Essential requirements on construction products and current European standardization of emission test methods

Hans Gustafsson

SP Technical Research Institute of Sweden, P. O. Box 857, SE-501 15 Borås, Sweden

INTRODUCTION

Since materials in building structures, and especially those applied to surfaces in large quantities, are permanently exposed to the indoor air, it is crucial to develop an understanding to what extent they contribute to indoor air pollution. Numerous laboratory investigations have been reported concerning releases of chemical substances from interior building materials. However, several of these substances are seldom associated with complaints or building related illness. It could therefore be worthwhile to give an overview of technically well-documented case studies where chemical substances have been positively identified in the indoor air in various types of buildings. This updated overview is limited to surface materials as primary emission sources and based on an international review of case studies (1) in which most of the cases not have been published earlier. The review is covering various materials containing more or less volatile functional chemicals or their degradation products.

The overview does not include radon or formaldehyde, as the sources of these substances can be regarded as well known. Since building materials can be considered as key emission sources, the current development of standardised emission test methods within the frame of the Construction Products Directive (2) are described.

INDOOR EXPOSURES TO VOCS AND GASEOUS CONTAMINANTS FROM BUILDING MATERIALS

The highest personal exposures to VOCs are often due to high residential indoor concentrations. Indoor sources for VOCs were identified in *source attribution analyses* on the *EXPOLIS*-Helsinki data. The most influential indoor sources identified were cleaning products (Acetone, Terpenes, Aldehydes, 1-Butanol, Hexanal), building products (Octanal, Formaldehyde, Terpenes, Acetaldehyde and Benzaldehyde) and air refresheners (Limonene) (3, 4).

Also Naphthalene has been associated with bitumen, which has been used in buildings for moisture sealing. In the *EXPOLIS* study, e.g., in the sub-sample of high Naphthalene exposures in Athens, all were due to high home indoor concentrations (100 - 1000 µg/m^3), and home - but not workplace - naphthalene concentrations were also the determining factors in the Naphthalene exposures of 2/3 of the other study subjects. (5) In most of these cases, however, the source of Naphthalene was mothballs, not building materials.

The simplest approach for assessing the relative contributions of indoor sources to indoor exposures, is to look at the indoor/outdoor (I/O) concentration ratios, which have been extensively reported since the late '70's. Ratios close to 1.0 indicate outdoor air as the main contributor to indoor exposure. High ratios up to 10 and above indicate that indoor sources dominate the indoor concentrations. This approach is, however, not reliable for compounds with high reactivity or affinity to indoor surfaces. In the *EXPOLIS* study in Helsinki, the mean I/O ratio was highest for acetone (145) from human metabolism followed by formaldehyde (6, 3).

Indoor sources contribute also to bioaerosols (Nevalainen 2007) and combustion products (Jantunen 2007), which are covered in these proceedings under session 3 (see page 90) and session 5 (see page 164) respectively.

CHEMICAL EMISSIONS FROM SURFACE MATERIALS - CASE STUDIES

Various types products have been reported as primary emission sources on-site in buildings, e.g. linoleum flooring (7), polysulfide sealant (8), alkyd paint for radiators (9, 10), water repellent (11) and preparations based on coal tar such as damp proof membranes (12), wood preservatives (1). A case example of xylamite as source of indoor air pollution is presented by Prejzner et al. 2007 on page 219 of these proceedings.

The following overview of technically well-documented case studies has been limited to polymeric surface materials in direct contact with the indoor air such as textile flooring, resilient flooring and latex paint. Polymeric materials may emit residual monomers, process solvents and other functional chemicals during the use-phase.

In most of the case studies complaints had been made of the indoor climate and the substances concerned had been released over periods of several years. Bearing in mind the fact that a chemical substance that has been proven to be present in indoor air may have various origins, the indicated source of emission have been identified by an unmistakable odour or by parallel investigations, e.g. in a climate chamber. If not otherwise stated the presented case studies are selected from European countries.

Cork tiles – phenol

Plastic-laminated cork tiles laid on the floor in an office had a very strong odour, even when being unpacked and laid. Chemical measurements in the newly-built part of the office a year later indicated the presence of **phenol** (15 $\mu g/m^3$) in the air. The cork tiles consisted of a wearing layer of transparent vinyl plastic, and the layer of cork beneath it contained phenolic resin. It was shown that phenol was released from the material, with the result that it was decided as essential to replace the floor covering. The manufacturer of the material has subsequently modified the phenol-based resin.

Vinyl flooring – dodecylbenzene

Alkyl benzenes, among them being **dodecylbenzene**, are common process solvents for plasticizers in the manufacture of **vinyl floor coverings**. This type of process solvent has been detected in several investigations of indoor air (100-200 µg/m^3) resulting in complaints about the air quality.

<u>Vinyl flooring – dodecene</u>

Shortly after moving into an office building several employees complained about the air quality. It can be estimated that the concentration of **dodecene** in the premises was 40 µg/ m^3. Dodecene is used as a solvent in the manufacture of **vinyl floor covering**, but in this particular case dodecene was a decomposition product produced during manufacture of the floor covering, and originating from dodecylbenzene.

<u>Vinyl flooring – TXIB</u>

In several public buildings "**TXIB**" have been present in the indoor air, in the range of 100 µg/ m^3. TXIB stands for 2,2,4-trimethyl-1,3-pentadioldiisobutyrate, which is used as a process solvent in the manufacture of certain **vinyl floor coverings**. Extraction of one type of vinyl floor covering showed that it contained 7% TXIB.

<u>Textile carpet - 4PC</u>

4-phenylcyclohexene (4-PC) has been detected in offices in which **wall-to-wall carpets** have been laid. Four weeks after laying, the concentrations where about 15 µg/ m^3. 4-PC can be formed by the manufacture of the layer of styrene-butadiene rubber used to bind the fibres to the carpet backing. This undesirable and odorous substance is formed of the reaction of styrene and 1,3 butadiene. The presence of the substance has aroused considerable attention in the USA, where 4-PC has been found in the EPA main building in Washington D.C. and elsewhere.

<u>Textile carpet – styrene</u>

Ever since it was built, a strong smell was noticeable in a school (Texas, USA). The average ventilation rate was 3 air changes/h. After eight years, measurements were made, and found up to 0.9 ppm of **styrene** in the air. The styrene was found to be coming from the underside of the **wall-to-wall carpet** that covered an area of 6500 m^2 where the underside consisted of foamed styrene-butadiene rubber.

<u>Vinyl flooring – EHA</u>

Chemical measurements in a school indicated the presence of **ethylhexyl acrylate** (about 30 µg/ m^3) in a corridor outside a classroom. Emissions from samples of **vinyl floor coverings** were also investigated, including those from a sample of unused floor covering that had been kept in the school store. More than five years after delivery, this floor covering was found to be releasing several different substances, including ethylhexyl acrylate. This was most probably coming from the surface varnish layer of the floor covering. This layer of varnish is intended to be cured by UV light, and ethylhexyl acrylate can remain as a residual monomer if the layer is incompletely cured.

Latex paint – butylphtalate

In over 60 both newly-built and older renovated residential buildings it was noticed that pot plants were not growing normally and that in many cases the new shoots were completely white. It was also noted that paint on the walls was not completely dry in the room where the affected plant were kept. The type of paint could be identified in several of the cases and in all of them a certain **latex paint** from one and the same manufacturer had been used.
Test plants were kept under glass covers, together with a sample of the latex paint. It was found that the white shoots occurred only in presence of those of the manufacturer's paints that contained **dibutyl phtalate** (DBP). The concentration of DBP in the room air where the discolouration first was observed was 40 $\mu g/m^3$.

Textile carpet - ethylhexanol etc

Two years after completion as a building in (Oregon, USA), complaints were received about a smell of **2-ethylhexanol** in concentrations ranging from 34 $\mu g/m^3$ to 138 $\mu g/m^3$. **Heptanol and nonanol** were also detected in the air. A laboratory investigation showed that the alcohols came from a PVC layer on the underside of the wall-to-wall carpet. The PVC layer contained the plasticizers (phthalate esters) diethylhexyl phthalate (DEHP) and heptylphthalate and nonylphthalate to make the carpet soft and easier to lay. Surprisingly, the PVC layer also contained a considerable amount of calcium oxide, resulting in a high pH in this layer and the phthalate esters were decomposed by alkaline hydrolysis. Complete hydrolysis of a phthalate plasticizer results in its irreversible decomposition to phthalic acid and a volatile and usually odorous alcohol. The ester hydrolysis is catalysed by hydroxyl ions.

MOISTURE INDUCED CHEMICAL DEGRADATION OF MATERIALS

Besides the release of substances to the indoor air due to primary emission, damp building materials give rise to volatile substances formed during secondary reactions. Moisture induced chemical degradation of e.g. vinyl flooring, floor adhesive and self-levelling flooring compound may give rise to the formation of odorous substances. In most of cases, the substances concerned have been released over a period of several years.

Flooring materials – ethylhexanol

The phthalate plasticizer content in resilient vinyl floor coverings can amount to 30% of the weight of the material. A large number of cases have been reported especially from the Nordic countries concerning a heavy, sickly smell from vinyl floor coverings as a result of hydrolysis of phthalate plasticizers (13). Until the last years the most common phthalate plasticizer in floor coverings has been diethylhexyl phthalate (DEHP) giving rise to 2-ethylhexanol, with reported indoor concentrations ranging from about of 10-30 $\mu g/ m^3$ in various premises.

The chemical degradation of the plasticizer is strongly accelerated by the presence of alkali.

All concrete is alkaline, and the dampness of the concrete is of considerable importance for the formation of odorous alcohols. Self-levelling flooring compounds based on Portlandcement, used to smooth and level the surface of concrete floors, do not differ significantly in chemical terms from concrete. This means that damp self-levelling compound ("smoothing compound"), too, can result in hydrolysis of the plasticizer in vinyl floor covering.

The types of adhesives most commonly used for laying floor coverings are **dispersion-based**. This type of adhesive is often based on acrylate copolymers of 2-ethylhexacrylate. **2-ethylhexanol** is a feedstock material (an estering alcohol) for the manufacture of 2-ethylhexylacrylate. In the same way as plasticizers, 2-ethylhexylacrylate can be hydrolysed by damp alkaline concrete, thus reforming the alcohol, which can be smelt.

Vinyl flooring – cresol

An unpleasant smell of horse stable was noticeable in newly-built extension to a hospital. Chemical measurements indicated the presence of **phenol and p-cresol**, both in concentrations around 10 $\mu g/m^3$. Laboratory investigation of the **vinyl floor covering** in the premises also indicated emission of these substances. p-cresol occurs naturally in horses' urine, which can explain the strange smell in the premises. The floor covering contained phosphorus plasticizer which degraded as a result of the effect of damp concrete. Cresol and phenol are decomposition products from this type of plasticizer. The same type of floor covering, lying on old and drier floor/ceiling structures, exhibited no noticeable release of phenol or cresol. Vinyl floor coverings containing phosphorus plasticizer are used in premises with particularly stringent fire safety requirements.

Self-levelling flooring compound – amines

Self-levelling flooring compounds based on Portlandcement and containing casein as a levelling agent may give rise to odorous substances when laid on a damp subfloor. In alkaline conditions, casein and other proteins break down as a result of hydrolysis to form volatile amines and other substances. A breakdown substance of casein, **ortho-aminoacetophenone**, has a very unpleasant odour that can be recognised at concentrations as low as of the order of nanogram/m^3. Ortho-aminoacetophenone, is formed as a result of breakdown of the amino acid tryptophane. Portland cement-based casein-containing self-levelling compound may also release ammonia, which is capable of discolouring oak parquet laid on top of it.

CHAMBER TESTING OF PRODUCTS AS A TOOL TO IDENTIFY CHEMICAL EMISSIONS

Building materials shall meet official requirements and market demands on e.g. mechanical strength, fire resistance and be resistant against humidity variations, abrasion and the influence of microbiological and chemical degradation. In order to maintain intended properties of the material various functional chemicals are used. For hygienic, health and environmental reasons and in order to maintain the properties of the material in the construction, it is important that the functional chemicals remain bound in the material during the use-phase and are not released into the ambient environment. Therefore, laboratory studies are needed, where the potential for release of substances under the intended use conditions are estimated. Such studies should preferably be performed under experimental conditions in test chambers corresponding to relevant exposure scenarios.

Test chambers methods are used for:

- official approval procedures
- voluntary labeling
- development of low-emitting products

The use of standardised scenarios and emission test methods are important, especially when building materials are examined in relation to official approval procedures. Test chambers are mainly used for the determination of area specific emission rate, (µg/m2 x h) at constant temperature, relative humidity and air exchange rate. Emission test chambers are designed to permit the testing of samples of various types of materials and can range in size from mL to several m^3 (14). Portable emission cells, where the test material becomes a part of the cell itself, can be used for identifying emission sources on-site in buildings (15, 16).

EC CONSTRUCTION PRODUCTS DIRECTIVE AND HEALTH PROTECTION

The Construction Products Directive (2) has been established to examine construction product safety and to break down technical barriers to trade in the European Economic Area. Construction products are defined as all products permanently incorporated in construction works, including both buildings and civil engineering works.

In addition to the traditional essential requirements such as safety in case of fire, stability and mechanical resistance, the CPD also requires that hygiene, health and environmental protection shall be considered. All essential requirements concerning the use-phase of the construction products are covered.

Essential requirements (ER) defined in CPD:
ER 1 Mechanical resistance and stability
ER 2 Safety in case of fire
ER 3 Hygiene, health and the environment (e.g. substances in indoor air)
ER 4 Safety in use
ER 5 Protection against noise
ER 6 Energy economy and heat retention

The essential requirements are intended to be incorporated in several hundred harmonised technical specifications [hTS] such as European standards [EN] and technical approvals [ETA] for construction products. After a transitional period, construction products may only be placed on the market if they conform to the specifications and display the CE mark.

The aim of the environmental requirement (ER3) is to protect the health of occupants and neighbours and the immediate environment, which essentially comprises the e.g. indoor air. The implementation of the environmental criteria requires test methods especially designed for the determination of the release of dangerous substances from construction products. The European Commission has therefore given the task to CEN to develop standardised test methods for the release of dangerous substances (17).

CURRENT STANDARDISATION OF EMISSION TEST METHODS

Since 2006 a Technical Committee (18) at CEN is responsible for developing environmental test methods under the CPD. The end objective is the elaboration of European standards [EN] for environmental properties. These standards will be supporting standards which are referred to in the forthcoming revised technical specifications for construction products. The methods shall take into account the intended conditions of use of the products (release scenarios) and address emission to indoor air, release to soil, surface water and ground water.

The environmental test methods shall preferably be applicable to all or most product groups ("horizontal methods") which may release substances under a specific release scenario. For the release into indoor air horizontal test methods applicable to several surface materials have been developed (19).

Within the framework of the REACH Regulation (20) e.g. the release of chemical substances from articles and preparations has to be assessed. Construction products are not exempted from this obligation. The horizontal release methods developed within the framework of CPD will therefore support the intention of REACH with respect of construction products.

CONCLUSIONS

Emission from construction products to the indoor air have been reported for a wide range of substances including those formed during secondary reactions causing complaint of irritation and odour. Emission testing of construction products is therefore an important tool to identify chemical emissions that could cause problems with the indoor air quality.

The Construction Products Directive [89/106/EEC] is now addressing emission of dangerous substances including to indoor air and standardised test methods are under development by CEN. The REACH regulation [EC1907/2006] being implemented from June 2007 will also impact on the control of emissions from articles including construction and consumer products.

REFERENCES

1. Gustafsson H: Building Materials Identified as Major Emission Sources for Indoor Air Pollutants - a critical review of case studies, D10:1992, Swedish Council for Building Research.

2. 89/106/EEC: Council Directive of 21 December 1988 on the approximation of laws, regulations and administrative provisions of the Member States relating to construction products (Construction Products Directive - CPD).

3. J.A. Jurvelin, R.D. Edwards, M. Vartiainen, P. Pasanen and M.J. Jantunen. Residential Indoor, Outdoor, and Workplace Concentrations of Carbonyl Compounds: Relationships with Personal Exposure Concentrations and Correlation with Sources. *J. Air & Waste Manage. Assoc.* 53 (2003) 560-573.

4. R.D. Edwards, J. Jurvelin, K. Koistinen, K. Saarela and M. Jantunen. VOC source identification from personal and residential indoor, outdoor and workplace microenvironment samples in EXPOLIS-Helsinki, Finland. *Atmos. Environ*. 35 (2001) 4829–4841.

5. R.D. Edwards, C. Schweizer, M.J. Jantunen, H.K. Lai, L. Bayer-Oglesby, K. Katsouyanni, M.J. Nieuwenhuijsen, K. Saarela, R. Srám and N. Künzli. Personal exposures to VOC in the upper end of the distribution — relationships to indoor, outdoor and workplace concentrations. *Atmos. Environ.* 39 (2005) 2299-2307.

6. J.A. Jurvelin, M. Vartiainen, P. Pasanen, M.J. Jantunen. Personal exposure levels and microenvironmental concentrations of formaldehyde and acetaldehyde in Helsinki Metropolitan Area, Finland. *J. Air Waste Manage. Assoc.* 51 (2001) 17-24.

7. Zellweger, C. et al. (1997): Emissions of Volatile Organic Compounds (VOC) from Building Materials - Methods and Results, pp. 74-80, p. A42, Bundesamt für Energiewirtschaft.

8. Wolkoff, P. (1987): Analyse af organiske luftforureninger i provesal B på det Kgl. teater, 86-423-2635, Arbejdsmiljøinstitutet, Danmark.

9. Ullrich, D, et al. (1982): Einfluss von Lackanstrichen auf die Innenraumluftqualität am Beispiel von Heizkörperlacken. In Luftqualität in Innenräumen, Ed by Aurand K. et al pp. 283-298. Gustav Fischer, Stuttgart.

10. Seifert, B. & Ullrich, D (1987): Methodologies for evaluating sources of volatile organic compounds (VOC) in homes, Atm. Envir. (21), pp. 395-404.

11. Bloeman, H.J. et al. (1990): Indoor Air Pollution after the Application of Moisture Repellent, Proc. 5 th Int. Conf. on Indoor Air Quality and Climate, Vol 3, 569-574, Toronto.

12. Brown, V. M. et al. (1990): Investigations of the volatile organic compound content of indoor air in homes with an odorous damp proof membrane, Proc. 5 th Int. Conf. on Indoor Air Quality and Climate Vol. 3, 575-580, Toronto.

13. Gustafsson, H: Einwirkung von feuchten Betonuntergründen auf Kleber und Fuβbodenbeläge - Übersicht und Kommentare zur Laboruntersuchungen über chemische Abbauprozeβe und Emissionen flüchtiger chemischer Verbindungen, SP Bericht 1996:25.

14. ECA: Guideline for the Characterization of Volatile Organic Compounds Emitted from Indoor Materials and Products Using Small Test Chambers, European Concerted Action - Indoor Air Quality and Its Impact on Man, COST Project 613, Report No 8, Commission of The European Communities, Joint Research Centre, 1994

15. Wolkoff, P. et al. (1991):. Field and Laboratory Emission Cell: FLEC, Conf. Proceedings "Healthy Buildings 91", Sept 4-8, pp. 160-165, Washington D.C.

16. Gustafsson, H. (1999): Field and Laboratory Emission Cell (FLEC), Chapter 12, pp. 143-152, In ”Organic Indoor Air Pollutants”, Ed. T Salthammer, Wiley-VHC.

17. Mandate/366EN: Horizontal complement to the Mandates to CEN/CENELEC, concerning the execution of standardisation work for the development of horizontal standardised assessment methods for harmonised approaches relating to dangerous substances under the construction products directive (CPD) - Emission to indoor air, soil, surface water and ground water, European Commission, Brussels, 16th March 2005.

18. CEN/TC 351: Construction Products: Assessment of Release of Dangerous Substances (www.centec351.org)

19. ISO 16009--11: Emission test chamber method - ISO16009; Emission test cell method - ISO16010; Sampling, storage of samples and preparation of test specimens - ISO16011.

20. Reach: Regulation (EC) No 1907/2006 of the European Parliament and of the Council of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), establishing a European Chemicals Agency, amending Directive 1999/45/EC and repealing Council Regulation (EEC) No 793/93 and Commission Regulation (EC) No 1488/94 as well as Council Directive 76/769/EEC and Commission Directives 91/155/EEC, 93/67/EEC, 93/105/EC and 2000/21/EC.