Determination of VOC emissions from French wood products

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Abstract

In order to improve indoor air quality (IAQ), it appears necessary to have analytical protocol for the measurement and health evaluation of building product emissions. In this context, the CTBA (French Wood Technical Centre) and the CSTB (French Building Technical Centre), set up and tested a methodology to characterise emissions of Volatile Organic Compounds (VOCs) from wood building materials. This methodological tool called "Bench of Quality", aims at measuring VOC emissions from building products with laboratory emission test chamber method (European Standard prEN 13419 : 2002) and evaluating the resulting health impact. In all, 15 wood building products were tested : solid wood, wood based panels (particleboards, particleboards with melamine on top, unsanded and painted plywood), glulam, laminate flooring. Results showed that Standard prEN 13419 can readily be applied to solid wood products. A standardised methodology is now available for French wood manufacturers to determine IAQ performances of their products and to confront them with other building products.

Introduction

Today, it is well known that building products may influence indoor air quality (IAQ). Indeed, building products are significant sources of Volatile Organic Compounds (VOCs) emitted in indoor air, which have the potential to affect human health and well-being. In order to improve IAQ, it is generally advised to optimise ventilation and to control sources by reducing pollutant emissions, particularly by the selection of "low emission" building products.

This second point is taking into account the European Directive 89/106/EEC on the provisions and regulations related to construction products [1]. This Directive, also called Construction Products Directive (CPD), demands that products placed on the market shall be designed, built and maintained in such a way that life, health and indoor environment are not damaged (annex 1 of the CPD – essential requirement n°3 "hygiene, health and the environment"). In particular, source control shall be performed by characterisation and quantification of indoor air pollutants, including VOCs and formaldehyde. In order to answer the technical DPC specifications, different testing VOC emission standards have been developed within the CEN committees (European Committee of Standardization).

At the same time, building product manufacturers need reliable tools enabling them to characterise their products and to communicate on their performances in an objective way. This is particularly important for French wood manufacturers to determine IAQ performances of their products and to confront them with other building products. Thus, VOC emission data from building materials based on solid wood and its derivatives (adhesives, coatings, preservatives) are needed.

However, there is a real lack of information on nature and concentration level of the pollutants emitted by wood products. Several data on formaldehyde emission from wood-based panels were available following regulations in certain countries (Germany). But VOC emission results are more difficult to obtain and even when data are available, they were not easily comparable due to lack of uniformity in the different National testing methods and the type of tested material [2].

In this context, French wood manufacturers need a standardised procedure to compare wood building products and to assess their health impact on indoor air quality. Therefore, they encouraged the CTBA to set up a standardised protocol to evaluate VOC emissions from building wood products. Thus, the CTBA and the CSTB have proposed and tested a methodological tool, called "Bench of Quality", to characterise pollutant emissions from wood building materials.

This study, supported by Environmental French Agency (ADEME) and French Building Federation, was based on a chemical analysis of pollutant emissions (COV and aldehydes) from building products according to the horizontal European Standard prEN 13419 and a toxicological evaluation of volatile pollutants according to an European classification scheme developed under the European Collaborative Action (ECA) “Indoor Air Quality and its impact on Man” [3, 4, 5, 6].

Standard prEN 13419 was developped by the European group of standardization CEN/TC264/WG7 ("Quality of the interior air"). This committee worked out an experimental standard in three parts to measure the emission of all volatile chemical substances from all building products [3, 4, 5]. These European Standards use the principle of the emission chamber test. They describe the procedure for sampling, transport, storage and preparation of building products (prEN 13419-3), and for conditionning test specimen under controlled conditions in test emission chamber (prEN 13419-1) or in test emission cell (prEN 13419-2). Test emission chambers allow the transposition of VOC test chamber concentration to VOC exposition concentration (real room conditions) by acting on several parameters (temperature, relative humidity, air exchange rate, loading factor of the product,…).

Standard prEN 13419 applications are specified for all building products, except wood-based panels. For the specific determination of formaldehyde release from wood-based panels, standard ENV 717-1, worked out by the CEN/TC112/WG5 ("wood-based panels"), should be used [7]. Today, it is defined as the reference method for the classification (EC mark) of wood-based panels according to the standard EN 13986 [8].

European standard prENV 13419 only presents the conditioning in test emission chamber of building products. Other Standards prepared by ISO/TC146/SC6 (“Air quality – indoor air) were developped for the measurement of pollutants in indoor air or at the output of test chambers.

About VOCs, the ISO produced standard ISO/DIS 16000-6 [9]. The method is based on active sampling of the chamber air through a solid adsorbent (Tenax TA), and analysis by thermal desorption and gas chromatography. VOC identification is carried out by mass spectrometry (MSD) and quantification by flame ionization detector (FID). After final vote, ISO 16000-6 will be taken over Part 4 of EN 13419.

Standard ISO/DIS 16000-6 can’t be used for formaldehyde analysis. Standard ISO 16000-3 was retained to measure formaldehyde and others carbonyl compounds in air [10]. Air is sampled through silica gel cartridges impregnated by 2,4-dinitrophenylhydrazine (DNPH) and analysed by liquid chromatography with UV detection (HPLC/UV).

Experimental

The principle retained for the "Bench of Quality" is to determine the emission of pollutant emissions (COV and aldehydes) from solid wood building products by test chamber

according to Standard prEN 13419-1 [3,11]. The test was carried out in two sorts of emission test chambers : a one cubic metre stainless steel chamber and a 51 litre glass chamber called CLIMPAQ (Figure 1).



Figure 1 : 51 litre glass chamber used for the test

The principle is to determine emission rate of VOCs emitted from products at controlled and constant environmental parameters (temperature, relative humidity, air exchange rate). Important parameters for controlling environmental and ventilation conditions are presented in Table 1.

Parameter	Analytical condition
Temperature	23 ± 2 °C
Relative Humidity	50 ± 5 %
Air flow velocity	0,1 - 0,3 m/s
Area-specific air flow rate	1,25 $m^3.m^{-2}.h^{-1}$
Air exchange rate	0,5 h^{-1}
Scenario	Floor and ceilings
Test duration	28 jours

Table 1: analytical parameters in emission test chamber

The area-specific air flow rate is the ratio between the air exchange rate in test chamber and the product loading factor (ratio between product surface area and chamber volume).

Analytical conditions simulate realistic indoor situation. Only real emissive face of building product in indoor air will be exposed in test chamber. Others faces will be sealed. A “Floor and ceilings” scenario has been applied to reflect exposure under residential practical conditions.

Wood building products were sampled ready to use and packaged just before merchandising. They were stored in wrapped plastic bags just before test. The beginning of the experiment is defined as the time at which the product is unwrapped and placed into the test chamber. Exemple of test specimens is presented in Figure 2.



Figure 2: test specimens prepared from particleboard with melamine on top

After the beginning of the test, emission rates from wood product were measured after 1, 3 and 28 days (VOCs according to ISO/DIS 16000-6 and aldehydes according to ISO 16000-3). Concentration in the air chamber of individual and total VOCs (TVOC) were determined. Identification is conducted on all individual compounds with a concentration greater than 2 $\mu g.m^{-3}$ and a retention time in apolar capillary column between n-hexane (C_6) and n-hexadecane (C_{16}) included. The TVOC parameter is the sum of these individual compounds after quantification in Toluene equivalent.

In this study, different wood building products were tested according to the "Bench of Quality": solid wood (pine, oak), wood based panels (E1 and E2 particleboards, particleboards with melamine on top, unsanded and painted plywood), glulam, laminate flooring with MDF substrate. Last test was carried out on composite product (multi-layer parquet floor glued on concrete substrate) to study influence of gluing. The aim was to compare main families of uncoated and coated wood products in order to validate the use of standard prEN 13419 to wood building products. Main results were presented hereafter.

Results

Firstly, solid wood emission was studied. Softwood (uncoated and coated maritime pine lamelling, uncoated Scots pine) and hardwood (uncoated and coated oak parquet floor) were tested. Maritime pine lamelling was coated by nitrocellulosic varnish and oak parquet floor by UV lacquer. TVOC emission is presented in Figure 3.

TVOC emissions decrease during the test. Whatever the test period, VOC and aldehyde emissions are significatively higher in coated than those in uncoated oak. After 28 days in test chamber, main compounds emitted from uncoated oak parquet floor are acetic acid, acetone, formaldehyde and acetaldehyde. The coated oak parquet floor is also mainly characterized by acetic acid emission. The UV lacquer of the parquet floor showed specific emissions of ketones (cyclohexanone, benzophenone) and benzaldehyde but its emission is low.

There's no real difference between uncoated and coated pine lamelling. Pine TVOC emission is widely larger than oak because of natural compounds emission like terpenes (alpha-pinene, beta-pinene, myrcene, limonene) and aldehydes (hexaldehyde). Many other natural compounds were not identified. However, specific pollutants of nitrocellulosic varnish were clearly identified, especially after 1 and 3 days (2-butanol, toluene, isomers of xylene).

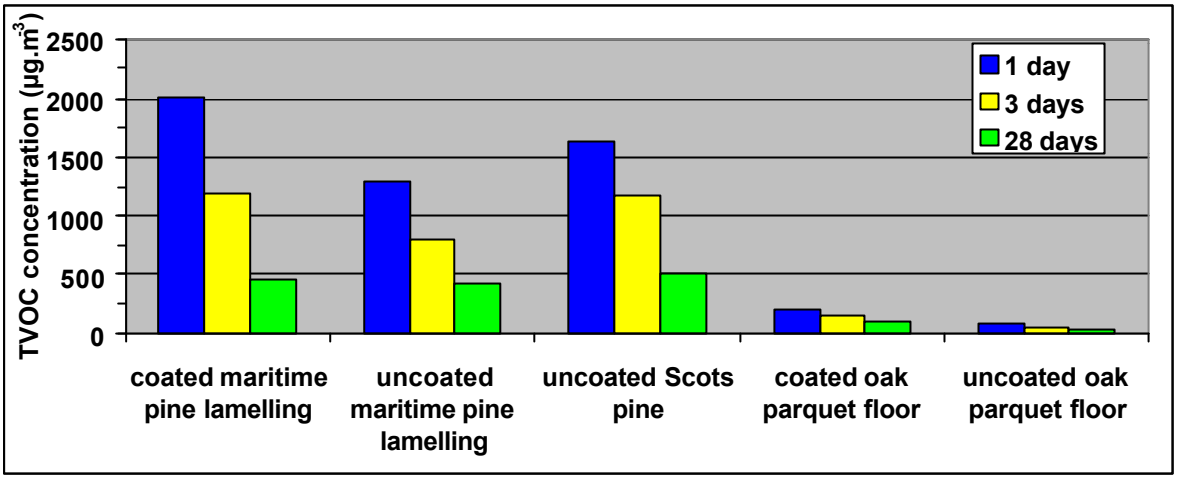


Figure 3 : TVOC emission from oak parquet floor, pine lamelling and Socts pine after 1, 3 and 28 days

Wood-base panels were also tested: plywood, E1 / E2 particleboards and MDF boards. To study influence of coating, unsanded plywood was compared to painted plywood (acrylic paint). Okoume panels were issued from the same batch and were glued with phenolic adhesive (3 veneer, 8 mm thickness). TVOC emissions are shown in Figure 4.

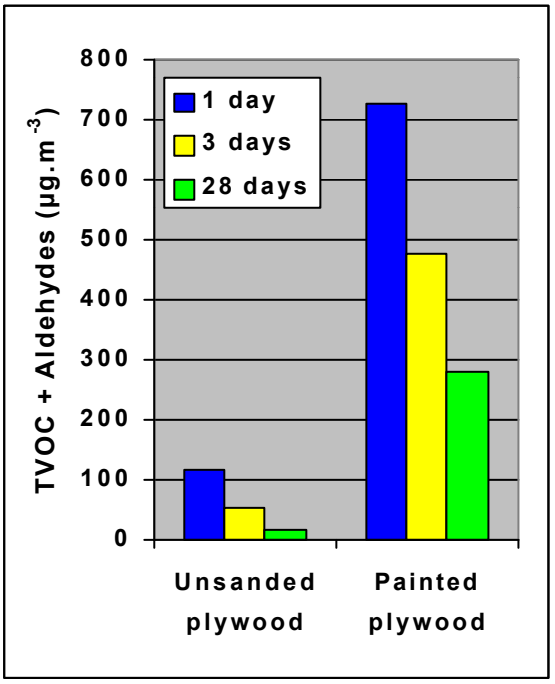


Figure 4 : pollutant emission (TVOC + Total aldehydes) from unsanded and painted plywood after 1, 3 and 28 days

Tested plywoods showed low TVOC and aldehyde levels (14 µg.m^{-3} for unsanded plywood and 280 µg.m^{-3} for painted plywood after 28 days), paint applied to the panel emitting additional compounds. After 28 days, unsanded plywood emits mainly acetic acid, furfural and formaldehyde. Formaldehyde emission is below 5 µg.m^{-3}. These results confirmed the weak formaldehyde emission of this type of wood-based panels (phenolic glue). Acetic acid and furfural were also identified in painted plywood but emission of specific compounds of the waterborne paintlike glycol ethers (2-butoxyethanol, 2-phenoxyethanol and and 1,2-propanediol) were also identified.

E1 and E2 class particleboards glued with UF adhesive were tested (16 mm thickness). The same E1 class wood-based panel was coated with melamine on top (white and oak pattern). TVOC emissions after 28 days are shown in Figure 5.

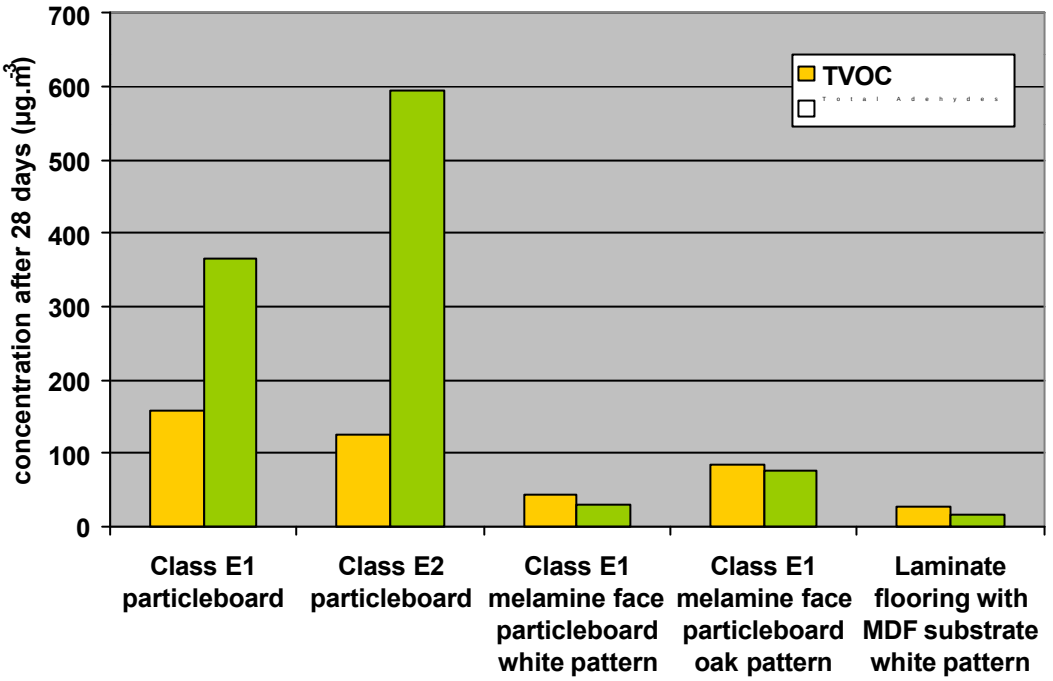


Figure 5 : TVOC and Total aldehyde emissions from particleboards and laminate flooring after 28 days

After 28 days, aldehyde emission showed lower results for E1 class panel, in comparison with E2 class panel. On the other hand, TVOC emission remains in the same order of magnitude. This confirms the influence of other compounds on the pollutant emission, when aldehydes are especially related to the UF glue. For particleboards, formaldehyde is the principal pollutant because of the UF glue composition. But other aldehydes like hexaldehyde, valeraldehyde and acetaldehyde were identified. VOCs like acetone, acetic acid, alpha-pinene and 1-pentanol were also identified. These compounds were already identified in solid wood (pine). They are thus probably emitted from natural softwood.

TVOC and Total aldehyde emissions of E1 class panels with melamine on top (white and oak pattern) are very low, in comparison with E1 class particleboard. Same compounds are emitted but at significantly lower concentrations. These results show the “barrier effect” of melamine on top.

After 28 days, formaldehyde concentration of E1 class particleboard was higher (210 µg.m^{-3}) than limit E1 specified by Standard ISO 13986 (124 µg.m^{-3}). Probably, stationary state described by ENV 717-1 is not reached after 28 days of test. However, results between prEN

13419 and ENV 717-1 are not comparable because of a different loading factor and a different relative humidity in test chamber (50 % HR in prEN 13419 instead of 45 % HR in ENV 717-1).

Another test was carried out on laminate flooring with MDF substrate (8 mm tickness) and white pattern (Figure 5). Among all tested building products, emissions of laminated flooring were the lower. Only the decorative phase is in contact with the air of the test emission chamber. The very weak TVOC emission confirmed the “barrier effect” of melamine facing. Indeed, formaldehyde concentration, component of UF glues, is lower than 5 $\mu g.m^{-3}$. In addition, coating does not involve specific emissions of volatile compounds.

All previous tests were carried out on solid wood products. A final test was realised to study a composite wood product. For that, two sorts of test were realised: a preliminar test on non-glued multi-layer parquet floor (oak face, softwood back, UV coating) in order to characterise emissions of the exposed side (in contact with indoor air), and a secondary test on multi-layer parquet floor glued on a concrete substrate (vinyl glue in petroleum solvent) in order to simulate real conditions of use of the parquet floor and to evaluate influence of gluing (Figure 6).

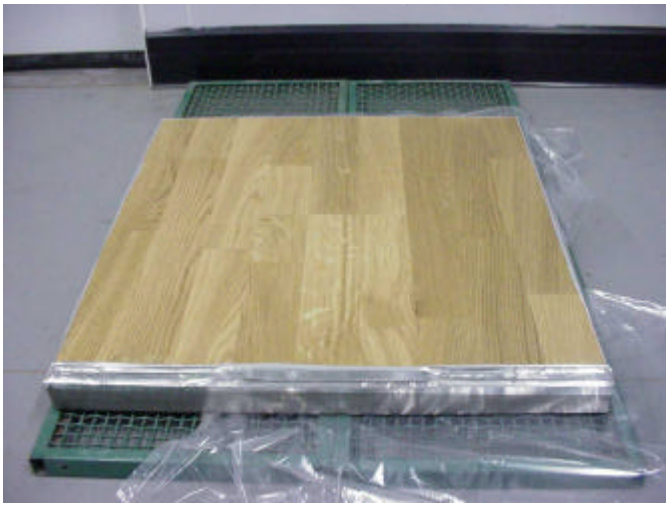


Figure 6 : multi-layer parquet floor glued on a concrete substrate

TVOC concentrations after 1, 3 and 28 days are presented in Figure 7. Multi-layer parquet floor emission is low in comparison with glued parquet floor, especially after 1 and 3 days of test.

Non glued multi-layer parquet floor emission showed results similar to those of pine lamelling. Terpenes are major compounds (alpha-pinene, limonene, 3-carene) and other compounds are also emitted (acetic acid, formaldehyde). Concentration levels of these compounds are relatively stable over the duration of test. The multi-layer parquet floor presents an oak facing. Some identified compounds are thus identical to those found during the test on the oak parquet floor. On the other hand, terpenes are generally specific to softwoods. Emission of these compounds must be related to the presence of the softwood back. It seems that UV coating does not create a barrier effect.

Emissions of multi-layer parquet floor glued on a concrete substrate are mainly characterised by solvents of vinyl glue (toluene, ethyl acetate, acetone, ethanol, alkanes) which are emitted by joints. After 3 days, vinyl glue emissions are still very important (TVOC concentrations of 4676 $\mu g.m^{-3}$ for the glued parquet floor against 364 $\mu g.m^{-3}$ for the non glued parquet floor).

Glue emission lasted after 28 days of test. Indeed, TVOC concentrations are always high (514 µg.m^{-3} for the glued parquet floor against 287 µg.m^{-3} for the non glued parquet floor). Probably, glue reticulation is not yet finished but solvent emission are definitely weaker (ethanol, acetone, ethyl acetate). Alkanes are not detected any more. Only toluene remains with a significant level of concentration.

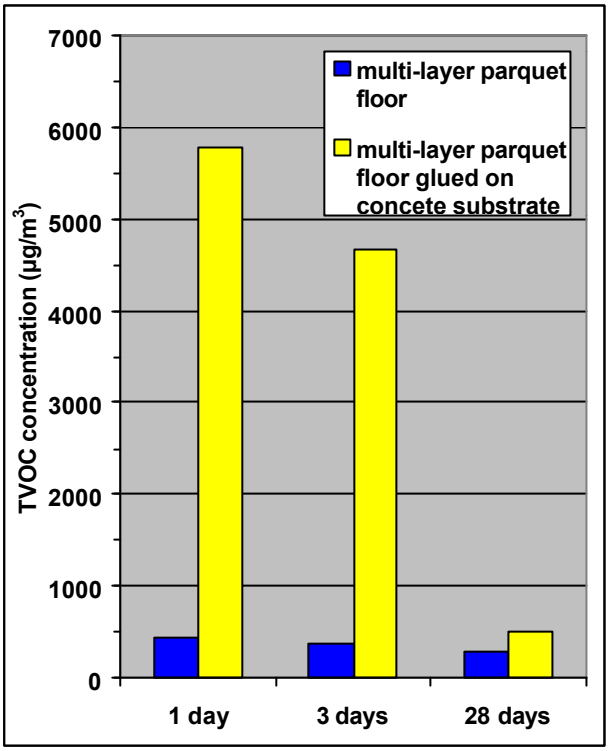


Figure 7 : TVOC emission from multi-layer parquet floor after 1, 3 and 28 days

Conclusion

The aim of this study was to test the principle of the " Bench of Quality " for wood building products. Therefore, test pieces were prepared and conditioned in test emission chamber according to horizontal standard prEN 13419. Air sampling and analyses were carried out using ISO/DIS 16000-6 for VOCs and ISO 16000-3 for aldehydes.

In all, 15 different products were tested : solid woods, glulam, laminate flooring with MDF substrate and wood based panels (E1/E2 particleboards, particleboard with melamine on top, unsanded and painted plywood).

Results confirmed that the European standard prENV 13419 can be readily applied to solid wood products from the point of view of their VOC and aldehyde emissions. Indeed, specificity of great families of wood products was identified: influence of natural compounds emitted by solid wood (terpenes, acetic acid, hexaldehyde), influence of coatings (specific emission of solvents, barrier effect), glue emission (formaldehyde, acetaldehyde).

However, this study showed that evaluation of building product emissions remains delicate because of the diversity of parameters to check. It is necessary to further the knowledge on emission data from wood products according to the "Bench of Quality". On the other hand, influence of wood product sampling should be discussed, this material presenting high natural heterogeneity. Finally, a preliminary step is to determine the reproducibility of prEN 13419 (Round Robin Test between National or European laboratories).

Another problem is specific to wood-based panels. Indeed, two Standards (ENV 717-1, prEN 13419) using test chamber principle should be available to determine pollutant emission from wood-based panels but they specify different analytical parameters (relative humidity, air exchange rate, loading factor). In order to realise one unique test to measure VOC and formaldehyde emission from wood-based panels, an harmonized standard must be discussed.

At present, the "Bench of Quality" is always in validation step. Its first interest is to characterise pollutant emission (VOC and aldehydes) according to the same methodology. Results obtained from 15 tested wood products are encouraging and this methodological tool should be proposed to French manufacturers to test and to compare their products with other building products and to select “low emission” products for specific use.

From now on, an additional assessment taking into consideration health criteria must be studied. In 1997, a detailed procedure for the assessment of chemical emissions from solid flooring materials was presented by the European Collaborative Action (ECA) “Indoor Air Quality and its impact on Man” [6]. By now, the German committee for health-related evaluation of buiding products (AgBB) establishs the fundamentals for the minimum requirement of VOC emission from building product for health protection. All these new health schemes must initiate manufacturers to declare performances of their products as regards VOC emission and to produce “low-emission” products.

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