8100 €t NMVOC** and operating costs **700 €t NMVOC**.

From these examples, the following costs have been adopted for this study :

Primary Measure 02 and secondary measure 02

A combination of different techniques is used and it is necessary:

- To collect emission discharges,
- To clean waste gases from thermal incineration (presence of chlorinated solvents),
- To be in compliance with emission requirements dedicated to waste incinerators when gaseous effluents are led to a waste incinerator.

Average costs from the two examples 1 and 2 presented above are assumed to represent the costs for Primary Measure 02 and secondary measure 02. These are as follows:

Investments: **8400 €/ t NMVOC**
 Fixed operation costs: 5 % of the investment
 Variable operating costs: **850 €/ t NMVOC**

7.4 Summary of costs derived

Table 7.4.1: Emission factors, investments, operating costs, abatement efficiencies, technical lifetimes for relevant combinations of reference installations and primary measures.

Combination Code	Emission Factor for NMVOC [kg/t solvent used]	Abate-ment efficiency [%]	Investment costs [€]	Fixed operating costs [€y]	Operating Costs [€y]	Tech. Life-time [y]	CI [%]
01 00 00	300						
01 01 01	80	73	2 640 000 (4000x660)	132 000	316 800 (480x660)	10	50
01 02 02	35	88	6 678 000 (8400x795)	333 900	675 650 (850x795)	10	50

Table 7.4.2: Marginal costs.

Combination Code	Emission factor for NMVOC [kg/t solvent used]	Emissions abated [t]	Annualised capital costs [€y]	Fixed operating costs [€y]	Operating costs [€y]	Total annual cost [€y]	Marginal annual cost [€t NMVOC]
01 00 00	300						
01 01 01	80	660	325 486 (2640000x0.1233)	132000	316800	774286	1173
01 02 02	35	795	823 331 (6678000x0.1233)	333900	675650	1832881	2305

Total annual cost ranges from 1173 to 2305 €t NMVOC abated. These costs are lower than costs presented in study [10] mainly used in the actual RAINS data. As explained in chapter 2, in the Entec study [10], costs of application of the NMVOC directive in England are compared to a reference base line, which integrates the application of the IPPC directive. The IPPC is assumed to abate emissions from this industry by 43 %. The application of the directive is assumed to abate NMVOC by 56 %. The costs presented in Entec Study are additional marginal costs involved to achieve the additional emission reduction of 13 points of %. This explains the average higher costs of study [10]. Compared

to measure 01 01, marginal costs of measure 02 02 are 7840 €/t NMVOC. The costs involved by the reduction of the first tonnes of NMVOC are lower than costs for reducing the last remaining tonnes of NMVOC.

8 Comments from industry and institutional experts
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No special comments have been received from SICOS: French Association of the Organic Chemistry and Biochemistry Industries.

Minor correction carried out on presentation in April 2005.

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