

Emission of Volatile and Semivolatile Organic Compounds from Waterborne Paints - The Effect of the Film Thickness

Per Axel Clausen¹

Abstract

A waterborne paint was applied to tin plated steel sheets with three different film thicknesses. The emission of volatile and semivolatile organic compounds ((S)VOCs) from the samples was measured in small climatic chambers under standard conditions over a two-week period. The purpose of the study was to evaluate the effect of the film thickness on the emission rate decay. First order decay models, including sink effects for the high boiling (S)VOCs, were fitted to the concentration versus time data. The results showed that the first order rate constants decrease with increasing film thickness. In addition, the results indicated that the emissions of the (S)VOCs in the waterborne paint film seemed to be controlled by evaporation. The thicknesses of paint films used in climatic chamber tests to estimate emission rates for product comparison or emission prediction must be known in order to prevent erroneous conclusions.

Introduction

Waterborne paints used for ceilings, walls and woods are major potential indoor sources of volatile and semivolatile organic compounds ((S)VOCs) (Hansen et al., 1987; Van Faassen and Borm, 1991; Clausen et al., 1991). (S)VOCs may be one factor that can cause the sick building syndrome (SBS) (e.g. Hodgson et al., 1991; Sundell et al., 1993). Semivolatile organic compounds (SVOCs) are defined as compounds with vapour pressures from approximately 10^{-2} to 10^{-8} kPa (10^{-1} to 10^{-7} torr) at ambient temperatures (Clements and Lewis, 1988); they exist to a high degree in the condensed phase. Nevertheless, SVOCs are easily emitted from waterborne paint films for more than a year (Clausen et al., 1991), they can be found in the indoor air more than a year after application (Wolkoff et al. 1991), and they can be found in floor dust (Wolkoff and Wilkins, 1993). These properties of SVOCs may be of importance in relation to the SBS.

The emission of (S)VOCs from waterborne paints can be evaluated in climatic chamber tests in which the environmental variables are controlled (air exchange rate, temperature, relative humidity and loading). An overlooked variable in emission testing is the thickness of the paint film. The film thickness has for several years commonly been used as a variable in paint research (e.g. Hansen, 1974; Sullivan, 1975; Newman and Nunn, 1975). The relevance of the thickness of waterborne paint films has only once been considered in the indoor air literature (Clausen et al. 1991).

The purpose of this paper is to demonstrate by modelling the effect of variation in the thickness of a waterborne paint film on the emission rate decay of (S)VOCs and to compare the results with the theory. SVOCs may have strong sink effects because of their ability to adsorb to surfaces. Therefore the models have to incorporate sink effects.

KEY WORDS:

Waterborne paints, Emission, Volatile organic compounds, Semivolatile organic compounds, Film thickness.

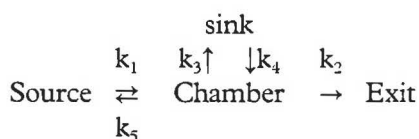
¹ National Institute of Occupational Health, Lersø Parkalle 105, DK-2100 Copenhagen Ø, Denmark
Fax no. +45 39 27 01 07

Theory

It is commonly assumed that the emission rates of (S)VOCs from finite thin film sources decrease approximately by a first order decay, as shown in Equation 1.

$$R = M_0 k_1 \exp(-k_1 t) \quad (1)$$

where R = emission rate ($\text{mg} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$), M_0 = initial (S)VOC mass in the source ($\text{mg} \cdot \text{m}^{-2}$), k_1 = first order rate constant (h^{-1}), and t = time (h). Models which mathematically try to decouple the true emission characteristics of an organic source from effects imposed by a climatic chamber have been proposed by Dunn and Tichenor (1988). These models assume that a climatic chamber consists of four (mathematical) compartments. These are the source, the homogeneous chamber air, an exit, and a sink. First order rate constants describe the flows among these compartments as illustrated in the following scheme:



The solution to the so-called dilution model (i.e. neglecting sink effects (k_3, k_4) and vapour pressure effect (k_5)), given that $C=0$ when $t=0$, gives the following expression for the chamber concentration:

$$C = AM_0 k_1 (\exp(-k_1 t) - \exp(-k_2 t)) / (V(k_2 - k_1)) \quad (2)$$

where C = chamber air (S)VOC concentration ($\text{mg} \cdot \text{m}^{-3}$), A = area of source (m^2), k_2 = air exchange rate (h^{-1}), and V = chamber volume (m^3). The solution to the so-called sink model (i.e. neglecting vapour pressure effect), given that $C=0$ when $t=0$, gives the following expression for the chamber concentration:

$$\begin{aligned}
 C = AM_0 k_1 \{ & (k_4 - k_1) \exp(-k_1 t) / [(r_1 - k_1)(r_2 - k_1)] \\
 & - (k_2 + k_3 - r_2) \exp(-r_1 t) / [(r_1 - r_2)(r_1 - k_1)] \\
 & + (k_2 + k_3 - r_1) \exp(-r_2 t) / [(r_1 - r_2)(r_2 - k_1)] \} / V,
 \end{aligned} \quad (3)$$

$$r_2, r_1 = \{k_2 + k_3 + k_4 \pm [(k_2 + k_3 + k_4)^2 - 4k_2 k_4]^{1/2}\} / 2$$

where k_3, k_4 = first order rate constants describing the sink effects (h^{-1}).

Emission of (S)VOCs from Waterborne Paints

The emission of (S)VOCs from a waterborne paint film prior to dry film formation may be regarded as being controlled by evaporation, i.e. limited by diffusion through a boundary layer above the paint film (Kornum, 1980). The emission after dry film formation may be controlled either by evaporation or by internal diffusion in the paint film. This depends upon the (S)VOC involved and the interactions which occur between different (S)VOCs (Sullivan, 1975).

Evaporation-Controlled Emission

The mass transport of a (S)VOC from the surface and through the boundary layer above a dry paint film becomes the limiting step of the emission when this mass transport is much slower than within the dry paint film. The emission can then be considered as evaporation from a liquid. Assuming that the diffusion in the boundary layer is controlled by Fick's first law, that the diffusion coefficient D ($\text{m}^2 \cdot \text{h}^{-1}$) is constant, and that a linear gradient has been established, the flux F ($\text{mg} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$) is given as:

$$F = D(C_{\text{sur}} - C) / \delta \quad (4)$$

where C_{sur} = (S)VOC concentration at the paint film surface, and δ = apparent laminar boundary layer thickness (m). Assume that the chamber (S)VOC concentration is much lower than the concentration at the paint film surface ($C \ll C_{\text{sur}}$) and that this concentration is directly proportional to the (S)VOC paint film concentration ($C_{\text{sur}} = \beta C_s$). Thus, expressing the (S)VOC paint film concentration by the mass per unit area and the film thickness ($C_s = M_s / L$) Equation 4 can be expressed as:

$$F = D\beta M_s / L\delta \quad (5)$$

where β = proportionality constant, M_s = (S)VOC mass in the source ($\text{mg} \cdot \text{m}^{-2}$), and L = thickness of the source (m). Defining that $k_{\text{E1}} = D\beta / \delta$, the mass balance for the (S)VOC in the paint film is:

$$A(dM_s / dt) = -AM_s k_{\text{E1}} / L \quad (6)$$

where k_{E1} = rate constant of evaporation-controlled

emission for $L = 1 \mu\text{m}$ ($\mu\text{m} \cdot \text{h}^{-1}$). The emission rate of (S)VOC from the paint film is then given as:

$$R = M_0(k_{E1}/L) \exp(-(k_{E1}/L)t) \quad (7)$$

This expression is analogous to Equation 1 and has previously been obtained empirically (Clausen et al. 1991). Dilution and sink model expressions for the evaporation-controlled emission can be obtained by substituting k_{E1}/L for k_1 in Equations 2 and 3.

Diffusion-Controlled Emission

The mass transport of a (S)VOC in a dry paint film becomes the limiting step of the emission when it is much slower than the mass transport from the surface and through the boundary layer above the paint film. Assume that the diffusion within the paint film is controlled by Fick's Second Law, that the diffusion coefficient D is constant, that a relatively stable concentration profile has been established in the source with a very low concentration at the surface (that can be approximated by zero), and that the flux is zero at the substrate. Using these assumptions and defining $k_{D1} = D(\pi/2)^2$, it has been shown (Clausen et al., 1993) that the emission rate can be expressed as:

$$R = M_0(k_{D1}/L^2) \exp(-(k_{D1}/L^2)t) \quad (8)$$

where k_{D1} = rate constant of diffusion-controlled emission for $L = 1 \mu\text{m}$ ($\mu\text{m}^2 \cdot \text{h}^{-1}$). This expression is analogous to Equation 1 and has previously been obtained empirically (Clausen et al., 1991). Dilution and sink model expressions for the diffusion-controlled emission can be obtained by substituting k_{D1}/L^2 for k_1 in Equations 2 and 3.

Materials and Methods

Selection of Paint

A common white pigmented waterborne wall paint containing a broad spectrum of (S)VOCs with respect to polarities and boiling points was selected.

(S)VOC Measurements

Before the chamber test, the paint was examined by headspace techniques using thermal desorption and gas chromatography combined with Fourier transform infrared spectrometry (Clausen et al., 1990). The measurements of the chamber concentrations were performed by chamber air sampling on Tenax TA, thermal desorption and subsequent analysis by

capillary gas chromatography with flame ionization detection (Wolkoff, 1990). The sampling flow was nominal 40 ml/min or 80 ml/min and the sampling volumes were from 600 ml to 9600 ml at the end of the test period. Breakthrough was checked for high volume samples with sample backup tubes. Complete desorption of the cold trap was checked by a succeeding run of a blank tube and complete desorption of sample tubes was checked by a cleaning procedure identical to the analysis.

Chambers

The three electropolished stainless steel chambers (without fans) had a volume of 0.234 m^3 resulting in a chamber loading of $0.36 \text{ m}^2/\text{m}^3$ for the paint samples. The test conditions were $23.0 \pm 0.6^\circ\text{C}$, $45 \pm 3\%$ RH, and an air exchange rate of $0.25 \pm 0.013 \text{ h}^{-1}$. The air exchange rate in the chamber of the $28 \mu\text{m}$ sample was estimated from the emission curves because the valve controlling the airflow through the chamber had failed. It was found to be approximately 0.21 h^{-1} . The exact value of the air exchange rate is of less importance for the estimated values of the first order rate constants (k_1) and is without any significance for the conclusions. The chambers complied with a Nordtest method (NT Build 358, 1990).

Sample Preparation

The paint was applied to tin plated steel sheets ($250 \text{ mm} \times 341 \text{ mm}$ each) with three different film thicknesses. The application was conducted by a short haired (synthetic) paint roller in order to obtain a surface similar to a painted wall. Before application the steel sheets were cleaned by heating to 150°C for 4 days. In order to obtain the intended film thicknesses of 20, 40, and $60 \mu\text{m}$, the substrate (the tin plated steel sheet) was placed on a balance, the calculated amount of paint was poured onto the substrate, and the paint was distributed with the paint roller which was pre-wetted with paint to get a zero uptake/delivery of paint from the substrate. The ratios of the amounts of paint applied to the substrates were approximately 1:1.3:2.6 which correspond to the ratios of the dry film thicknesses. Based on previous emission experiments, the reproducibility of the (S)VOC measurements, and the theory, it was estimated that these ratios were sufficiently large to obtain a different result for each sample. Immediately after application the samples were placed in a large office for conditioning for 24 hours in order to avoid the strong emission during the

drying phase (Clausen et al., 1991). The background concentrations of the relevant (S)VOCs in the office were below the detection limits and the temperature and the relative humidity varied in the range from 22 to 23°C and from 37 to 40% RH, respectively.

Chamber Tests

The tests were conducted in compliance with a Nordtest method (NT Build 358, 1990). Prior to the tests the chambers were cleaned and background measurements were carried out. The samples were placed on the bottom part of the chambers. The test start (time = 0) was established when the doors to the chambers were closed. During 2 weeks 12 duplicate samples for each paint sample were taken. When the tests were terminated, 100 measurements of the dry film thickness on each paint sample were carried out by use of a calibrated coating thickness gauge which operates by means of electromagnetic induction (Elcometer 300).

Data Treatment and Calculations

The model expressions were fitted to the concentration versus time data with STATGRAPHICS routine for nonlinear regression using the standard regression parameters of the program (STATGRAPHICS, 1987). The duplicate samples were used as two individual data points at the nonlinear regression. The average film thicknesses of the samples were used in the calculations.

Results

Table 1 shows the broad spectrum of (S)VOCs with respect to physical/chemical properties identified in the paint and the abbreviations used in this paper.

Table 1 The (S)VOCs identified in the paint and their vapour pressures

| (S)VOC | Abbreviation | Vapour pressure (torr, 23°C) |
|--|---------------------|------------------------------|
| white spirit | white spirit | 0.69–3.9 ^a |
| 1,2-propanediol | propanediol | 0.11 ^b |
| 2-(2-butoxyethoxy)-ethanol | butoxyethoxyethanol | 0.014 ^b |
| 2,2,4-trimethyl-1,3-pentenediol mono-isobutyrate | Texanol | 0.07 ^c |

^a Calculated approximate value based on the boiling points.

^b DIPPR Data Compilation of Pure Compound Properties, American Institute of Chemical Engineers, 1989.

^c Calculated approximate value based on the boiling point and a vapour pressure of 1 torr at 87°C.

Figure 1 shows the concentration versus time data for the three samples. The 24 hours' conditioning of the samples prior to insertion into the chambers reduced the white spirit content below the detection limit in the chambers and reduced the contents of the other (S)VOCs more than expected. This expectation was based on a previous experiment in which the paint sample was inserted into the chamber immediately after application of the paint. This resulted in high initial (S)VOC concentrations (200 mg/m³) which may have suppressed the initial emission by vapour pressure effect, and thus resulted in a relatively high (S)VOC content in the paint film after 24 hours. This effect was overlooked when the chamber concentrations were estimated for the actual experiment, resulting in the observed low concentrations. The low propanediol concentrations are very uncertain because they were very close to the detection limit. Some concentration data of the (S)VOCs are missing because they were below the detection limits.

Table 2 shows the measured film thicknesses of the paint samples. The large standard deviations

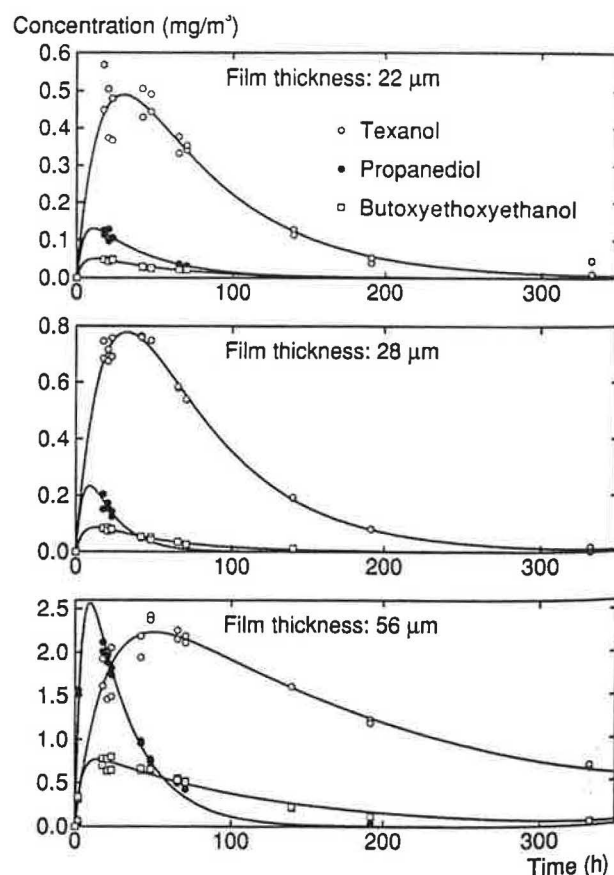


Fig. 1 The concentration versus time data of the three paint samples with different film thicknesses and the modelled emission curves

Table 2 The average film thicknesses of the three paint samples.

| Sample | Average film thickness (μm) | Standard deviation | Number of observations |
|------------------|--|--------------------|------------------------|
| 22 μm | 22.02 | 11.04 | 100 |
| 28 μm | 27.66 | 15.17 | 100 |
| 56 μm | 56.27 | 19.11 | 100 |

illustrate the very uneven surfaces caused by the paint roller application.

Table 3 shows the estimated model parameters and Figure 1 the corresponding model curves. It was found that the chamber concentrations of propanediol and butoxyethoxyethanol did not suffer significantly from sink effects and therefore Equation 2 was used to model the emission of these (S)VOCs. On the contrary, the Texanol concentrations suffered from sink effects which are reflected by the delayed summits of the model emission curves shown in Figure 1. In addition, it was found that the sink rate constants k_3 and k_4 were much larger than k_1 . Thus the sink effects are governed by the ratio of k_3 and k_4 only. The model parameters of the Texanol emission shown in Table 3 were estimated by Equation 3 using a fixed value of $k_4 = 10$.

Discussion

Emission Rate Constant and Film Thickness

According to the theory, an increased film thickness should result in a decreased emission rate constant (k_1) because $k_1 = k_{E1}/L$ for the evaporation-controlled emission or $k_1 = k_{D1}/L^2$ for the diffusion-controlled emission (L = the film thickness). The estimate of k_1 for propanediol of the 22 μm sample in Table 3 is low compared to the 28 μm and 56 μm

Table 4 Mean and relative standard deviation of the different first order rate constants (k_1 , k_{E1} , and k_{D1}) for three (S)VOCs based on three samples with different film thicknesses

| (S)VOC | Propanediol | Butoxyethoxyethanol | Texanol |
|------------------------------|-------------|---------------------|-------------|
| Mean of k_1 (rel. std.) | 0.039 (41%) | 0.016 (38%) | 0.012 (52%) |
| Mean of k_{E1} (rel. std.) | 1.4 (54%) | 0.48 (16%) | 0.35 (27%) |
| Mean of k_{D1} (rel. std.) | 57 (92%) | 17 (57%) | 12 (36%) |

samples. This is probably due to the very uncertain measurements of the low concentrations. A decrease of k_1 for the 56 μm sample compared to the 28 μm sample is observed. For butoxyethoxyethanol and Texanol no difference is found between the k_1 estimates of the 22 μm and 28 μm samples. This may be due to a too large uncertainty of the (S)VOC measurements in proportion to the thickness difference of these samples. A strong decrease of the k_1 estimates for the thick 56 μm sample for butoxyethoxyethanol and Texanol is observed.

Emission Mechanism and Reproducibility

The values in Table 3 of the first order rate constants k_{E1} and k_{D1} for the evaporation- and diffusion-controlled emission, respectively, were estimated in the same way as described for k_1 . The estimates of k_{E1} should be reproduced for the three different film thicknesses if the emission is controlled by evaporation. Also, this should be the case for k_{D1} if the emission is controlled by diffusion. The reproducibility of k_1 , k_{E1} , and k_{D1} for the three samples expressed by the relative standard deviation is shown in Table 4.

For propanediol, k_1 is best reproduced but assuming that the emission must be either evaporation- or diffusion-controlled, it is most probable

Table 3 The estimated first order decay model parameters for three different (S)VOCs from the three samples (the standard error of the last digit is in brackets)

| (S)VOC | Sample | k_1 (h^{-1}) | k_{E1} ($\mu\text{m} \cdot \text{h}^{-1}$) | k_{D1} ($\mu\text{m}^2 \cdot \text{h}^{-1}$) | M_0 ($\text{mg} \cdot \text{m}^{-2}$) | k_3/k_4 | N | r^2 |
|---------------------|------------------|------------------------------|---|---|--|-----------|----|-------|
| Propanediol | 22 μm | 0.025 (2) | 0.56 (5) | 12 (1) | 4.6 (3) | – | 12 | 0.98 |
| | 28 μm | 0.057 (9) | 1.6 (3) | 44 (7) | 3.9 (2) | – | 10 | 0.96 |
| | 56 μm | 0.036 (2) | 2.0 (1) | 114 (6) | 68 (2) | – | 20 | 0.98 |
| Butoxyethoxyethanol | 22 μm | 0.018 (1) | 0.40 (3) | 8.7 (1) | 2.4 (1) | – | 16 | 0.97 |
| | 28 μm | 0.020 (1) | 0.55 (3) | 15.1 (8) | 3.2 (1) | – | 18 | 0.99 |
| | 56 μm | 0.0089 (7) | 0.49 (4) | 28 (2) | 68 (4) | – | 24 | 0.97 |
| Texanol | 22 μm | 0.015 (3) | 0.33 (7) | 7 (2) | 35 (3) | 2.9 (9) | 22 | 0.94 |
| | 28 μm | 0.016 (1) | 0.46 (4) | 13 (1) | 46 (1) | 2.9 (3) | 22 | 0.99 |
| | 56 μm | 0.0048 (6) | 0.27 (4) | 15 (2) | 410 (30) | 3.8 (6) | 24 | 0.96 |

that the emission is evaporation-controlled. For butoxyethoxyethanol it is most probable that the emission is controlled by evaporation. For Texanol it is weakly indicated that the emission may be controlled by evaporation. Wolkoff and Wilkins (1993) have also found that the Texanol emission from a waterborne paint film was controlled by evaporation because the first order rate constant increased with increasing air exchange rate (air velocity).

Sample Preparation, Test Conditions, and Model

A thick film of a given paint has a larger (S)VOC mass than a thin film but they have the same surface concentration. Assuming no influence of the chamber (S)VOC concentration they will have identical initial emission rates but the thick film emission decays slower because of the larger (S)VOC mass. An uneven paint film surface (e.g. caused by paint roller application) can have local very different film thicknesses (see the large standard deviations in Table 3) but the evaporation-controlled emission rate decay is the same all over the surface because of the homogeneous concentration. This is not the case for the diffusion-controlled emission where the areas with thin film have a faster decrease of (S)VOC content than areas with thick film. The emission cannot be considered as the sum of several independent sources because the (S)VOCs may diffuse from the thick to the thin film areas. Thus the paint roller application may have caused problems for the model if the emission had been diffusion-controlled. But the low air exchange rate and thereby the low air velocity probably results in a thick boundary layer, thus supporting the evaporation-controlled emission.

The 24 hours' storing prior to insertion into the chambers ensures that a stable paint film has been formed that does not change emission during the chamber test, as has been observed previously (Clausen et al., 1991). It probably would have been better to precondition the samples under conditions identical to the test conditions. The estimated initial masses in Table 3 would then have been comparable.

Application of the Chamber Tests and Results

The method described can be used for comparison of the emission from different paints as previously described (Clausen et al. 1991). It may not

be used for prediction of the emission in the indoor environment for at least two reasons. First, the air exchange rate and thereby the air velocity used is constant and low compared to the indoor environment and we do not know the relation between air velocity and the first order rate constant of the evaporation-controlled emission. (Also, at higher air velocities the emission may change to diffusion control.) Secondly, we do not know the effect of application on porous substrates e.g. wall paper. If the thickness of a paint film applied to a porous substrate influences the emission significantly, it may be recommended indoors to use waterborne paints with strong film and high covering power. Thus only one or two thin layers of the paint need to be applied. This reduces the initial amount of emitting compounds and increases the emission rates, reducing the long-term emission of (S)VOCs.

Conclusion

It has been demonstrated, in theory and practice, that the thickness of waterborne paint films may have a strong influence upon the emission rates of (S)VOCs emitted from the waterborne paint films. Thus the thicknesses of paint films used in climatic chamber tests to estimate emission rates for product comparison or emission prediction must be known in order to prevent erroneous conclusions. It has been demonstrated, also, that an intended variation of the film thickness (if it is large) may be used to determine whether the emission is controlled by evaporation or internal diffusion.

The results show that it may be very difficult to reproduce results of chamber tests. Thus it is important to know the reproducibility of a given chamber test before it is used for comparison or prediction.

It is important that the sample preparation and test conditions are fitted to the properties of the model. In addition, it is recommended that sufficient data be sampled to estimate sink effects when the emission contains SVOCs which strongly adsorb to surfaces.

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