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Emission of chemicals By air fresheners

Tests on 74 consumer products sold in Europe January 2005



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Summary

The health and comfort of the occupants of a home depends heavily on the quality of the air and the presence of pollutants. These pollutants come from a multitude of sources, such as furniture, paints, varnishes, cleaning products, glues, etc. In this document, we shall be looking at polluting emissions from **air fresheners**. Laboratory tests have been carried out at the initiative of BEUC (Bureau européen des Unions de Consommateurs) by ICRT (International Consumer Research & Testing) on air fresheners sold in Europe. The results of these tests were published in December 2004 in the magazines of the consumers' organisations which are members of ICRT: Altro Consumo (Italy), Compra Maestra (Spain), Pro Teste (Portugal), Test Achats (Belgium) and Que Choisir (France).

This report records the chemicals emitted by air fresheners. The tests, simulating common use of such products by consumers, were carried out on 74 products belonging to different categories (incense, natural products, scented candles, aerosols, liquid diffusers, electric diffusers and gels); for each product, the concentration of Volatile Organic Compounds (VOCs) and aldehydes in the air after the use of the product was measured.

The results are systematically compared against the guideline values provided by the reference organisations such as the Centre International de Recherche contre le Cancer (CIRC), the World Health Organisation (WHO), the American Environmental Protection Agency (US-EPA) and the US Agency for Toxic Substances and Disease Registry (ATSDR). The guideline values in the occupational sector, although cited, are not used as a reference because they are not suited to the general population.

The conclusions demonstrate total VOC emissions which are often very much higher than 200 μ g/m³, the value considered to be significant, substantially increasing the background indoor pollution. The number of different molecules emitted by the 74 products studied under the survey is over 350.

Among the substances emitted we find the presence of **allergens** in the majority of the products tested. Certain products combine up to 3 molecules emitted (cinnamaldehyde, eugenol and coumarin), while others show high concentrations of a single molecule (in particular limonene, which is an allergen in its oxidized form).

A more detailed analysis of the concentrations is proposed for several substances of interest on account of their known toxicity or impact on health: benzene, formaldehyde, terpenes, styrene, diethylphthalate and toluene.

Very high levels of **benzene**, which is a high priority because it is carcinogenic, are found in incense in particular (with a maximum concentration at 221 μ g/m³). These, with incense paper (papier d'Arménie), also produce high emissions of formaldehyde, equally classed as carcinogenic, at concentrations sometimes 6 to 7 times higher than the guideline values: in the case of incense, a maximum value of 69 μ g/m³ and for incense paper (papier d'Arménie), 42 μ g/m³. These levels of **formaldehyde** can be increased by the chemical reactions between primary VOCs, such as terpenes, and other molecules present in the atmosphere, such as ozone, causing the formation of new molecules of formaldehyde.

Worrying levels of exposure can be recorded in the case of molecules such as styrene and diethylphthalate (DEP), by virtue of inadequate data: incense emits more than 60 μ g/m³ of styrene and values in excess of a milligram for DEP.

Only toluene does not seem to be emitted at concentrations which cause concern for the health of the consumer.

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Preface

"There were a number of factors in the decision to test air fresheners. Many scientific studies indicate actual and potential dangers to human health in the use of air fresheners in enclosed spaces - especially for vulnerable groups including pregnant women and children (and pets). Air freshener use is increasing. The sector is virtually unregulated; there are no direct legislative controls or standards for the emissions, primary or secondary, of air fresheners. Labelling requirements are minimal and lists of ingredients not required. There is also increasing concern about the total exposure to chemicals to which consumers may be subject from multiple sources. Furthermore, much of the marketing of air fresheners gives the impression that they 'purify' or improve the quality, in some objective sense, of the air we breathe.

Against this background, the tests presented in the present report were carried out in order to find out more about air fresheners on the market, to inform consumers about the situation through our members' magazines and to contribute to our work on the REACH proposal for a programme of assessment of the tens of thousands of chemicals to which consumers are exposed in everyday life".

Jim Murray Director of Beuc Brussels, 3 January 2005

INTRODUCTION

Air fresheners today belong to a range of common consumer products. Marketed as products associated with comfort and well-being, they allegedly create a relaxing, clean and sophisticated atmosphere in our homes. Moreover, scientific studies show the impact of odours on mood and their beneficial effects. Some advertisements in Europe even focus on the purifying properties of these products.

Yet, whether of natural or synthetic origins, perfumes are chemical substances which are liable to interact with other biological systems than the sense receptors. More and more publications focus on the risks and dangers to human health of the molecules emitted by these perfumes and air fresheners, while indoor air quality is becoming a subject of growing concern. However, no regulatory framework is in place to define the safety of these products, this being left to the goodwill of the manufacturers.

Despite being called fresheners, these products actually work thanks to the fact that they contain perfuming molecules capable of masking 'bad odours' by desensitising our sensory system. What about the effects of these molecules on our health?

With little data available on such products offered to consumers in Europe, Beuc has taken the initiative of entrusting a comparative study into the emissions of 74 air fresheners with several objectives: to gain precise results as to their emissions of chemical substances, to get the public at large to be aware of their impact on air quality and finally to put pressure on the Community health authorities and professionals to ensure that air quality is finally at the heart of their concerns.

This document details the results of measurements of concentrations in the air of the VOCs obtained for every category of products, and identifies the substances of priority interest in the light of the toxicological data available.

1 Objectives of the study

Our objective was to identify and quantify the polluting chemicals emitted by air fresheners. The point is that the scientific literature on the subject mentions multiple pollution, liable to have an adverse effect on the health of the occupants of homes where these products are used. However, no complete information has been available until now on the products marketed in France or in the other countries in Europe, especially in Southern Europe. Yet with the commercial development of these products being very significant these days, consumers' exposure to the chemicals that they emit is on the increase.

The studies conducted in various countries, in the United States or in Europe, on air quality in homes, show worrying levels of pollution linked to the various activities carried out indoors, such as cleaning, decorating, DIY, cooking and so on. Several national or international bodies, plus a number of consumers' associations, have published test results on sources of pollution such as chipboard furniture¹, paints² and cleaning products³. Certain studies even show a link between various illnesses and consumers' exposure to air fresheners⁴.

Our study has therefore focused on measuring the concentrations of pollutants emitted by certain air fresheners, natural or not, such as incense paper (papier d'Arménie), incense, scented candles, and air fresheners in liquid, gel or spray form.

The values measured have then been interpreted in the light of the work of the bodies below:

- the Centre International de Recherche contre le Cancer (CIRC) has evaluated the carcinogenic potential of chemical substances in man; it proposes a classification of the substances depending on the strength of the proof of their carcinogenic nature, but does not propose any Toxicological Reference Values (TRVs).
- the World Health Organisation (WHO) has drawn up a document called 'Guidelines for Air Quality', a reference work in terms of air quality; the WHO suggests guideline values in the air depending on variable lengths of exposure (30 minutes, 8 hours, 24 hours, 7 days, 1 year, etc), and these values may be assimilated to TRVs; for a given dangerous agent, the WHO can offer two types of TRV, for carcinogenic effects and for non-carcinogenic effects.
- the American Environmental Protection Agency (US-EPA) proposes TRVs for chronic carcinogenic and non-carcinogenic effects, but none for short exposures; like the CIRC, it suggests a classification of the carcinogenic power of substances.
- the Agency for Toxic Substances and Disease Registry (ATSDR), an American body, publishes toxicological indexes only for non-carcinogenic effects. The nature of the data used and the arrangements for the derivation of these toxicological indexes make them into a separate set of TRVs. This agency rates TRVs as acute (exposure of less than 15 days), sub-chronic or intermediate (exposure of between 15 and 365 days) and chronic (exposure of over 1 year).

The reference values published by these bodies are systematically used in the framework of our study in order to evaluate the risks associated with exposure to the emissions from the products tested.

The reference values used in occupational regulations are cited for information purposes. However, these do not take account of populations such as babies, children, pregnant women, the elderly or those suffering from asthma or a depressed immune system. Moreover, exposures in the workplace are shorter than in the domestic framework. There has thus been a focus on the Lowest Concentration of Interest (LCI) in order to take account of the particular situation of consumers in their domestic environment.

2 **Definitions of Volatile Organic Compounds (VOCs)**

The term VOC refers to any organic compound with a vapour pressure of 0.01 kPa or higher at a temperature of 293.15 K or with a corresponding volatility under particular conditions of use³.

In France, for example, a ministerial decree dated 11 March 1993 likewise defines VOCs as 'any compound which, with the exception of methane, contains C and H which can be substituted for other atoms (X, O, S, N, P) with the exception of CO and carbonates²⁶.

Volatile organic compounds (or VOCs) cover a multitude of substances which may be of natural or artificial origin (synthetic chemistry). They are always made up of the element carbon and other elements such as hydrogen, halogens, oxygen, sulphur, etc. Their volatility gives them the ability to propagate themselves at greater or smaller distances from their place of emission, thus leading to direct and indirect impacts on man or the environment.

VOCs do not include compounds which do not react photochemically, such as methane, ethane and chlorofluorocarbons (CFCs). Aldehydes (such as formaldehyde) are not VOCs but are ubiquitous pollutants: they have been included in the scope of our study.

2.1 Sources of pollution

According to the EPA, the principal sources of VOCs in indoor air are, aside from the sources of combustion, paints, aerosols such as disinfectants, insecticides and air fresheners⁷.

2.2 Exposure inside buildings

In 1996, the UK's Building Research Establishment⁸ (BRE), conducted a study into VOCs and other polluting substances present in the indoor air in apartments. It seems that the British are exposed to several hundred μg per m³ of VOCs, with significant variations depending on the housing, while the concentration of VOCs outdoors is a few tens of µg per m³:

Average annual concentrations of IVOC in the air $(\mu g/m^2)$					
Compounds	Concentrations (µg/m ³)				
TVOC	n	Average	Standard	Minimum	Maximum
			deviation	4.0	
Bedroom	173	415	323	40	2051
Lounge	173	406	314	51	1799
Outdoors	13	40	30	14	120

 a_1 concentrations of TVOC in the sin $(u, a/m^3)$

TVOC: total volatile organic compounds

In 1991, the teams of Wallace L et al.⁹ at the US-EPA and the California Air Resources Board (CARB) studied the exposure of residents of Los Angeles (California) to 25 different VOCs, in indoor air over an entire year. The results showed up a greater concentration of VOCs in indoor air than in outdoor air, with concentrations (indoors or outdoors) that were higher in winter than in summer; and only in winter, concentrations of VOCs outdoors that were higher at night than during the day.

Similar data were obtained in France by the work conducted by the consumers' association UFC-Oue Choisir in 1999¹⁰ and by the Observatoire de la Qualité de l'Air Intérieur¹¹: the air is 'more polluted indoors than outdoors' by a factor of 1 to 10, or ten times more polluted indoors than outdoors in the case of certain pollutants.

2.3 Impact of VOCs on health

The direct impact of VOCs on the body may stem from their inhalation or their contact with the skin. Consumer products (household products, toiletries and cosmetics, paints and printing inks) individually represent minor sources of emissions of VOCs, but they all make a significant contribution to the total load of VOCs and the problems associated with air quality.

Very recently, the American team of Rumchev K. et al.¹² (2004) showed the existence of a link between domestic exposure to volatile organic compounds and asthma in young children. In this study, the VOCs taken individually represent a significant risk factor for asthma, with the likelihood highest for benzene followed by ethylbenzene and toluene. For every increase in the concentration of toluene and benzene (μ g/m³) of 10 units, the risk of driving up asthma incidence is multiplied by a factor of two and three respectively.

As long ago as 1997, an American animal experiment study, conducted by Anderson laboratories¹³ sought to denounce the toxic effects of emissions from air fresheners. According to this study, exposure of mice for one hour to classic levels of air freshener use triggers pulmonary irritations, respiratory difficulties (asthma type, loss of 50% of respiratory capacity in 10 minutes of exposure) and behavioural anomalies explicable by neurological weakening, with some of the mice dying. Yet the analysis of the VOCs present in the air during the tests shows the presence of substances such as benzene, toluene, limonene, terpene derivatives and other aldehydes (formaldehyde) considered as damaging.

Recently, in Great Britain, the Avon Longitudinal Study of Parents and Children¹⁴ (ALSPAC) also known as 'Children of the 90s', tracked the health and development of 14,000 children throughout their prenatal and postnatal period. This study was the first on the effects of VOCs on very young children. The researchers randomly chose 170 mothers, and monitored them through their pregnancies and the first year of their children's lives. They studied the levels of VOCs in the home over the course of a year and sought to determine which household products were very probably likely to increase the levels of VOCs in the air.

The researchers found that frequent use of air fresheners or aerosols during the mothers' pregnancies was associated inter alia with frequent diarrhoea among the very young children. As to the mothers, headaches and signs of depression were attributed to the regular use of air fresheners. An increase of 32% in babies suffering diarrhoea was recorded in homes where air fresheners (sticks, sprays and aerosols) were used every day, compared to homes where they were used once a week or less. In the case of the mothers, 16% who had used air fresheners suffered from depression, compared to 12.7% who used them only rarely.

Following these publications on emissions of toxic compounds by air fresheners, more and more countries are adopting provisions designed to protect consumers. For example, in the United States, the California Air Resources Board¹⁵ (CARB, 2001) has adopted a regulation to reduce emissions from consumer products, including air fresheners, because of the chemical compounds considered toxic that these products emit. In that context, the Office of Environmental Health Hazard Assessment (OEHHA) in California publishes a list (Proposition 65) of the substances which are regulated because they are recognised to be carcinogenic with no authorised exposure limit; these include benzene.

3 The test

What we did was to simulate the use of air fresheners by consumers, and thus to test them under realistic conditions of use. We staged tests in rooms located in an empty and unoccupied building. The levels of pollutants were measured using air samples from the room, the same air that the consumer would breathe when using the products. (We did not choose to conduct the laboratory tests in emission chambers: because of their small dimensions, emission chambers are not suitable for certain products, in particular aerosols, whose droplets might land on the walls; this method might, however, be suitable for certain air fresheners and its reliability is proven.)

3.1 Implementation of the products

The methodology of application and sampling (i.e. recovery of the pollutants for subsequent analysis) has been designed according to the 5 different categories of products:

-	Candles	sampling 2 hours after ignition
_	Incense	sampling after complete combustion i.e. 1 hour 30
_	Essential oils	sampling after 2 hours
_	Electric diffusers	sampling 2 hours after switching on (position max)
_	Sprays	3 puffs spaced at 15 minutes, sampling 1 minute after the final puff

- Slow-release gels and liquids sampling 2 hours after opening

3.2 Methodology

The tests were conducted in several newly built rooms in the laboratory. Seven rooms were selected in regard to their low background levels (chemicals and odours).

The products were placed on the floor in the middle of the rooms. All the doors were kept closed during testing.

The samples were taken at a distance of 2 metres away from the product. As soon as a test was completed, we opened the doors and made a forced draught through the rooms.

We observed that a delay of at least 3 days was necessary to get the rooms clean and odourless.

3.3 Sampling methods

3 different sampling methods were used for these tests:

- direct sampling in Tedlar® bags (without preconcentration) was performed according to NF-X 43-104 and EN13725. This method is the one used for odour quantification by dynamic olfactometry. For low odour levels, we observed that the bag had to be flushed 3 times prior to the final sampling (in order to avoid adsorption loss).
- VOCs were collected on Tenax TA cartridges (Gerstel, Germany) according to ISO16000-6, at a flow rate of 100ml/min with Gilian Gilair 5 pumps (with low-flow adapter). A sampling duration of 50 minutes (5L/cartridge) was necessary to reach low detection thresholds and avoid MS saturation.
- specific sampling was applied for carbonyl derivatives by using DNPH cartridge 'Xposure Sep-pak' supplied by *Waters* (ref WAT047205). The cartridge content is silica impregnated

with DNPHydrazine and orthophosphoric acid. Carbonyl bonds react with DNPH to produce the stable hydrazone derivatives. For the sampling, an air volume was flushed through the cartridge by using a high precision pump (*Gilian*, Gilair 5). The flow rate was fixed at 1.5 l/min and the sampling time was 20 minutes. This method has been validated according to the NIOSH guidelines for STEL measurements (short-term exposure).

3.4 VOC assay by TDS-GC-MS

- Gas chromatography was used for the analysis of individual traces of VOCs (volatile organic compounds) following EPA method TO17 (see also ISO16000-6). Our GC was coupled to a mass spectrometry (MS) detector and thermal desorption (TDS from *Gerstel*) as injection device.
- Chromatographic separation was achieved on a non-polar Supelco Petrocol (DH 50m x 0.25 mm ID, 0.5µm film thickness). Identification of the chemicals was based on the specific retention time (RT) and the mass spectrometry (by comparison to the data in the literature). External standard curves were used for the semi-quantification in regard to peak height.

3.5 C=O assay by HPLC-UV

Derivatised carbonyls (Hydrazone) were desorbed with 3 ml acetonitrile and the eluate was analysed using an HPLC system equipped with UV detection. The external standard method was used for the quantification. A high-purity (99%) DNPH derivatives solution (available from Supelco ref 47285-U) was used as stock solution to prepare a set of 6 calibration standards in the same range as the expected concentration of the samples (20 µg/m³ to 1000 µg/m³). Calibration fit and quantification are based on the peak area.

3.6 Olfactometric measurements

- Odour intensity measurements were also made: we describe 1 operational method for information purposes because the results of these tests are not discussed in the present document. Accurate and objective odour quantification can be performed according to the new European standard (EN 13725) which describes a method based on dynamic olfactometry at detection threshold with human assessors.
- The odour concentration determinations were performed with the ODILE olfactometer from *Odotech* (Montreal, Quebec). Odourless air supply of ODILE was ensured by a WS15 compressor (oil-free system from *Compair*). Our testing room was neutral (white walls) and odour-free.
- An air-conditioning unit in the room gave the guarantee of an optimised measuring environment (21°C, 50% rh). The bags were placed in a pressurised cylinder in such a way that the odour was pushed into the dilutor (without pumps). The odour was then distributed to the 6 sniffing ports. We worked with 6 panel members who were previously calibrated in regard to their sensitivity to n-butanol. They all had their own board for the vote equipped with 3 outlets (2 for neutral air and 1 for the odour). Presentations of the odour lasted 15 seconds, with a delay of one minute between each presentation. The sequence started at a low concentration of the odour (high dilution factor without perception), then the concentration was doubled for the next presentation until all the panel members were able to completely identify the odour. We ran 3 sequences per sample.
- The odour level was calculated according to the standard, on all the individual results (max. 3 sequences x 6 panellists). The standard recommends a minimum of 10 ITE.

4 Total VOCs

The sum of the concentrations of all the VOCs, or total VOCs, allows an initial interpretation of the level of pollution created by air fresheners. So this is the first criterion which we have taken as an indicator of the level of pollution and which we will discuss in this chapter. We will discuss in the following chapters how to take account of the individual compounds emitted.

4.1 Guideline values

There is no regulatory limit for the maximum concentration in the air in non-industrial premises. However, many national or international bodies have put forward limit values of between 200 and $500 \ \mu g/m^3$.

Value	Category of value	Source
$< 200 \ \mu g/m^{3}$	Comfort level	Commission of the European
		communities; Molhave 1990 ¹⁶
$200 \ \mu g/m^3$	Maximum level	Finnish Society of Indoor Air
		Quality and Climate, 2001 ¹⁷
$200 \ \mu g/m^3$	Maximum level	Public works and Government
		Services – Canada
$< 200 \ \mu g/m^3$	EPA RTP Campus	American Environmental Protection
		Agency (EPA)
$300 \ \mu g/m^3$	Maximum level	Commission of the European
		Communities; Seifert 1990 ¹⁸
$400 \ \mu g/m^3$	Maximum level – provisional	Ministry of Health, Labour and
	guideline	Welfare, Japan, 2001 ¹⁹
$500 \ \mu g/m^3$	Maximum level for one hour	National Health and Medical
		Research Council, Australia, June
		1993 ²⁰
$500 \ \mu g/m^3$	Maximum level	State of Washington ²¹

In light of these data, valid for the <u>total</u> concentration of pollutants in indoor air, or the sum from all sources, we have taken the acceptable limit value of $200 \ \mu g/m^3$, ie the lowest of these values. This value will serve here to evaluate the <u>contribution</u> of the individual product to the total pollution.

The limit values taken in the specifications of the quality labels for emissions from materials are of the same order of magnitude: thus the German quality label GUT (*Gemeinschaft umweltfreundlicher Teppichboden*) applicable to floor coverings provides, three days after it is installed, a total VOC emission limit value of 300 μ g/m³; the values in this set of specifications are compatible with those published in Germany by the Committee for Health-Related Evaluation of Building Products²².

4.2 Secondary pollutants

VOCs emitted by products are known as 'primary pollutants': they can be converted by reaction with photochemical oxidizing agents, under the influence of the sun's rays and heat, into so-called 'secondary' pollutants such as ozone and other photochemical pollutants (aldehydes, ketones, organic acids, etc). These secondary pollutants often have more alarming effects than the primary pollutants. Account will thus need to be taken, in the case of products emitting VOCs at high concentrations, of the risk of the production of secondary pollutants.

The commonest reaction observed involves VOCs and the radical hydroxyl (OH), formed from ozone and water vapour under the influence of the sun's rays. Studies are currently underway to extend our understanding of the speeds of the reactions between certain VOCs and radical hydroxyl, in order to evaluate the life spans of VOCs in the atmosphere, estimated at a few hours to several months.

Recent studies tend to measure and identify, in indoor air, the products generated by the reaction between ozone, radical hydroxyl and $VOCs^{23}$ ²⁴ ²⁵, likewise considered as dangerous. It has been shown that many toxic organic acids and aldehydes are formed in the course of reactions between ozone and organic alkenes (C=C). Radical hydroxyl in a reaction with VOCs is apparently responsible for the formation of 56 to 70% of the formaldehyde²⁶ present in indoor air.

It has been amply demonstrated²⁷ that terpenes (limonene, pinene isoprene), very widespread VOCs in everyday consumer products, in particular air fresheners, react with the ozone present in indoor air. These reactions lead to exposure to new particles, whose structure is little understood, which can trigger respiratory problems²⁸. These reactions likewise occur in the absence of ozone.

A team of American researchers²⁹ has noted that a potentially harmful mini 'smog' can form inside homes as a result of reactions between air fresheners and ozone. These reactions generate the production of formaldehyde. Other teams³⁰ likewise insist on the formation of aldehydes such as formaldehyde by the oxidization of organic compounds, present in air fresheners, containing double bonds (C=C), by ozone.

4.3 Reservoir effect: adsorption

The reservoir effect refers to the ability of materials to adsorb and release chemicals. These materials, carpets and rugs or wall coverings, paper, bedding, etc, thus act like reservoirs for contamination.

Several studies³¹ describe the phenomena of the adsorption of volatile molecules present in indoor air on carpets and rugs in houses. This effect has not been taken into account in the present study, but its existence needs to be borne in mind in the study of indoor pollution phenomena: the point is that it increases the time of exposure of consumers to the substances adsorbed and re-emitted, and under certain circumstances it permits skin contact with the pollutants.

4.4 Results

4.4.1 Incense paper (papier d'Arménie) and other natural products

In the case of incense paper (papier d'Arménie), 13 molecules of different VOCs are found at a total concentration of 78 μ g/m³. In comparison, other air freshening products classed as natural products contain 16 to 24 VOCs at concentrations of 1484 and 1668 μ g/m³, for Aromatic refreshner oil rose and Florame diffuseur d'arôme, respectively, or well beyond the limit of 200 μ g/m³ that we had set.

Papier d'Arménie	78 μg/m ³
Florame diffuseur d'arôme	$1484 \ \mu g/m^3$
Aromatic refreshner oil rose	1668 μg/m ³

4.4.2 Scented candles

Of the 16 scented candles tested, the emissions represent a total number of molecules of VOCs of between 3 and 36 different molecules, for concentrations ranging from 12 to 327 μ g/m³, respectively, for AIRWICK vanille caramel and AIRWICK épices et cannelle. The product emitting the largest number of VOCs is not the one with the greatest concentration, since AMBI

PUR lueur de parfum Odyssey delivers a total concentration in VOCs in the air of over 670 μ g/m³ with 16 different molecules.

Nine products exceed the limit value of total VOCs of 200 μ g/m³.

$12 \ \mu g/m^3$	3 molecules
$22 \mu g/m^3$	5 molecules
$32 \mu g/m^3$	8 molecules
71 μg/m ³	11 molecules
$134 \ \mu g/m^3$	9 molecules
136 μg/m ³	9 molecules
153 μg/m ³	25 molecules
222 μg/m ³	14 molecules
233 μg/m ³	19 molecules
248 μg/m ³	23 molecules
281 μg/m ³	20 molecules
327 μg/m ³	36 molecules
375 μg/m ³	26 molecules
586 μg/m ³	14 molecules
620 μg/m ³	34 molecules
670 μg/m ³	16 molecules
	$12 \ \mu g/m^3 \\ 22 \ \mu g/m^3 \\ 32 \ \mu g/m^3 \\ 71 \ \mu g/m^3 \\ 134 \ \mu g/m^3 \\ 136 \ \mu g/m^3 \\ 136 \ \mu g/m^3 \\ 222 \ \mu g/m^3 \\ 233 \ \mu g/m^3 \\ 233 \ \mu g/m^3 \\ 248 \ \mu g/m^3 \\ 248 \ \mu g/m^3 \\ 327 \ \mu g/m^3 \\ 375 \ \mu g/m^3 \\ 586 \ \mu g/m^3 \\ 620 \ \mu g/m^3 \\ 670 \ \mu g/m^3 \\ 670 \ \mu g/m^3 \\ \end{array}$

4.4.3 Incense

Among the types of incense tested, none emits fewer than 22 molecules of VOCs, at total concentrations among the highest of all the products tested, namely more than 415 up to 1725 $\mu g/m^3$. All the total emissions of VOCs are higher than 200 $\mu g/m^3$.

DRAKE floralies fragrance incense cones	415 $\mu g/m^{3}$	27 molecules
MONOPRIX Bleu d'évasion figuier des cyclades	935 μg/m³	22 molecules
SARATHI Incense sticks camomilla	1232 μg/m ³	23 molecules
USHUAIA fleur de vanille	1725 μg/m ³	29 molecules

4.4.4 Gel fresheners

Of the nine products tested, the total number of molecules measured was between 10 and 46 VOCs, the latter value being the highest total so far. Total VOC concentrations across these minimum and maximum molecules range from over 100 to $1172.4 \,\mu g/m^3$, respectively.

The highest number of molecules does not represent the highest concentration, since the highest concentration is over $1203 \ \mu g/m^3$ for 28 molecules. Three products out of nine emit under 200 $\mu g/m^3$.

AIRWICK Crystal'air fleur de coton	76 μg/m ³	11 molecules
BRISE Victorian Rose	$95 \mu\text{g/m}^3$	21 molecules
IL GIGANTE fiorito	$100 \ \mu g/m^3$	10 molecules
AIRWICK Aroma Mangue et citron vert	$222 \mu g/m^3$	18 molecules
BRISE Lavanda	$481 \ \mu g/m^3$	25 molecules
AMBI PUR Golden Sun New July	743 μg/m ³	21 molecules
IBA Ibana citron vert	997 μg/m ³	14 molecules
AIRWICK Crystal'Air Fleurs de Pêcher	1172 μg/m ³	46 molecules
AIRWICK Crystal'Air lavande gardenia	$1203 \ \mu g/m^3$	28 molecules

4.4.5 Liquid air fresheners

The 10 liquid air fresheners tested released 10 to 35 different VOC molecules, with total concentrations of VOCs from 145 up to more than 1956 μ g/m³ respectively. In this case, the

highest number of molecules represents the highest concentration measured so far with the products tested. Only five of the ten products emit under $200 \ \mu g/m^3$.

AUCHAN limone	$78 \ \mu g/m^3$	12 molecules
ADRITT désodorisant mèche peach	$129 \mu g/m^3$	31 molecules
IBA Tikala thé vert passion	$145 \mu g/m^3$	10 molecules
AIRWICK pomme-chèvrefeuille	$153 \mu g/m^3$	19 molecules
IBA Sanaga pyramide épices marines	$194 \mu g/m^3$	22 molecules
AIRWICK décosphère mangue-citron vert	$480 \ \mu g/m^3$	34 molecules
CAMPAGNIA DEL INDIE new age	590 $\mu g/m^{3}$	15 molecules
AIRWICK decosphère ambiance vanille-orchidée	$629 \ \mu g/m^3$	26 molecules
Ambria vanilla	$1637 \mu g/m^3$	16 molecules
Lampes Berger orange de cannelle	$1956 \mu g/m^3$	35 molecules

4.4.6 Electric diffusers

Among the 16 electric diffusers tested, the total number of VOCs emitted was between 9 and 29 for concentrations of 55 μ g/m³ and over 2284 μ g/m³, respectively.

The highest total concentration of VOCs was over $3163 \ \mu g/m^3$, for a diffuser containing only 11 different molecules. Only two of the 16 products emit under 200 $\mu g/m^3$.

Kill Paff perfume diffusor + recambio	$55 \mu\text{g/m}^3$	9 molecules
AIR WICK Mobil'air vanilla-orchidée	$146 \mu g/m^3$	13 molecules
AIRWICK mandarine thé vert	$252 \mu g/m^3$	15 molecules
AMBIPUR harmony baunilha & lis	$633 \mu g/m^3$	22 molecules
CARREFOUR frutas citricas	1149 $\mu g/m^3$	20 molecules
AMBIPUR April Thé vert	1161 $\mu g/m^3$	27 molecules
BRISE circul'air plaisir d'été	$1268 \ \mu g/m^3$	25 molecules
GREY rillassante vanilla-lily	1595 μg/m ³	13 molecules
AUCHAN marino	$1662 \ \mu g/m^3$	27 molecules
CONTINENTE canela	2185 μ g/m ³	24 molecules
CARREFOUR terre	2284 $\mu g/m^3$	29 molecules
AUCHAN cesta floral	2752 μg/m3	23 molecules
COOP bouquet di orchidée	3163 μ g/m ³	11 molecules

4.4.7 Sprays

The sprays tested in the course of this study are among the air fresheners containing the greatest total number of VOC molecules, but also the greatest concentrations measured in indoor air, after use. Out of the 21 products, we count up to 42 molecules in total, and a maximum concentration in indoor air measured at 7228 μ g/m³. Five of the twenty-one products emit under 200 μ g/m³.

GLADE white freesia & grapefruit	63 $\mu g/m^{3}$	9 molecules
Bonaria (Yplon) Lavande	$68 \mu\text{g/m}^3$	5 molecules
GLADE green apple	$104 \mu g/m^3$	9 molecules
BRISE Orange Jasmin	$187 \mu g/m^3$	30 molecules
BRISE Jasmin et pétales verts	$198 \mu g/m^3$	14 molecules
BRISE Lavanda	$332 \mu g/m^3$	21 molecules
AIRWICK Régén Air	$361 \mu g/m^3$	28 molecules
AIRWICK Lavande	$378 \mu g/m^3$	18 molecules
Lampe Berger Les ambiances vanille	$400 \ \mu g/m^3$	10 molecules
AIRWICK Ambiance mandarine/thé vert	$788 \mu g/m^3$	42 molecules
CARREFOUR Flores silvestres	915 $\mu g/m^3$	13 molecules
AMBI PUR Instant parfum cashmere	920 $\mu g/m^{3}$	36 molecules
CONTINENTE Orquidea Oriente	$1073 \ \mu g/m^3$	27 molecules
AMBIPUR Limon Mandarina	$1198 \mu g/m^3$	32 molecules
GREY Deo'aromatherapy limoni in fiore	$1324 \mu g/m^3$	17 molecules

AUCHAN lavanda	$1622 \ \mu g/m^3$	38 molecules
AIRWICK click spray rosa bouquet	$2018 \mu g/m^3$	20 molecules
Phytaromasol bergamote lemon grass	$2856 \mu g/m^3$	27 molecules
Maison parfum natural spray style colonial	$4596 \mu g/m^3$	27 molecules
BRISE Touch & Fresh brin de muguet	$4655 \mu g/m^3$	33 molecules
Royale Ambree Legrain	$7228 \mu g/m^3$	36 molecules

4.5 Conclusion

Most of the products contribute strongly to the increase in the pollution of indoor air, with a total of VOCs often very much higher than 200 μ g/m³. Given the large number of molecules and the complex character of reactions which may give rise to the formation of secondary pollution, the total levels of VOC concentrations recorded are cause for concern.

Given the wide diversity between the number of different VOC molecules present in each of the products, the concentration emitted of each of these molecules and their more or less toxic character, a more detailed analysis of these results is called for. The point is that even if certain products do not emit a large number of molecules and do not attain a high level of total VOCs emitted, the strong concentration of certain substances may prove very harmful.

In the following sections, we shall look at these molecules and their potentially irritant, allergic or, in the long term, carcinogenic, mutagenic or reprotoxic effects.

5 Allergens

5.1 An allergic mechanism specific to perfuming substances

Among the list of the VOCs emitted by air fresheners we find some potentially allergenic substances. In particular, perfuming substances are present.

The compounds which cause allergies do so via mechanisms which are significantly different from the common allergies to pollens and other protein allergies. The point is that allergies to proteinbased substances occur when the body registers these substances, considers them as harmful and unleashes its defence mechanisms. Yet the majority of VOCs are too small to be detected by the body as allergens. But these substances have the ability to bond with skin proteins. Once modified in this way, these latter are registered by the body, which considers them to be foreign substances, thereby triggering the allergic reaction.

Once a person is sensitised, the only way to avoid the allergy is to avoid exposure. These types of allergies are usually for life, and worsen with each new exposure.

5.2 Respiratory allergies and skin allergies to perfumes

The SCCNFP (Scientific Committee on Cosmetic Products and Non-Food Products) divides allergens into two categories, the substances <u>most frequently</u> reported as contact allergens and those less frequently reported as allergens. This list serves as a basis for the new labelling rules introduced at Community level for cosmetics, and by October 2005, for detergents. Among these allergens, the majority are present in the emissions of the products tested (linalol, geraniol, D-limonene, citral, coumarin, etc).

Allergy specialists in various countries in Europe are finding a significant increase in contact allergies to the fragrance mix, the blend of perfumes commonly used in the skin reaction tests. A study in Germany puts the fragrance as the prime skin allergen, responsible for 15.9 % of the positive reactions among the population studied.³²

Terpenes, in particular limonene, are ever-present in air fresheners: they lead to the formation of oxidization products in indoor air, which are significant allergens in our European context³³. The importance of allergies triggered by the appearance of secondary compounds also needs to be taken into consideration.

Accordingly, the chemicals whose concentration is measured in the emissions from air fresheners are likely to provoke respiratory allergies. But these gaseous compounds can also be absorbed by the skin and lead to skin allergies. Moreover, through phenomena of adsorption on materials and objects in the rooms (walls, rugs, carpets, etc), these primary and/or secondary compounds increase the risks of contact with the skin and the emergence of skin allergies.

5.3 Results

5.3.1 Natural products

- Among the natural products, only Florame diffuseur d'arôme emits limonene: Florame diffuseur d'arôme 911 µg/m³ (D-limonene) 243 µg/m³ (L-limonene)
- Only one natural product contains two allergens rated as 'frequent', <u>citral</u> and <u>coumarin</u>. Aromatic refreshner oil rose $48 \mu g/m^3$ (citral) $20 \mu g/m^3$ (coumarin)

Papier d'Arménie

48 μ g/m³ (citral) 20 μ g/ r 5 μ g/m³ (coumarin)

5.3.2 Scented candles

Among scented candles, the most frequently emitted allergens are cinnamaldehyde (a frequent allergen), linalol and limonene in the majority of products (cf. chapter on terpenes):

 $5 \,\mu\text{g/m}^3$ (linalol)

 $66 \mu g/m^3$ (cinnamaldehyde) $3 \mu g/m^3$ (<u>cinnamaldehyde</u>) $47 \mu g/m^3$ (linalol)

Brise pomme cannelle AIRWICK épices et canelle Ambi pur lueur de parfum Odyssey Natura velas perfumadas vanilla

5.3.3 Incense

Among the types of incense, three out of four emit limonene (cf. chapter on terpenes) and only one emits linalol.

81 μ g/m³ (linalol) DRAKE floralies fragrance incense cones

5.3.4 Gel air fresheners

Six of the nine gel air freshener products tested essentially emit limonene (cf. 8. terpenes), and one emits linalol.

BRISE Lavanda

 $37\mu g/m^3$ (linalol)

5.3.5 Liquid air fresheners

One of the liquid air fresheners essentially emits cinnamaldehyde in high concentrations, and nine of the ten products tested emit limonene (cf. chapter on terpenes). Coumarin, eugenol (allergen plus) and lilial are emitted only at low concentrations and by very few products. 146 μ g/m³ (cinnamaldehyde)

Lampes Berger orange de cannelle

	8 μg/m ³ (<u>coumarin</u>)
	$16 \mu g/m^3 (eugenol)$
AUCHAN limone	$2 \ \mu g/m^3$ (lilial)

5.3.6 Electric diffusers

All of the electric diffuser products tested emit limonene (cf. chapter on terpenes). Linalol is emitted by eight of the thirteen products tested, lilial by two and cinnamaldehyde and benzyl alcohol by just one.

CARREFOUR terre	$25 \ \mu g/m^3$ (linalol)		
AIRWICK mandarine thé vert	46 μ g/m ³ (linalol)		
CARREFOUR frutas citricas	$65 \ \mu g/m^3$ (linalol)		
AUCHAN marino	75 μ g/m ³ (linalol)	$30 \mu\text{g/m}^3$	(lilial)
AUCHAN cesta floral	107 μg/m ³ (linalol)	41 $\mu g/m^{3}$	(lilial)
CONTINENTE canela	111 μg/m ³ (linalol)	$14 \mu g/m^3$	(cinnamaldehyde)
COOP bouquet di orchidée	112 μg/m ³ (linalol)		
Ambipur April Thé vert	146 μg/m ³ (linalol)		
AMBI PUR harmony baunilha & lis	22 µg/m ³ (benzyl alcohol))	

5.3.7 Sprays

Sprays emit 9 allergenic molecules. Limonene is emitted by seventeen out of twenty-one of the products tested (cf. chapter on terpenes). Linalol is emitted by nine of the products tested and lilial by eight. Hydroxycitronellal, geraniol, coumarin, citral, benzyl benzoate and cinnamaldehyde are emitted by only one product.

BRISE Jasmin et pétales verts	$22 \ \mu g/m^3$ (linalol)	$9 \ \mu g/m^3$ (benzyl benzoate)
AIRWICK click spray rosa bouquet	49 μ g/m ³ (linalol)	

AIRWICK Ambiance mandarine/thé vert	$50 \ \mu g/m^3$ (linalol)	$2 \mu g/m^3$ (citral)
AMBIPUR Limon Mandarina	$65 \mu \text{g/m}^3$ (linalol)	$6 \mu g/m^3$ (lilial)
AIRWICK Lavande	$68 \mu \text{g/m}^3$ (linalol)	$3 \mu g/m^3$ (lilial)
CONTINENTE Orquidea Oriente	$77 \mu \text{g/m}^3$ (linalol)	
Maison parfum natural spray style colonial	$78 \mu\text{g/m}^3$ (linalol)	$22 \ \mu g/m^3$ (coumarin)
Royale Ambree Legrain	$750 \mu\text{g/m}^3$ (linalol)	51 μg/m ³ (<u>hydroxycitronellal</u>)
Phytaromasol bergamote lemon grass	$103 \ \mu g/m^3$ (linalol)	$40 \mu g/m^3$ (geraniol)
GLADE white freesia & grapefruit	$9 \mu g/m^3$ (lilial)	$9 \mu\text{g}/\text{m}^3$ (cinnamaldehyde)
AUCHAN lavanda	$45 \ \mu g/m^3$ (lilial)	
AIRWICK Régén Air	$24 \mu g/m^3$ (lilial)	
CARREFOUR Flores silvestres	$115 \mu \text{g/m}^3$ (lilial)	
BRISE Touch & Fresh brin de muguet	$310 \mu\text{g/m}^3$ (lilial)	

5.3.8 Conclusion

In conclusion, the majority of the products tested emit allergens, at concentrations ranging up to 911 μ g/m³ in the case of limonene in one natural product (Florame diffuseur d'arôme). Certain products tested combine allergenic substances emitted such as the Lampe Berger orange de cannelle, in which three different allergenic molecules were measured, with a combined concentration reaching 170 μ g/m³.

These results are corroborated by the study by the Danish Environmental Protection Agency³⁴ seeking to provide a chemical characterisation of air fresheners in order to explain the upward trend in allergies to perfumes among the Danish population using 19 air fresheners*. The concentration of allergens was measured at concentrations up 16% of the total weight of the air freshener, especially in the liquid forms.

* 6 car products (suspended, plug-in or sprays) and 13 home air fresheners (4 electric ones for all around the home, 3 cartridges for vacuum cleaners, 2 sprays for all around the home, and 4 for bathrooms).

The nature of the emissions measured in our study shows that these products are likely to provoke reactions in the form of respiratory and skin allergies: the worrying nature of these phenomena and their increase among the general population in Europe, in particular among children, thus makes air fresheners into a significant source and needs to be taken into account, by professionals and the health authorities, with regard to both the formulation of the products and their labelling.

6 First substance of interest: benzene (CAS 71-43-2)

Benzene (C_6H_6) is an aromatic compound belonging to the family of Volatile Organic Compounds (VOCs) taken into account in many indoor air pollution reduction programmes (see below). It is present in the air primarily in vapour form. Its concentration in the air depends on several factors, notably environmental factors (urban or rural) and climatic factors.

6.1 Sources

The principal sources of the potential production of benzene are oil refineries, the petrochemical industry, petrol, the synthesis of chemical products (for example: phenol, nitrobenzene, chlorobenzene, etc), solvents used in the rubber, leather, footwear and paint industries, and chemical and biological laboratories (usage currently regulated). For the public at large, benzene comes mainly from petrol (car pollution) and cigarette smoke and the air inside homes, furniture polishes, glues, paints, and solvents for domestic use.

6.2 Exposure and impact on health

The average annual concentrations of benzene in the outdoor air registered in Europe by the air quality monitoring associations stand at between $1 \ \mu g/m^3$ in background urban sites and more than $10 \ \mu g/m^3$ in sites close to traffic. As an hourly average, these concentrations can reach $30 \ \mu g/m^3$.

The European Environment Agency has laid down a 5 μ g/m³ limit for benzene in air (Directive 2000/69/EC of the European Parliament and of the Council of 16 November 2000 relating to limit values for benzene and carbon monoxide in ambient air). At present, the European regulation sets the limit concentration value at a concentration of 10 μ g/m³ as an annual average.

For indoor air, the scientific team at the Observatoire de la Qualité de l'Air Intérieur³⁵ (OQAI) in France classes benzene as a priority substance given the health issues recognised by the toxicological databases systematically consulted. The preliminary results obtained thanks to a pilot study by the OQAI conducted between March and July 2001 identified a concentration of benzene in homes and schools of an average of $0.18 \ \mu g/m^3$. A quality objective for indoor air was defined by the Conseil Supérieur d'Hygiène Publique in France at $2 \ \mu g/m^3$. This objective is far exceeded in the case of consumers using certain of the products that we have tested.

In 1996, the UK Building Research Establishment³⁶ (BRE) conducted a survey into the VOCs and other polluting substances present in indoor air in apartments. The measurements obtained on benzene in different rooms are detailed in the table below:

Average annual concentrations of benzene in an (µg/in)					
Compounds	Concentrations (µg/m ³)				
TVOC	n	Average	Standard deviation	Minimum	Maximum
Bedroom	173	8	4	2	32
Lounge	173	8	6	2	46
Outdoors	13	5	1	3	8

Average annual concentrations of benzene in air $(\mu g/m^3)$

6.3 Toxicological values

6.3.1 Occupational regulations

The OSHA (Occupational Safety & Health Administration) in the United States defines the permissible exposure limits (PELs) as follows:

Employers shall ensure that no employee is exposed to a concentration of benzene in air of more than 1 ppm across an average of eight hours' exposure. For the short-term exposure limit (STEL), the concentration of benzene in the air must not exceed 5 ppm for 15 minutes.

At the European Union level, and in the framework of the protection of workers against the risks associated with exposure to carcinogenic or mutagenic agents, regulatory limit values have been established for benzene. The value laid down for the European Union is 1 ppm or 3.25 mg/m³ (across 8 hours - Council Directive 1999/38/EC of 29 April 1999), a value that has applied, for example, to France since June 2003 (art. R.231-58 of the 'Code du Travail') and to Belgium (royal decree of 11 March 2002 relating to the protection of the health and safety of workers against the risks associated with chemical agents in the workplace, annex I).

In Italy, an updating of the limits on occupational exposure (directives 97/42/EC and 1999/38/EC, amending directive 90/394/EEC) on the protection of workers from the risks related to exposure to carcinogens at work and extending it to mutagens, made it possible to reduce the value from 9.75 mg/m³ to 3.25 mg/m³.

The indicative limit value proposed in the United States by the American Conference of Governmental Industrial Hygienists (ACGIH) is 0.5 ppm (1.625 mg/m³), half the European regulatory value. A downward revision of the EU value would be warranted.

A study³⁷ (*Q. LAN. et al.*, 2004)³⁸ recently conducted in China in fact shows that this limit of 1 ppm (3.25 mg/m^3) does not offer sufficient protection, since damage appears at lower levels of exposure; the observations related to 390 Chinese workers exposed (250 workers) or not exposed (140 workers) to quantities of benzene lower than or equal to 1 ppm, for a short part of their lives (3 to 6 years). This research showed haematotoxicity in the workers exposed. In the second group, compared to the control group, we see a significant drop in all types of white cells (global level, granulocytes, lymphocytes, B cells), and platelets. A correlation between the symptoms of the workers and the inhalation of the benzene is shown.

6.3.2 The Observatoire de la Qualité de l'Air Intérieur in France

Under the OQAI ratings, benzene is classified among the 6 'high-priority' substances (*Group A*) because of its acute immunological and chronic carcinogenic effects (leukaemia). This body uses as the Toxicological Reference Value for benzene, in the case of acute exposure (duration of exposure less than 15 days) by inhalation, that of the Agency for Toxic Substances and Disease Registry) (ATSDR), namely 160 μ g/m³.

On the other hand, in the case of chronic exposure (more than one year) by inhalation, the Toxicological Reference Values taken are those of the US-EPA and the WHO, or, respectively, 2.2 – 7.8 x $10^{-6} / \mu g/m^3$ and 4.4 – 7.5 x $10^{-6} / \mu g/m^3$ (expressed as the factor of the linear regression slope between the level of exposure and the frequency of the occurrence of cancer, designating the additional probability, compared to an unexposed individual, of contracting cancer for a subject exposed throughout his life to one unit of measurement of the substance).

6.3.3 The US-EPA

In the same way, the US-EPA is currently proceeding with a rating of the individual chemicals found in indoor environments. It seeks to define the priority substances for which action needs to

be undertaken (Johnston, 2002)³⁹. The first substances recognised as being a priority, in addition to acetaldehyde, formaldehyde, toluene and xylenes, include benzene. In 1999, the EPA⁴⁰ defined an index for the risk of cancer through absorption of benzene by inhalation of 2.2×10^{-6} to 7.8×10^{-6} for exposure to $1 \mu g/m^3$ over a lifetime.

6.3.4 The WHO

The WHO places benzene in the group of substances carcinogenic to man, without any concentration limit.

As to the carcinogenic effects, the concentration of benzene in the air associated with various excess risk values gives the following figures:

- excess risk of 10^{-4} (1 case in 10,000) is 17 µg/m³, for chronic exposure (24 hours a day over a lifetime), a value reached by several of the products we tested.
- excess risk of 10⁻⁵, the concentration value is 1.7 μg/m³
 excess risk of 10⁻⁶, the concentration value is 0.17 μg/m³.

The WHO considers between 4.4 and 7.5 cases of leukaemia per 1 million individuals, exposed to $1 \,\mu\text{g/m}^3$ of benzene over a lifetime. In short, levels of exposure to benzene need to be limited as far as possible. Yet these relatively low values are achieved by several products in our test.

6.3.5 Others

In Denmark, the air quality criteria for emissions from polluting sources are stricter, since the limit for benzene is set at 0.125 μ g/m³ (for chronic effects).

6.4 Results

6.4.1 Incense paper (papier d'Arménie) and other natural products

In the course of our tests, even though it is presented as natural, incense paper (papier d'Arménie) proved to generate pollution: for every incense paper (papier d'Arménie) burned, we found, in a room with initially low background pollution, over $3 \mu g/m^3$ of benzene in the indoor air. The other natural products tested does not emit benzene in detectable quantities.

Aromatic refreshner oil rose	ND μg/m ³
Florame diffuseur d'arôme	ND $\mu g/m^3$
Papier d'Arménie	$3 \mu g/m^3$

6.4.2 Scented candles

Of the 16 candles tested, only one, Brise pétales de fraîcheur, emits almost 3 μ g/m³ after having been burned for 2 hours.

 $3 \mu g/m^3$ BRISE pétales de fraîcheur

Recently, scientist David Krause⁴¹ (1999) in the United States showed emissions of benzene, among other Volatile Organic Compounds, from scented candles. In his report, exposure to benzene for one candle burned per day reached 0.02 to 1.6 μ g/m³, or less than the product above. For a lifetime exposure to these levels, risk indexes of cancer by inhalation are determined: namely 3.3×10^{-8} to 1.2×10^{-7} for the candles with the lowest emissions up to 3.5×10^{-6} to 1.2×10^{-5} for candles with higher emissions. The method for the determination of this factor is based on the relative efficiency of the absorption of benzene via the pulmonary and gastro-intestinal barriers.

6.4.3 Incense

All the products tested in our study are far in excess of the limit of 5 μ g/m³ fixed by the European Environment Agency.

Monoprix Bleu d'évasion figuier des cyclades	19 μg/m ³
DRAKE floralies fragrance incense cones	27 μg/m ³
SARATHI Incense sticks camomilla	137 μg/m ³
USHUAIA fleur de vanille	221 μg/m ³

The types of incense tested released an average of 101 μ g/m³ of benzene after 1 hour 30 minutes of burning (min.: 19 μ g/m³ for the incense Bleu Évasion figuiers des cyclades from Monoprix, max.: 222 μ g/m³ for Ushuaïa fleur de Vanille).

It should be noted that following the publication of the results of the present study, the Belgian Ministry of the Environment and Health has taken the decision to withdraw DRAKE floralies fragrance cones and USHUAIA fleur de vanille sticks from the market, after conducting three tests:

- search for formaldehyde and benzene in the original product (negative test)
- chamber emission test (WHO limits exceeded)
- emission test under real conditions (ditto)

Other scientific studies had already revealed high emissions of benzene by incense.

The most complete is a Danish study⁴² conducted into chemical emissions from incense sticks. Benzene emissions were measured that ran from $11 \ \mu g/m^3$, for a Japanese incense, to $281 \ \mu g/m^3$ for a stick from Hong Kong. These values are maximum concentrations for a stick burned in a room measuring 20 m³ where the air is renewed at a rate of 50% (half the air renewed in one hour). The average for 6 types of incense tested (European, Asian and Indian) is 69 $\mu g/m^3$. These values are at least double those fixed by the European Environment Agency (5 $\mu g/m^3$). According to this study, burning one incense stick is equivalent to 0.5 to 4 cigarettes smoked in a room measuring 20 m³ where the air is renewed 0.5 times/hour.

Let us also cite the works of Löfroth G. et al.⁴³ in 1991 relating to emissions from incense cones. This article is cited as a reference in a report by the United States Environmental Protection Agency (US-EPA) dating from January 2001⁴⁴. Emissions of benzene from incense cones ranging up to 440 μ g/g of incense burned have been measured. These emissions are, of course, in addition to the quantity of benzene already present in the air (1 to 10 μ g/m³). Once again, the reference values are very significantly exceeded.

All these values are all the more alarming for the fact that American⁴⁵ and Chinese⁴⁶ epidemiological studies respectively show the existence of significant correlations between exposure to smoke from incense and cases of leukaemia or symptoms of respiratory diseases in children.

6.4.4 Gel air fresheners

No emission of benzene measured by the gel air fresheners tested.

6.4.5 Liquid air fresheners

In our study, with regard to liquid air fresheners, the AIRWICK décosphère ambiance vanilleorchidée, the lampes Berger orange de cannelle, the ADRITT mèche peach and the pyramides épices marines IBA Sanaga are among the products to emit benzene.

ADRITT désodorisant mèche peach	$4 \mu g/m^3$
Lampes Berger orange de cannelle	$7 \ \mu g/m^3$
IBA Sanaga pyramide épices marines	8 μg/m ³
AIRWICK decosphère ambiance vanille-orchidée	$8 \mu g/m^3$

Out of 10 products tested, 4 produce an average of $7 \mu g/m^3$ after two hours of emission. The maximum concentration emitted is over $8 \mu g/m^3$, by the AIRWICK décosphère, and the minimum concentration almost $4 \mu g/m^3$ by the ADRITT mèche peach air fresheners. The value established by the European Environment Agency of $5 \mu g/m^3$ is therefore exceeded in the majority of cases.

6.4.6 Electric diffusers

Benzene is not emitted by any of the electric diffusers tested.

6.4.7 Sprays

None of the sprays tested leads to the diffusion of benzene in indoor air.

6.5 Conclusion

Given the very high levels of benzene recorded when incense is burned, confirmed by previous studies, these products should be the subject, at the very least, of appropriate labelling. The point is that the comparison with emissions from cigarettes which are at the same level can only encourage the public authorities to take steps. The labelling should, as the consumers' associations have recommended, not only include information relating to the emission of a carcinogenic compound, but also advise that the product be kept away from children, babies and pregnant women, and that rooms should be aired thoroughly after use. The manufacturers of candles, and particularly of incense, should be working, for their part, to reduce as far as possible the emission of benzene, which is an unavoidable combustion sub-product of the use of their products.

As to the other categories, in particular liquid air fresheners, given the permanent character of the emission and the carcinogenic nature of benzene, it is essential for the professionals to conduct checks before air fresheners are placed on the market, in order to ensure that the emissions contain no benzene.

7 Second substance of interest: formaldehyde (CAS 50-00-0)

7.1 Sources

Formaldehyde (CAS 50-00-0) is used in the textile, dye and cosmetics industries as a disinfectant, germicide, insecticide, fungicide and preservative. It is also used in the production of resins as an adhesive and binder in timber and paper products; this makes it omnipresent in our environment at work and at home alike. It is also found in many household products and combustion products. The commonest sources of exposure are combustion gases from boilers and motorised vehicles, cigarette smoke, wallpapers, varnishes and paints, as well as chipboard wood resins and urea-formaldehyde insulation foams, which constitute the principal sources.

7.2 Exposure and impact on health

According to the CIRC, the levels of exposure in ambient air are generally low, but higher levels can be found in indoor air inside homes. According to the ATSDR, most of the exposure to formaldehyde occurs through inhalation or by contact with skin and eyes. Formaldehyde is readily absorbed into the lungs. The odour of formaldehyde is detectable by most individuals at a concentration between 0.06 and 0.22 mg/m^3 .

According to the WHO, the average concentration of formaldehyde in an older home, not containing urea-formaldehyde insulating foams, is less than 0.1 ppm (115 μ g/m³). In contrast, in an interior containing for example recent chipboard, the levels may be above 0.3 ppm (345 μ g/m³). The WHO further states that average exposures to formaldehyde are measured at 0.02 to 2.4 mg/m³, with peaks at between 5 and 18 mg/m³. The WHO provides values for average concentrations in exposures to formaldehyde and the contribution of the various atmospheric environments (below):

Source	Concentration (mg/m ³)
Outdoor air	0.001 - 0.02
Indoor air	
- Classic	0.03 - 0.06
- Mobile home	0.1
- Environment with cigarette smoke	0.05 - 0.35
Smoking (20 cigarettes per day)	60 - 130

Moreover, the study by the OQAI⁴⁷, the average concentration of formaldehyde collected during the pilot campaign in France is 24 μ g/m³, with maximum concentrations being as high as 74.8 μ g/m³. According to the classification method used by the working group, formaldehyde is listed among the 6 substances (8.4%) out of the 70 initially selected, which are 'high priority' (*Group A*) with or without taking account of the index of frequency in indoor air.

In 1991, a Danish team^{48, 49} had already shown emissions of formaldehyde through an entire year, in two identical apartments, one occupied and one not. In the empty apartment, the concentration of formaldehyde is as high as 400 μ g/m³ during the warmer seasons. This concentration is more variable in the occupied apartment, but the authors show that human activity in the apartment introduces new VOCs likely to react with the compounds emitted by the construction materials.

In conditions where incense is used regularly (a ritual of faith in Asia, for example), concentrations of formaldehyde increase considerably. A study⁵⁰ into emissions of formaldehyde in houses or temples in Hong Kong during the use of incense has recently been published.

Exposure to low doses may result in headaches, rhinitis and dyspnoea; higher doses can cause serious irritations of the mucous membranes with burning, and lead to bronchitis, pulmonary oedema or pneumonia. Sensitive individuals may also have attacks of asthma and dermatitis, even at very low doses. Persons sensitised to formaldehyde may suffer headaches and minor irritations of the eyes and respiratory tract at very low concentration levels.

7.3 Toxicological values

In the report on pollution of indoor air inside buildings⁵¹ by the Fondation Universitaire Luxembourgeoise, the toxicity of formaldehyde led many countries to regulate its utilisation. The United States, Canada, Germany and various Scandinavian countries began taking steps in the early 80s to reduce and regulate emissions of formaldehyde deriving notably from urea-formaldehyde foams and chipboards containing urea-formaldehyde resins.

7.3.1 Occupational regulations

The French circular DRT n° 93-18 dated 12 July 1993, modifying and supplementing the modified circular dated 19 July 1992, relating to the permissible values for concentrations of certain dangerous substances in the atmosphere in the workplace, lays down for formaldehyde an AEV of 0.5 ppm (600 μ g/m³). This value is a value calculated for 8 hours of work per day, 5 days per week.

In Spain, the limit for short exposures in the workplace, laid down by the National Institute for Safety and Health at Work⁵², is 0.3 ppm (0.37 mg/m^3) .

In Belgium, the royal decree of 11 March 2002 relating to the protection of the health and safety of workers against the risks associated with chemical agents in the workplace lays down the limit for occupational exposure to formaldehyde at a STEL of 0.38 mg/m^3 (short-term exposure).

In France, the Ministry of Labour has laid down an indicative exposure limit (STEL) for formic aldehyde and an indicative average exposure value (AEV) which are permissible in the air in the workplace. These values correspond respectively to 1 ppm (1.23 mg/m³) and 0.5 ppm (0.61 mg/m³).

In France, there is no general regulation on the limit values for formaldehyde in indoor air in houses. However, the order dated 6 May 1988 (N° 88-883) lays down the maximum level of formaldehyde coming from the injection of urea-formaldehyde foams in buildings used as dwelling places or intended for permanent or semi-permanent human occupation. This order sets the maximum differential value for the concentration of formaldehyde after application of the urea-formaldehyde insulating foam at 0.2 ppm (230 μ g/m³).

7.3.2 The WHO

We took into consideration the limit values fixed by the WHO (World Health Organisation), namely a value of 100 μ g/m³ for exposure of a duration of half an hour⁵³ and 10 μ g/m³ for sensitised persons⁵⁴, still for a duration of half an hour. Considering that formaldehyde is ubiquitous and that there are very few places where the concentration in indoor air is lower than

this limit of 10 μ g/m³, sensitised persons can run up against considerable difficulties in their everyday lives.

7.3.3 The US-EPA

The US-EPA has rated the individual chemicals found in indoor environments. It seeks to define the priority substances for which action needs to be undertaken [Johnston, 2002]. Formaldehyde is rated among the top priority substances. According to the EPA, the Toxicological Reference Value for chronic exposure by inhalation to formaldehyde is $1.3 \ 10^{-5} \ \text{mg/m}^3$ (0.013 $\mu\text{g/m}^3$).

7.3.4 The ATSDR

The American Agency for Toxic Substances and Disease Registry (ATSDR), which depends on the American Ministry of Health, for its part, proposed in 1999 an MRL (minimum risk level) by inhalation of 50 μ g/m³ under acute exposure, 40 μ g/m³ under subchronic exposure and 10 μ g/m³ under chronic exposure.

These values are justified in scientific terms by the Agency that drew them up. This Agency provides the references of the OSHA (Occupational Safety and Health Administration) with regard to the PEL (permissible exposure limit), namely 0.75 ppm or 862.5 μ g/m³ (for exposure for eight hours per day) and the STEL (short-term exposure limit) = 2 ppm or 2300 μ g/m³ (for 15 minutes). The National Institute for Occupational Safety and Health (NIOSH) value for the IDLH (immediately dangerous to life or health) values given as a reference by the ATDSR is set at 20 ppm or 23 mg/m³.

7.3.5 Others

In California, the Ministry of Health recommends reducing concentrations of formaldehyde in homes to less than 0.1 ppm ($120 \ \mu g/m^3$), even in the absence of particular symptoms among the occupants. There is even a recommendation that a concentration of 0.05 ppm ($60 \ \mu g/m^3$) or lower be aimed at.

For the OQIA, in the case of substances presenting both carcinogenic effects and non-carcinogenic effects, the concentration in the air associated with an Excess Unit Risk of cancer of 10^{-6*} is given for formaldehyde at a concentration of 7.7 x $10^{-2} \,\mu g/m^3$.

*or an excess of one additional case of cancer per 1,000,000 subjects exposed to a unit of measurement over an entire lifetime

In Norway, the guideline value for formaldehyde in the domestic environment has been 60 μ g/m³ since 1991. In Sweden it is 130 μ g/m³.

7.4 Results

7.4.1 Incense paper (papier d'Arménie) and other natural products

The concentration of formaldehyde measured is 42 μ g/m³, or 4 times more than the WHO value for 30 minutes and as much as the ATSDR subchronic exposure value (40 μ g/m³). As to the natural products, only the Florame diffuseur d'arôme does not generate any emissions of formaldehyde.

Florame diffuseur d'arôme	ND $\mu g/m^3$
Aromatic refreshner oil rose	$2 \mu g/m^3$
Papier d'Arménie	$42 \ \mu g/m^3$

7.4.2 Scented candles

Fourteen candles out of 16 of the products tested emitted formaldehyde, at an average concentration of 6 μ g/m³. The highest concentration is measured from Spanish and Portuguese candles such as *Natura velas perfumadas vanilla* and *Rogitto Tribo vanilla* respectively with 13 μ g/m³, which is already beyond the limit values laid down by the WHO. Only some Belgian candles, such as *Delhaize anti-tabac* and *Ushuaia fleur de passion*, emit only 1 μ g/m³.

Natura velas perfumadas vanilla	$13 \ \mu g/m^3$
Rogitto Tribo vanilla	$13 \mu g/m^3$
Auchan Lirio dos vales	$10\mu g/m^3$
Glade sun dream	$9 \ \mu g/m^3$
Continente floral	$8 \mu g/m^3$
Brise pomme cannelle	$6 \mu g/m^3$
Brise 3 parfums	$6 \mu g/m^3$
Ambi pur lueur de parfum Odyssey	$4 \mu g/m^3$
Brise pétales de fraîcheur	$3 \mu g/m^3$
GB Vanille	$3 \mu g/m^3$
AIRWICK vanille caramel	$3 \mu g/m^3$
FLAME homeware angelic vanilla	$3 \mu g/m^3$
Delhaize anti-tabac	$1 \mu g/m^3$
Ushuaia fleur de passion	$1 \mu g/m^3$
-	

7.4.3 Incense

The concentrations of formaldehyde in the air are remarkable for the three products tested:

SARATHI Incense sticks camomilla	51 µg/m ³
DRAKE floralies fragrance incense cones	$60 \ \mu g/m^3$
USHUAIA fleur de vanille	69 μg/m ³

A Danish study⁵⁵ published in 2004 has already shown strong emissions of formaldehyde out of 6 types of incense tested. The average concentration of formaldehyde measured in the air is 5.5 mg per hour or 3.6 mg per unit of incense burned. Certain types of incense deliver concentrations of over 11 mg per hour and almost 6 mg per stick or cone burned. These values correspond to average concentrations of formaldehyde of 140 μ g/m³, with a minimum of 49 μ g/m³ for a Chinese incense and a maximum of 210 μ g/m³ for an Indian incense.

7.4.4 Gel air fresheners

No trace of formaldehyde in the air was measured during the use of the 9 products tested.

7.4.5 Liquid air fresheners

Only the lampe Berger, among the 10 products tested, released formaldehyde when burning.

Lampe Berger orange de cannelle $6 \mu g/m^3$

7.4.6 Electric diffusers

Some notable concentrations of formaldehyde were measured from the 13 products tested, with an average of 5 μ g/m³, a minimum of 2 μ g/m³ and a maximum of 13 μ g/m³.

AUCHAN marino	$2 \mu g/m^3$
BRISE circul'air plaisir d'été	$2 \mu g/m^3$
AIRWICK mandarine thé vert	$2 \mu g/m^3$
CARREFOUR frutas citricas	3 μg/m ³
AMBI PUR harmony baunilha & lis	4 μg/m ³
CONTINENTE canela	$4 \ \mu g/m^3$
AIR WICK Mobil'air vanilla-orchidée	5 μg/m ³
COOP bouquet di orchidée	5 μg/m ³
GREY rillassante vanilla-lily	7 μg/m³
Kill Paff perfume diffusor + recambio	7 μg/m ³
Ambipur April Thé vert	7 μg/m ³
AUCHAN cesta floral	9 μg/m ³
CARREFOUR terre	$13 \mu g/m^3$

7.4.7 Sprays

Only one of the 21 products tested emits formaldehyde into the indoor air, at a concentration of 1 $\mu g/m^3$.

AIRWICK Ambiance mandarine/thé vert $1 \mu g/m^3$

7.5 Conclusion

Out of all the air freshening products tested, all the types of incense emitted formaldehyde at levels of concentration 6 to 7 times higher than the limit reference value laid down by the WHO. With these being values that are in addition to the domestic background pollution, the values attained are worrying, even if none of the products tested attained emissions of more than 100 μ g/m³. However, it should be borne in mind that formaldehyde may be present at higher concentrations at certain times of the year, as a secondary pollutant, in the summer when high ozone concentrations are recorded in Europe.

Given the concentration values measured, formaldehyde is likely to pose a risk to the health of consumers, both in the case of incense and incense paper (papier d'Arménie) (high values) and for electric diffusers (low values, but continuous diffusion adding to background noise). In the case of incense, in particular, it would be advisable to test a wider selection of products in order to gain a better estimate of the exposure of consumers to formaldehyde through the use of incense. The recent classification of formaldehyde by the CIRC as carcinogenic to man should encourage the public authorities and manufacturers to reduce emissions of these products, in particular electric diffusers and incense, and to review the labelling.

8 Third subject of interest: terpenes

Terpenes are hydrocarbons, and there are numerous derivatives (alcohols, aldehydes, ketones, acids) of a similar structure, known as terpene derivatives. Among the oxygenated terpene compounds emitted by air fresheners in which we are interested is limonene (a monocyclic compound with two double bonds).

8.1 Sources

Terpenes are present in plants, of which they often form the 'fragrance' constituents (turpentine, camphor, menthol, citronella); they are extracted in the form of essential oils for the perfume industry. Essential oils and wood are therefore the principal sources of terpenes. They are used to perfume domestic products such as household cleaners, for example.

8.2 Exposure and impact on health

In Europe, the average exposure to limonene (the commonest terpene in the air) is estimated at 32-83 μ g/m³ (min–max) in indoor air, 11-23 μ g/m³ in the workplace and 5-9 μ g/m³ outside.

In 2002, an American team⁵⁶ measured the concentration of limonene in indoor air inside a house measuring 160 m² (in Florida, in March): the indoor emissions reached 40.3 μ g/m³ (with emissions of 4.4 mg/h) while outside, the concentration remained below 0.5 μ g/m³.

In 2000, a Danish team⁵⁷ had shown that four terpenes, including limonene emitted by wood, were irritating to the eyes. Irritations were observed at concentrations below 1250 μ g/m³. In its oxidized form, D-limonene is an allergen (see above).

The team of Wolkoff et al. in 2000⁵⁸ had already shown the respiratory problems caused by the formation of secondary molecules following reactions between ozone and certain terpenes.

8.3 Guideline values

No limit value for chronic exposures (inhalation or orally) has been established by the European Union, the European countries or the United States with regard to D- or L- limonene.

In terpenes, limonene (CAS 5989-27-5) is considered by the IARC (International Agency for Research on Cancer) as unclassifiable with regard to its carcinogenicity to man (the carcinogenic effect in man cannot be measured). In the same way, the US-EPA rates limonene in group D: not classed as carcinogenic to man.

The CIRC (Centre International de Recherche sur le Cancer) has shown cases of tumours in the renal tubules of male rats caused by exposure to D-limonene. However, the carcinogenicity mechanisms would not be transposable to man.

The Observatoire pour la Qualité de l'Air Intérieur in France rates limonene in group C ; a priority compound after a close examination of the potential dangers.

8.4 Results

8.4.1 Incense paper (papier d'Arménie) and other natural products

Incense paper (papier d'Arménie) does not emit any significant terpenes (limonene). Among the natural products tested, only the Florame diffuseur d'arômes gives rise to the emission of terpenes of type D-limonene of over 911 μ g/m³ and L-limonene of more than 243 μ g/m³.

Aromatic refreshner oil rose	ND μg /m ³
Papier d'Arménie	ND $\mu g/m^3$

Florame diffuseur d'arôme 9

911 $\mu g/m^3$

243 μ g/m³ (L-limonene)

8.4.2 Scented candles

Fifteen of the sixteen candles tested emit D-limonene, with an average of 5 μ g/m³, a minimum of 1 μ g/m³ and a maximum of 31 μ g/m³. Only 2 candles of the 16 tested emit L-limonene at a maximum of 5 μ g/m³.

AIRWICK vanille caramel	ND $\mu g/m^3$	
SPAAS aromatherapy relaxing lila	$1 \mu g/m^3$	
Rogitto Tribo vanilla	$1 \mu\text{g/m}^3$	
Natura velas perfumadas vanilla	$1 \ \mu g/m^3$	$5 \mu g/m^3$ (L-limonene)
Delhaize anti-tabac	1 μg/m ³	
GB Vanille	$2 \mu g/m^3$	
Continente floral	$2 \mu g/m^3$	
AUCHAN Lirio dos vales	$2 \mu g/m^3$	
BRISE pétales de fraîcheur	$2 \mu g/m^3$	
AIRWICK épices cannelle	$3 \mu g/m^3$	
FLAME homeware angelic vanilla	5 μg/m ³	
USHUAIA fleur de passion	6 μg/m ³	
BRISE pomme cannelle	6 μg/m ³	
GLADE sun dream	8 μg/m ³	
BRISE 3 parfums	9 μg/m ³	
AMBIPUR lueur de parfum Odyssey	31 μg/m ³	$1 \mu g/m^3$ (L-limonene)

8.4.3 Incense

Three of the four types of incense tested create emissions of D-limonene at a concentration of over $19 \,\mu\text{g/m}^3$; the only incense not emitting D-limonene emits L-limonene.

SARATHI Incense sticks camomilla	$1 \ \mu g/m^3$
USHUAIA fleur de vanille	$3 \mu g/m^3$
DRAKE floralies fragrance incense cones	$4 \mu g/m^3$ (L-limonene)
MONOPRIX bleu d'évasion figuiers des cyclades	19 μg / m ³

8.4.4 Gel air fresheners

Six of the nine products tested emit an average of over 266 μ g/m³ of D-limonene with a minimum of 2 μ g/m³ and a maximum of 735 μ g/m³. Three of the gels emitting substantial concentrations of D-limonene also emit appreciable concentrations of L-limonene, up to almost 92 μ g/m³ in the case of the gel emitting the highest concentration of D-limonene.

AIRWICK Crystal'Air lavande gardenia	ND $\mu g/m^3$	
AMBI PUR Golden Sun New July	ND $\mu g/m^3$	
BRISE Lavanda	ND $\mu g/m^3$	
IL GIGANTE fiorito	$2 \mu g/m^3$	
AIRWICK Crystal'air fleur de coton	$4 \mu g/m^3$	
BRISE Victorian Rose	8 μg/m ³	
AIRWICK Aroma Mangue et citron vert	$156 \mu g/m^3$	$5 \ \mu g/m^3$ (L-limonene)
AIRWICK Crystal'Air Fleurs de Pêcher	695 μg/m ³	$32 \mu\text{g/m}^3$ (L-limonene)
IBA Ibana citron vert	735 μg/m ³	92 μ g/ m ³ (L-limonene)

8.4.5 Liquid air fresheners

Nine of the ten air fresheners produce emissions of D-limonene at an average concentration of 36 $\mu g/m^3$ with a minimum of 1 $\mu g/m^3$ and a maximum of 107 $\mu g/m^3$. Some which emit no D-limonene emit the L form, and some gels emit both forms.

Lampes Berger orange de cannelle	ND $\mu g/m^3$	26 μg/m ³ (L-limonene)
IBA Sanaga pyramide épices marines	$1 \mu g/m^3$	
ADRITT désodorisant mèche peach	$1 \mu g/m^3$	
AIRWICK pomme-chèvrefeuille	$2 \mu g/m^3$	
AUCHAN limone	$5 \mu g/m^3$	$0.4 \mu\text{g/m}^3$ (L-limonene)
AIRWICK décosphère mangue-citron vert	$9 \mu\text{g/m}^3$	
Ambria vanilla	$32 \mu g/m^3$	$2 \mu g/m^3$ (L-limonene)
CAMPAGNIA DEL INDIE new age	$77 \ \mu g/m^3$	$2 \mu g/m^3$ (L-limonene)
IBA Tikala thé vert passion	$93 \mu\text{g/m}^3$	
AIRWICK decosphère ambiance vanille-orchidée	$107 \ \mu g/m^3$	

8.4.6 Electric diffusers

All the electric diffusers tested emit D-limonene at an average of 113 μ g/m³ with a minimum of 1 μ g/m³ and a maximum of 498 μ g/m³. Four of the diffusers emitting the highest concentrations of the D form also emit L-limonene.

COOP bouquet di orchidée	$1 \ \mu g/m^3$	
Kill Paff perfume diffusor + recambio	$1 \mu g/m^3$	
AUCHAN cesta floral	$2 \mu g/m^3$	
Ambipur April Thé vert	16 μg/m ³	
AIR WICK Mobil'air vanilla-orchidée	16 μg/m ³	
CARREFOUR terre	42 μg/m ³	
BRISE circul'air plaisir d'été	45 μg/m ³ .	
AUCHAN marino	46 μg/m ³ .	
AIRWICK mandarine thé vert	75 μg/ m ³	$2 \ \mu g/m^3$ (L-limonene)
AMBI PUR harmony baunilha & lis	102 μg/m ³	5 μg/ m ³ (L-limonene)
GREY rillassante vanilla-lily	176 μg/m ³	
CARREFOUR frutas citricas	450 μg/m ³	$26 \ \mu g/m^3$ (L-limonene)
CONTINENTE canela	499 μg/m ³	21 μ g/m ³ (L-limonene)

8.4.7 Sprays

Seventeen of the 21 sprays tested emit D-limonene at an average concentration of 301 μ g/m³ with a minimum of almost 1 μ g/m³ and a maximum of 2003 μ g/m³. Nine of the products also emit the L form.

AIRWICK Régén Air	ND μ g/m ³	
Bonaria (Yplon) Lavande	ND $\mu g/m^3$	
CONTINENTE Orquidea Oriente	ND $\mu g/m^3$	
Lampe Berger Les ambiances vanille	ND $\mu g/m^3$	
GLADE white freesia & grapefruit	$1 \mu g/m^3$	
GLADE green apple	$2 \mu g/m^3$	
BRISE Jasmin et pétales verts	$9 \mu\text{g/m}^3$	
AIRWICK Lavande	$10 \ \mu g/m^3$	
BRISE Lavanda	$10 \mu g/m^3$	
CARREFOUR Flores silvestres	$13 \mu\text{g/m}^3$	
AIRWICK click spray rosa bouquet	$14 \ \mu g/m^3$	$1 \ \mu g/m^3$ (L-limonene)

BRISE Orange Jasmin	$42 \ \mu g/m^{3}$	$1 \ \mu g/m^3$ (L-limonene)
BRISE Touch & Fresh brin de muguet	53 μ g/m ³	
AIRWICK Ambiance mandarine/thé vert	$80 \ \mu g/m^3$	$3 \mu\text{g/m}^3$ (L-limonene)
AMBI PUR Instant parfum cashmere	147 $\mu g/m^3$	5 μg/m ³ (L-limonene)
AUCHAN lavanda	$165 \mu g/m^3$	1 μg/m ³ (L-limonene)
Maison parfum natural spray style colonial	$177 \ \mu g/m^3$	8 μg/m ³ (L-limonene)
AMBIPUR Limon Mandarina	$450 \ \mu g/m^3$	$10 \mu\text{g/m}^3$ (L-limonene)
Phytaromasol bergamote lemon grass	$676 \mu g/m^3$	130 µg/m ³ (L-limonene)
GREY Deo'aromatherapy limoni in fiore	$967 \mu g/m^3$	$42 \mu g/m^3$ (L-limonene)
Royale Ambree Legrain	$2003 \ \mu g/m^3$	

8.5 Conclusion

Out of all the products tested, only one spray exceeds the value, by almost a factor of two, of 1250 μ g/m³ considered as irritating. The documented capacity of terpenes to form formaldehyde as a secondary pollutant must lead to the reduction of their concentration in indoor air. Moreover, the allergenic character of D-limonene in its oxidized form represents a risk to consumers' health. Concentrations of a few hundred microgrammes, frequently found in the emissions from the products tested, appear excessive and need to be reduced and systematic labelling introduced.

9 Fourth molecule of interest: styrene (CAS 100-42-5)

Styrene (or cinnamene, cinnamol, phenylethylene, vinylbenzene) is one of the major compounds in the series of benzene monomers. It is widely used in the production of polymers and copolymers, as well as in the manufacture of polyesters in the composite industry, as a solvent and polymerisation trigger for thermosetting resins, which contain between 30% and 50% styrene. Styrene is an aromatic hydrocarbon synthesised from ethylbenzene by dehydrogenation.

9.1 Sources

From the domestic point of view, cigarette smoke represents the main source of styrene. But domestic products including plastics based on styrene, such as ink, plastic kitchen utensils, food packaging, PVC, etc are also sources of exposure. In addition, styrenes are also used as solvents in certain household products.

Moreover, styrene is transformed in the air by a photo-oxidization reaction with the radicals OH and with ozone. Half-lives of 7.2 hours during a reaction with radicals OH and 9.2 hours during reactions with ozone have been measured. Utilisation combined with the degradation speeds of these two phenomena gives a half-life of 4 hours (EU, 1999). This means that we see a certain persistence of styrene in the air. In addition, the oxidization reactions lead to the formation of secondary compounds recognised as dangerous: the most important degradation products of styrene are formaldehyde and benzaldehyde.

9.2 Exposure and impact on health

According to the WHO, exposures to styrene are an average of $1 \ \mu g/m^3$ in rural areas. In urban areas, the values may reach 20 $\mu g/m^3$, with higher concentrations in new homes built with styrene-based materials.

According to the ATSDR, in the case of a classic population, the average concentrations in indoor air^{59} are between 1 and 9 μ g/m³, concentrations which are attributable to emissions of styrene from construction materials, domestic consumption products and tobacco smoke.

However, the greatest exposures occur in industry. A recent Canadian report⁶⁰ by the Institut de Recherche en Santé et Sécurité du Travail into the effects of exposure to styrene in the workplace reveals symptoms such as irritation of the eyes, the nasal mucous membranes and the throat, in the case of exposures below 1244 μ g/m³.

The CIRC classifies styrene as potentially carcinogenic in man (in general according to the facts considered as credible in man but for which other explanations cannot be ruled out).

Moreover, studies⁶¹ by the INRS in France are currently underway to research the possible effects of styrene on the central nervous system. There will be a transverse study which will compare exposed and non-exposed individuals by reference to their performances in neuro-behavioural and audiological tests.

Exposure to excessive concentrations of styrene vapours (above 200 ppm in air, or 860 mg/m³) leads to depression of the central nervous system which may translate into alertness problems, headaches, dizziness, nausea and fatigue, or even loss of consciousness at very high concentrations. The vapours, as from 100 ppm in air, or 430 mg/m³, are also irritating to the respiratory tract (including the nose and the throat) and to the mucous membranes of the eyes. A toxicological impact on the central nervous system after repeated exposures to styrene has not been clearly established.

9.3 Toxicology

9.3.1 Occupational regulations

The French AEV (Average Exposure Value) permitted in the workplace for a duration of exposure of 8 hours is 50 ppm, or 215 mg/m³ in the case of styrene. For other countries, such as the United States, Germany, Luxembourg or Sweden, this limit value is set at 20 ppm (86 mg/m^3). Harmonisation of the regulations is currently underway.

In Belgium, the royal decree of 11 March 2002 relating to the protection of the health and safety of workers against the risks associated with chemical agents in the workplace lays down the occupational exposure limit value for styrene at an AEV of 216 mg/m³. For short-term exposure, this limit value is set at 432 mg/m³.

In the United States, the OSHA has signed an agreement with the styrene industry to limit exposure to styrene to 50 ppm for exposures of 8 hours per day and 100 ppm (430 mg/m^3) for 15 minutes.

9.3.2 The WHO

The toxicological reference value applied by the WHO in 2000 for exposure by inhalation is a maximum of 70 μ g/m³ for 30 minutes.

9.3.3 The US-EPA

The reference value taken by the US-EPA in 1993 is 1 ppm (4.3 mg/m^3) for exposures by inhalation.

The EPA and the IARC classify styrene among the molecules in group B2, considered as potentially carcinogenic to man. The American agency sets an exposure of 700 ppm (3010 mg/m^3) as the level considered as dangerous for human life and health.

The reference value for chronic non-carcinogenic exposures established by the California Air Pollution Control Officers Association Air Toxics is set at 700 μ g/m³ revised in 1992⁶².

9.3.4 The European Union

Styrene is not classified and is under evaluation.

9.3.5 Others

In Denmark, the air quality criteria for emissions from polluting sources are stricter, since for styrene the limit is set at 0.2 mg/m^3 (for chronic effects).

In Germany, the specifications of the GUT, a label applicable to floor coverings, fixes a limit for emissions of styrene of 5 μ g/m³.

9.4 Results

9.4.1 Incense paper (papier d'Arménie) and other natural products

Incense paper (papier d'Arménie) emits 1.3 μ g/m³ of styrene. One of the two products tested emits over 60 μ g/m³ of styrene.

Florame diffuseur d'arôme	ND $\mu g/m^3$
Papier d'Arménie	$1.3 \ \mu g/m^3$
Aromatic refreshner oil rose	60.5 μg/m ³

9.4.2 Scented candles

Twelve of the 16 products tested emit an average of 43 μ g/m³ of styrene with a minimum of 1 μ g/m³ and a maximum of 112 μ g/m³.

AIRWICK vanille caramel	ND $\mu g/m^3$
GLADE sun dream	ND $\mu g/m^3$
FLAME homeware angelic vanilla	ND $\mu g/m^3$
Rogitto Tribo vanilla	ND $\mu g/m^3$
Natura velas perfumadas vanilla	$1 \mu g/m^3$
BRISE pétales de fraîcheur	$2 \mu g/m^3$
AUCHAN Lirio dos vales	$2 \mu g/m^3$
AMBIPUR lueur de parfum Odyssey	$3 \mu g/m^3$
Continente floral	$5 \mu g/m^3$
BRISE pomme cannelle	$10 \ \mu g/m^3$
AIRWICK épices cannelle	$28 \ \mu g/m^3$
USHUAIA fleur de passion	$35 \mu\text{g/m}^3$
Delhaize anti-tabac	99 $\mu g/m^{3}$
GB Vanille	$104 \ \mu g/m^3$
SPAAS aromatherapy relaxing lila	$109 \ \mu g/m^3$
BRISE 3 parfums	$112 \mu g/m^3$

9.4.3 Incense

All the types of incense tested release emissions of styrene averaging 26 μ g/m³ with a minimum of about 1 μ g/m³ and a maximum of almost 78 μ g/m³.

MONOPRIX bleu d'évasion figuiers des cyclades	1 μg/m ³
USHUAIA fleur de vanille	$10 \ \mu g/m^3$
SARATHI Incense sticks camomilla	$13 \ \mu g/m^3$
DRAKE floralies fragrance incense cones	78 μg/m ³

9.4.4 Gel air fresheners

Six of the nine products tested emit an average of 8 μ g/m³ with a minimum of about 3 μ g/m³ and a maximum of almost 18 μ g/m³.

AIRWICK Aroma Mangue et citron vert	ND $\mu g/m^3$
IL GIGANTE fiorito	ND $\mu g/m^3$
BRISE Lavanda	$3 \ \mu g/m^3$ $5 \ \mu g/m^3$
BRISE Victorian Rose AIRWICK Crystal'Air lavande gardenia	$6 \mu g/m^3$ 8 $\mu g/m^3$
AIRWICK Crystal'Air Fleurs de Pêcher AMBI PUR Golden Sun New July	$18 \ \mu g \ m^3$ 8 \ \ \ \ \ \ \ \ \ \ m^3

9.4.5 Liquid air fresheners

Eight of the ten products tested emit styrene at average concentrations of 8 μ g/m³ with a minimum of about 2 μ g/m³ and a maximum of almost 99 μ g/m³.

Ambria vanilla	ND $\mu g/m^3$
AUCHAN limone	ND $\mu g/m^3$
CAMPAGNIA DEL INDIE new age	2 μg/m ³
IBA Tikala thé vert passion	3 μg/m ³
Lampes Berger orange de cannelle	5 μg/m ³
ADRITT désodorisant mèche peach	8 μg/m ³
AIRWICK decosphère ambiance vanille-orchidée	9 μg/m ³
AIRWICK décosphère mangue-citron vert	19 μg/m ³
AIRWICK pomme-chèvrefeuille	71 μg/m ³
IBA Sanaga pyramide épices marines	98 μg/m ³

9.4.6 Electric diffusers

Seven of the thirteen products tested emit average concentrations of styrene of 10 μ g/m³ up to about 39 μ g/m³ with a minimum of 2 μ g/m³.

AMBI PUR harmony baunilha & lis	ND μg/m ³
AUCHAN cesta floral	ND $\mu g/m^3$
CARREFOUR frutas citricas	ND $\mu g/m^3$
COOP bouquet di orchidée	ND $\mu g/m^3$
GREY rillassante vanilla-lily	ND $\mu g/m^3$
Kill Paff perfume diffusor + recambio	ND $\mu g/m^3$
AIR WICK Mobil'air vanilla-orchidée	$2 \mu g/m^3$
Ambipur April Thé vert	$2 \mu g/m^3$
CARREFOUR terre	$4 \mu g/m^3$
AUCHAN marino	5 μg/m ³
AIRWICK mandarine thé vert	$8 \mu g/m^3$
CONTINENTE canela	$9 \mu g/m^3$
BRISE circul'air plaisir d'été	39 μg/m ³
-	

9.4.7 Sprays

Twelve of the 21 sprays tested emit an average of 27 μ g/m³ of styrene with a minimum of about 2 μ g/m³ and a maximum of almost 185 μ g/m³.

AIRWICK Lavande	ND $\mu g/m^3$
AIRWICK click spray rosa bouquet	ND $\mu g/m^3$
Bonaria (Yplon) Lavande	ND $\mu g/m^3$
CARREFOUR Flores silvestres	ND $\mu g/m^3$
GLADE green apple	ND $\mu g/m^3$
GLADE white freesia & grapefruit	ND $\mu g/m^3$
GREY Deo'aromatherapy limoni in fiore	ND $\mu g/m^3$
Lampe Berger Les ambiances vanille	ND $\mu g/m^3$
Phytaromasol bergamote lemon grass	ND $\mu g/m^3$
AIRWICK Ambiance mandarine/thé vert	$2 \mu g/m^3$
BRISE Touch & Fresh brin de muguet	$4 \mu g/m^3$
AIRWICK Régén Air	4 μg/m ³
Maison parfum natural spray style colonial	5 μg/m ³

$5 \mu g/m^3$
$5 \mu g/m^3$
9 μg/m ³
10 μg/m ³
11 μg/m ³
34 μg/m ³
$48 \ \mu g/m^3$
185 μg/m ³

9.5 Conclusion

Because of its effects on health, its status as a substance not evaluated in Europe and finally its persistence and its reactivity in the air, styrene should be the subject of particular supervision in air-freshening products: the point is that it is found at concentrations in the air that are higher than $70 \ \mu g/m^3$ in some liquid air fresheners, in some aerosols and in incense.

10 Fifth molecule of interest: Diethylphthalate (CAS 84-66-5)

Diethylphthalate (DEP) is used to denature alcohols, in other words to make them unsuited for oral consumption. It belongs to the phthalate family. Its CAS number is 84-66-2. Its liquid form is colourless and almost odour-free.

10.1 Sources

DEP is widely found in consumer products as a plasticiser in packaging, car parts, toys, tools and so on. It is also found as a solvent, a denaturing agent, a filmogenic agent in cosmetics, in particular in perfumes, and in mosquito repellents. It is likewise found in piping for medical material.

10.2 Exposures

DEP is found at different levels in our environment: in our food (because of migration, the most likely sources of exposure to DEP for the general population are the inhalation of contaminated particles and vapours, the ingestion of contaminated water or seafood), or by skin contact through the use of consumer products such as insecticides or aerosol repellents, or perfumes. People may also be exposed to DEP through contact with plasticised products such as inflatable vinyl paddling pools, vinyl seats in furniture or cars, or certain types of clothing such as jackets, rainwear or boots.

10.3 Toxicology

The critical effect of phthalates occurs in terms of their effects on the reproductive system, including effects on fertility and on the development of the foetus and the newborn child. There are differences in effects in terms of both fertility and development, depending on the individual phthalate considered. Phthalates are endocrine disrupters⁶³ but their estrogenic potential is apparently considered to be low compared to that of 17β -estradiol⁶⁴, which is used as a reference. An in-vitro study gave DEP a relative activity* of 10^{-3} compared to 3 x 10-10 for 17β -estradiol. * it takes a concentration of 10-3 to obtain 10% of activity of 10-7 M 17- β -Estradiol.

Compared to the other phthalates, DEP thus has a lower activity and a lower toxicity. However, it does cross the cutaneous barrier and can then spread through the body, although without accumulating in the tissues.

A study involving aquatic animal life indicates that DEP triggers significant changes to certain enzymes in the muscles and the liver. These disturbances to enzyme activity may have long-term effects on organisms exposed continuously to DEP in the aquatic environment⁶⁵.

Another study, using rats, shows that long-term exposure to DEP and ethanol leads to significant adverse effects to lipidic metabolism and toxic damage to the liver. The authors conclude that simultaneous exposure to DEP and ethanol may have a major impact on human health in the event of exposure to these two substances⁶⁶. Low direct mutagenic activity has been demonstrated (on Salmonella typhimurium).

DEP also causes other effects on the body: polyneuritis and impaired balance are reported in the literature.

10.4 Regulation

In occupational medicine, the exposure limit proposed by the American Ministry of Labor to protect workers from effects such as polyneuritis is $5 \text{ mg/m}^{3 67}$; this is the permissible exposure limit (PEL) for 8 hours.

The maximum permitted content of DEP in cosmetics in most countries in Europe is 15%: this recommendation applies to cosmetics liable to be applied on the body and the face ⁶⁸. The EPA has not defined any concentration reference in the case of chronic exposure by inhalation.

10.5 Results

10.5.1 Candles

10.5.2 Incense

Of the four types of incense tested, all emit DEP, two of them at substantially higher levels:

2 1	<i>p</i>
Sarathi Incense sticks camomilla	952 μg/m ³
Ushuaia Incense sticks fleur de vanille	1251 μg/m ³
Drake floralies fragrance incense cones	$19 \mu g/m^3$
Monoprix Bleu d'évasion figuier des cyclades	$2 \mu g/m^3$

10.5.3 Electric diffusers

Of the thirteen products tested, DEP is found in the air with only three: Brise Circul'Air Plaisir d'Eté $7 \mu g/m^3$ Kill Paff perfume diffusor + recambio $4 \mu g/m^3$ Auchan cesta floral $4 \mu g/m^3$

10.5.4 Aerosols

Of the 21 products tested, DEP is emitted into the air by the following thirteen, two at substantially higher levels:

0	
Brise Touch & Fresh brin de muguet	571 μg/m ³
Maison parfum natural spray style colonial	349 μg/m ³
Ambi pur Limon Mandarina	46 μg/m ³
Continente Orquidea Oriente	41 μg/m ³
Royale Ambree Legrain	15 μg/m ³
Auchan lavanda	14 μg/m ³
Carrefour Flores silvestres	13 μg/m ³
Airwick Lavande	8 μg/m ³
Ambi Pur Instant parfum cashmere	6 μg/m ³
Brise Jasmin et pétales verts	6 μg/m ³
Airwick Régén Air	5 μg/m ³
Grey Deo'aromatherapy limoni in fiore	5 μg/m ³
Airwick Ambiance mandarine/thé vert	$4 \mu g/m^3$

10.5.5 Liquid diffusers

Of the ten products tested, only three emit DEP, and only one of these at anything above a negligible level:

00	
Lampes Berger orange de cannelle	67 μg/m ³
Campagnia Del Indie new age	6 µg/m ³
IBA Sanaga pyramide épices marines	$2 \mu g/m^3$

10.5.6 Gels

10.5.7 Natural products

DEP is not emitted by any of the three products tested in this category.

10.6 Conclusion

In light of the data available, DEP is not among the most worrying substances; however, in the absence of a regulatory value and in the expectation of more in-depth studies into its estrogenic potential, it is advisable to limit the contributions to indoor air, notably in the case of incense which emits quantities approaching a milligramme.

11 Sixth molecule of interest: toluene (CAS 108-88-3)

The synonyms for toluene are methylbenzene, phenylmethane and methylbenzol.

11.1 Sources

The principal sources of toluene are adhesives (glues), some paints, varnishes or lacquers and their associated cleaning solvents, nail varnish, art supplies, products for motor vehicles; certain stain removers; pesticides and waxes.

11.2 Exposure and impact on health

According to the WHO, the concentrations of toluene in rural zones are in general lower than $5 \ \mu g/m^3$, whereas in urban areas, the concentrations may be as high as 5 to $150 \ \mu g/m^3$. These concentrations can prove higher in industrial areas.

In terms of indoor air, construction materials, household products and cigarette smoke are sources of toluene. This means that the concentration of toluene in indoor air is often higher than in the air outside. For example, according to a Swedish study⁶⁹ involving 40 volunteers in Göteborg (Sweden), exposures to toluene average 12 μ g/m³ in indoor air, with an average for outdoor air of 2.8 μ g/m³.

Toluene has a low acute toxicity; its prime target is the central nervous system⁷⁰. In that context, a reference concentration for inhalation is set by the US-EPA to protect populations at chronic exposures to toluene, or 0.4 mg/m^3 . Toluene is also an irritant of the eyes, the skin and the respiratory system.

According to the WHO, toluene has potential effects on reproduction, causing hormonal problems in both men and women. Toluene apparently causes spontaneous abortions in women, in the course of exposure at an average concentration of 332 mg/m^3 .

Proof of the carcinogenicity of toluene to man is insufficient: it is rated in group 3 (unclassifiable in terms of its carcinogenic effects in man) by the CIRC.

The European Union has not rated toluene as carcinogenic since 2004. The US-EPA rates toluene as an unclassifiable substance in terms of its carcinogenicity to man.

11.3 Toxicological values

11.3.1 Occupational regulations

In France, the weighted average exposure value (AEV) (8 hours/day) in the air in the workplace is set at 375 mg/m³. The short-term exposure limit (STEL) (15 minutes) is 550 mg/m³. In Germany and Belgium, this same value is set at 190 mg/m³. Europe sets the AEV at 192 mg/m³ and the STEL at 306 mg/m³.

In the United States, the short-term exposure value recommended by the American Conference of Governmental Industrial Hygienists (ACGIH) is set at 191.5 mg/m³ for an exposure of 8 hours per day.

11.3.2 The WHO

According to the WHO, toluene is classed in group IV (probably not carcinogenic to man). The guideline calculated for non-carcinogenic effects is set at a concentration of 3.8 mg/m^3 .

11.3.3 The US-EPA

The US-EPA (source: Integrated Risk Information System) proposes a short-term exposure limit by inhalation for non-carcinogenic effects of 400 μ g/m³. According to the US–EPA, toluene is a pulmonary irritant causing toxic effects for the central nervous system.

11.3.4 The ATSDR

In 2000, the ATSDR established a minimum risk level (MRL) for acute exposure by inhalation, at a concentration of toluene of 3.8 mg/m^3 . For chronic exposure by inhalation, the ATSDR set a minimum risk level of $300 \text{ }\mu\text{g/m}^3$.

11.3.5 Others

In 1991, in Canada, Health Canada established a value of 3.75 mg/m^3 in the case of chronic exposures by inhalation. The National Institute of Public Health and the Environment (RIVM, the Netherlands) proposes a value of $300 \text{ }\mu\text{g/m}^3$ for chronic exposure by inhalation. This value is likewise the limit taken by the US-EPA.

In Denmark⁷¹, the indoor air quality criteria lay down a value of 400 μ g/m³ for chronic effects. Since 2003, the Office of Environmental Health Hazard Assessment (OEHHA) in California has also proposed a value of 300 μ g/m³, still in the context of chronic exposures by inhalation.

11.4 Results

11.4.1 Incense paper (papier d'Arménie) and other natural products

Incense paper (papier d'Arménie) causes no emission of toluene and of the two natural products tested, only one emits a quantity higher than $2.5 \ \mu g/m^3$.

Florame diffuseur d'arôme	ND $\mu g/m^3$
Papier d'Arménie	ND $\mu g/m^3$
Aromatic refreshner oil rose	$3 \ \mu g/m^3$

11.4.2 Scented candles

Half of the candles tested emit concentrations of toluene averaging 6 μ g/m³ with a minimum of over 3 μ g/m³ and a maximum of over 15 μ g/m³.

ND $\mu g/m^3$
ND $\mu g/m^3$

$3 \mu g/m^3$
$4 \mu g/m^3$
4 μg/m ³
5 μg/m ³
5 μg/m ³
5 μg/m ³
$7 \mu g/m^3$
15 μg/m ³

11.4.3 Incense

Three of the four types of incense tested emit concentrations of toluene averaging 19 μ g/m³ with a minimum of under 8 μ g/m³ and a maximum of almost 33 μ g/m³.

DRAKE floralies fragrance incense cones	ND $\mu g/m^3$
MONOPRIX bleu d'évasion figuiers des cyclades	$8 \mu g/m^3$
SARATHI Incense sticks camomilla	15 μg/m ³
USHUAIA fleur de vanille	33 µg/m ³

11.4.4 Gel air fresheners

Seven of the nine products tested emit concentrations of toluene averaging $8 \mu g/m^3$ with a minimum of under $2 \mu g/m^3$ and a maximum of over $18 \mu g/m^3$.

IBA Ibana citron vert	ND $\mu g/m^3$
IL GIGANTE fiorito	ND $\mu g/m^3$
AIRWICK Aroma Mangue et citron vert	$2 \mu g/m^3$
BRISE Lavanda	5 μg/m ³
AIRWICK Crystal'air fleur de coton	6 μg/m ³
BRISE Victorian Rose	$7 \ \mu g/m^3$
AMBI PUR Golden Sun New July	$8 \ \mu g/m^3$
AIRWICK Crystal'Air Fleurs de Pêcher	9 μg/m ³
AIRWICK Crystal'Air lavande gardenia	18 μg/m ³

11.4.5 Liquid air fresheners

Six of the ten air fresheners tested emit concentrations of toluene averaging $6 \,\mu g/m^3$ with a minimum of under $2 \,\mu g/m^3$ and a maximum of over $15 \,\mu g/m^3$.

AIDWICK nomma abduratouilla	$ND u \alpha /m^3$
AIK WICK poinine-chevieleunie	ND μg/m
AUCHAN limone	ND µg/m ³
CAMPAGNIA DEL INDIE new age	ND $\mu g/m^3$
IBA Sanaga pyramide épices marines	ND $\mu g/m^3$
IBA Tikala thé vert passion	$2 \mu g/m^3$
Ambria vanilla	$4 \mu g/m^3$
AIRWICK décosphère mangue-citron vert	$4 \mu g/m^3$
ADRITT désodorisant mèche peach	$4 \mu g/m^3$
Lampes Berger orange de cannelle	$6 \mu g/m^3$
AIRWICK decosphère ambiance vanille-orchidée	$15 \mu g/m^3$

11.4.6 Electric diffusers

Six of the thirteen diffusers tested emit concentrations of toluene averaging 8 μ g/m³ with a minimum of under 4 μ g/m³ and a maximum of over 14 μ g/m³.

AIRWICK mandarine thé vert	ND $\mu g/m^3$
AIR WICK Mobil'air vanilla-orchidée	ND $\mu g/m^3$
AMBI PUR harmony baunilha & lis	ND $\mu g/m^3$
AUCHAN marino	ND $\mu g/m^3$.
BRISE circul'air plaisir d'été	ND $\mu g/m^3$.
GREY rillassante vanilla-lily	ND $\mu g/m^3$
Kill Paff perfume diffusor + recambio	ND $\mu g/m^3$
Ambipur April Thé vert	$4 \mu g/m^3$
COOP bouquet di orchidée	4 μg/m ³
CARREFOUR frutas citricas	6 μg/m ³
CONTINENTE canela	8 μg/m ³
AUCHAN cesta floral	9 μg/m ³
CARREFOUR terre	14 μg/m ³

11.4.7 Sprays

Ten of the twenty one sprays tested emit concentrations of toluene averaging $8 \mu g/m^3$ with a minimum of $2 \mu g/m^3$ and a maximum of over $21 \mu g/m^3$.

AIRWICK click spray rosa bouquet	ND $\mu g/m^3$
AIRWICK Lavande	ND $\mu g/m^3$
AIRWICK Régén Air	ND $\mu g/m^3$
AMBIPUR Limon Mandarina	ND $\mu g/m^3$
BRISE Jasmin et pétales verts	ND $\mu g/m^3$
BRISE Orange Jasmin	ND $\mu g/m^3$
CARREFOUR Flores silvestres	ND $\mu g/m^3$
GLADE white freesia & grapefruit	ND $\mu g/m^3$
GREY Deo'aromatherapy limoni in fiore	ND $\mu g/m^3$
Maison parfum natural spray style colonial	ND $\mu g/m^3$
Royale Ambree Legrain	ND $\mu g/m^3$
AMBI PUR Instant parfum cashmere	$2 \mu g/m^3$
Bonaria (Yplon) Lavande	$2 \mu g/m^3$
BRISE Lavanda	$3 \mu g/m^3$
GLADE green apple	$4 \mu g/m^3$
Phytaromasol bergamote lemon grass	5 μg/m ³
BRISE Touch & Fresh brin de muguet	7 μg/m ³
AUCHAN lavanda	$8 \mu g/m^3$
AIRWICK Ambiance mandarine/thé vert	$8 \mu g/m^3$
CONTINENTE Orquidea Oriente	$18 \mu g/m^{3}$
Lampe Berger Les ambiances vanille	$21 \ \mu g/m^3$

11.5 Conclusion

Under the test conditions used in the course of our study, no product emits concentrations of toluene in excess of the lowest concentration of interest of $300 \,\mu\text{g/m}^3$. The pollution of the air by toluene from these products is thus not a matter for concern.

12 CONCLUSION

This test conducted on 74 air fresheners, incense types, natural products, scented candles, electric diffusers, liquids, gels and aerosols and the associated bibliographical research have allowed us to reach the following important conclusions:

- Total VOC emissions from virtually all the products tested occur at a concentration higher than 200 μg/m³ after 2 hours of use. This represents a substantial increase in indoor air pollution. Notable quantities are emitted by aerosols and electric diffusers, at concentrations of over 7200 μg/m³ and 3100 μg/m³ in the case of Royal Ambree Legrain and Coop bouquet orchidée respectively. In these categories of products tested, 5 aerosols out of 21 and 4 electric diffusers out of 16 exceeded 2000 μg/m³ of emission of VOC.
- Moreover, the literature indicates that the spread of these molecules can be delayed over time because of the phenomenon of adsorption of the VOCs by materials present in homes, which act as a reservoir. Under certain circumstances, these adsorption phenomena may lead to skin allergies as a result of contact between the skin and objects or materials.
- The literature also describes the formation of secondary pollutants, from the primary VOCs, under the action of hear, light and oxidizing agents such as ozone.
- Emissions of allergens from perfumes at often high concentrations are a matter for concern, and need to be looked at in the context of the increase in allergies to perfumes in the various countries in Europe.
- Among the substances emitted, the presence of several carcinogens, benzene and formaldehyde, is a matter for concern.

Benzene is emitted, after 1 hour 30 minutes' burning, at a concentration of over 220 μ g/m³ by the Ushuaia fleur de Vanille stick. It is also emitted by the scented candles and liquid air fresheners in a high concentration. Given the particularly high levels, it seems necessary for work to be undertaken on a wider selection of types of incense.

Formaldehyde poses the same problem as benzene. High concentrations are emitted by incense (> 65 μ g/m³), while the toxicological reference value for chronic exposure by inhalation is set at 10 μ g/m³ by the ATSDR. Three candles tested and one electric diffuser returned a high concentration of formaldehyde.

• As to the emission of molecules of the family of **terpenes**, **styrene** and **DEP**, of which the health impact is amply described in the literature, the concentrations recorded in the air are certainly not negligible: for example, the concentrations of terpene emitted by certain aerosols are twice the doses considered to be irritating in the literature. Moreover, the oxidization reactions of the terpenes can form allergenic compounds (D-limonene in oxidized form) or secondary pollutants such as formaldehyde. As to styrene, certain natural products, scented candles, incense, liquid air fresheners and aerosols exceed the limit value of 70 µg/m³ for 30 minutes of exposure set by the US Environment Agency. This substance is not evaluated in Europe but deserves to have its health impact explored. Another molecule like DEP does not have a guideline value and studies are awaited into its

health impact. For all these non-evaluated substances, it would be wise to limit the concentrations in indoor air.

• Finally, only toluene (unclassifiable in terms of its carcinogenic effects in man) does not seem to pose any problem with high emissions.

Several molecules are thus emitted at concentrations which are alarming because of their recognized carcinogenic or allergenic character to man. It is necessary for the professionals and the health authorities to take account of the results of our trials, in order to improve the formulations and if appropriate to review the labelling of the products. We have not manipulated our interpretations and conclusions: when it comes to benzene, burning a stick of incense corresponds to 0.5 to 4 cigarettes being smoked in a room measuring 20 m³ where half the air is renewed in one hour. This comparison should encourage the public authorities to take steps, such as the introduction of labelling carrying a warning of the emission of a carcinogenic compound, plus a warning that the product should be avoided by children and pregnant women, and that the room should be intensively aired after its use.

At the end of this study and in light of its results, other tests should be conducted:

- testing of a wider range of products, types of incense in particular;
- seeking to gain a better understanding of the real conditions of use by consumers and thus to define their exposure;
- studying the elimination kinetics of certain priority molecules, so as to specify the levels of exposure and to understand the effects of aeration;
- specifying certain unidentified substances, such as ethers of glycol. Certain ethers of glycol in series E are proven reprotoxins and there is a need for precise identification;
- looking at the phenomena of interactions between the molecules and measuring the secondary pollution by varying the conditions of duration, temperature, ventilation, hygrometry, luminosity, etc, in the room.

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