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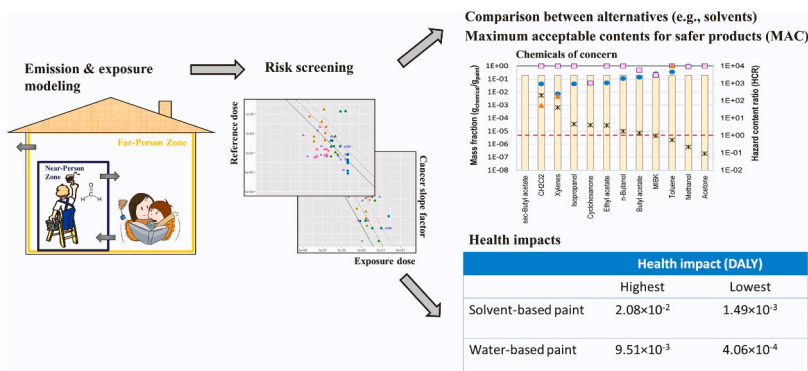
Near-field exposures and human health impacts for organic chemicals in interior paints: A high-throughput screening

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HIGHLIGHTS

- High-throughput (HT) approach to assess organic chemicals in interior paints.
- Mass-balance models predict near-field exposures during wet and dry phases.
- Screening of 65 chemicals in water- and 26 chemicals in 12 solvents-based paints.
- Several biocides identified as Chemicals of concern in generally safer water paints.
- Estimated Maximum content MACs and human health impacts of formulations in DALYs.

GRAPHICAL ABSTRACT



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ABSTRACT

Interior paints contain organic chemicals that might be harmful to painters and building residents. This study aims to develop a high-throughput approach to screen near-field human exposures and health impacts related to organic chemicals in interior paints. We developed mass balance models for both water- and solvent-based paints, predicting emissions during wet and dry phases. We then screened exposures and risks, focusing on Sri Lanka where residential houses are frequently repainted. These models accurately predict paint drying time and indoor air concentrations of organic chemicals. Exposures of both painter and household resident were estimated for 65 organic chemicals in water-based and 26 in solvent-based paints, considering 12 solvents. Chemicals of concerns (CoCs) were identified, and maximum acceptable chemical contents (MACs) were calculated. Water-based paints generally pose lower health risks than solvent-based paints but might contain biocides of high concern. The total human health impact of one painting event on all household adults ranges from 1.5×10^{-3} to 2.1×10^{-2} DALYs for solvent-based paints, and from 4.1×10^{-4} to 9.5×10^{-3} DALYs for water-based paints. The

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present approach is a promising way to support the formulation of safer paint, and is integrated in the USEtox scientific consensus model for use in life cycle assessment, chemical substitution and risk screening.

1. Introduction

Interior paints are commonly used to protect, color or provide texture to objects such as indoor walls. It is recommended that the interior of homes is repainted every 5–7 years [1]. Application and use of interior paints include two phases: the wet phase during which the paint is freshly applied to a substrate and dries as the solvents or water evaporate, and the dry phase during which the dried paint serves as a solid film on top of the substrate. While a household resident's exposure to wet painting is once every few years, a professional painter is exposed to painting processes on a near daily basis. In addition, in certain areas the interior of homes needs to be repainted more frequently due to weather conditions, such as Sri Lanka where homes are repainted every year, leading to higher exposure of the residents to wet paintings. After the paint is dried, additional masses of volatile chemicals can slowly off-gas or leach out from the dried paint or come off to form dust particles, further exposing the residents.

Paints contain a number of chemicals that are known for causing negative health effects in humans, both inorganic and organic. Lead-based paint has been a major concern historically and in developing countries [2], but it is commonly found in older homes as most developed countries banned decorative lead-based paint over 40 years ago [3]. In recent decades, a class of chemicals in paints has been of particular concern, which is known as volatile organic compounds, or VOCs. VOCs are especially prevalent in solvent-based paints, also known as alkyd paints [4], which are mainly found in the solvent portion of the paint, but are also found in pigments and binders [5]. Some common VOCs found in interior paints are benzene, toluene, xylene, naphtha, formaldehyde [4] and other chemical families like alkylphenol ethoxylates (APEOs) [6]. VOCs can lead to various adverse health effects, including respiratory irritation and sensitization, damage to liver, kidney and central nervous system, and cancer [7]. Due to VOCs' health concerns, solvent-based paints are more and more replaced by e.g. water-based paints characterized by low VOC emissions [8]. Besides lead and VOCs, although a limited number of studies have reported exposures to preservatives in paint (e.g., phenylmercuric acetate, isothiazolinones) in interior paints [9,10], other interior paints constituents such as binders and colorants are less studied, and their related human exposures and health risks are largely unknown.

To assess the emission of hazardous chemicals from interior paints, various chamber test studies have been conducted to measure their emission rates, especially for VOCs [11–13]. Several mathematical models have also been developed to estimate the VOC emissions from paints, as a more cost effective and faster alternative to chamber test experiments. For example, the ConsExpo Web developed by RIVM (the Dutch National Institute for Public Health and the Environment) employs an evaporation model to estimate the release of chemical from wet painting, which includes two coupled differential mass balance equations that need to be solved together [14]. However, the ConsExpo model only addresses the wet phase of interior paints, considers the air is instantaneously well-mixed in the painted room, and the equations need to be solved numerically due to increasing mass of wet paint during the painting process. The Consumer Exposure Model (CEM) developed by U.S.EPA uses a double exponential model for latex paint which considers both the wet phase (fast release governed by evaporation) and the dry phase (slower release dominated by diffusion), but it empirically assumes that at maximum only 25% of the applied chemical mass would be released, and it requires empirical estimation of certain rate constants [15]. The Wall Paints Exposure Assessment Model (WPEM) developed by U.S.EPA uses an incremental source model for paints, which assumes a constant application rate over time coupled with an emission rate for

each instantaneously applied segment that declines exponentially. However, it also only considers the wet phase and assumes well-mixing air in the room [16]. Li et al. developed a physically-based model which considers both the evaporation of VOCs from wet paints and the diffusion of VOCs into the painted substrate, but it also considers well-mixed air and requires numerical solutions [17]. Overall, the existing models/tools for predicting chemical emission from paints cannot differentiate the exposure for the painter and for the resident (i.e., assuming air is well-mixed), and are not suitable for efficiently screening dozens if not hundreds of chemicals (i.e., high-throughput screening) due to their complexity.

To address the needs of differentiating resident and painter exposures as well as efficiently assessing many chemicals, the present study aims to propose a high-throughput screening (HTS) approach for estimating emissions and related exposures and impacts during both wet and dry phases for a wide range of chemicals in interior paints, to identify chemicals of concern (CoCs) and inform risk reduction efforts. The study targets Sri Lanka where residential houses are frequently repainted. We focus on organic chemicals, since current modeling approaches on indoor emission, transport and fate are only valid for organics [18–20]. To achieve these aims, we focus on the following specific objectives:

- (1) Characterize the chemical composition in residential interior paints, including water-based and solvent-based paints;
- (2) Develop a mass balance-based, high-throughput suited model for predicting the chemical emissions from interior paints during both the wet phase and the dry phase;
- (3) Estimate multi-pathway near-field human exposures and related human health risks for organic chemicals present in interior paints, considering differences between the painter and the residents;
- (4) Screen and prioritize human health risks to identify chemicals of concern and maximum acceptable contents for chemicals in paints.
- (5) Calculate the near-field human health impacts resulting from organic chemicals in paints, and compare between different compositions of paints.

This study combines exposure estimates with available toxicity data and high-throughput toxicity estimates to inform decision makers and paint manufacturers of the potential human health risks and impacts for the various chemical constituents in paints. It determines risks besides commonly studied VOCs, pinpoints priority chemicals for future development of safer paint products, and educates the consumers and professional painters of the best practices for reducing exposures. The model for interior paints presented here has been integrated in USEtox 3 as a module to assess consumer exposures (both painters and residents) during the use phase of paints, and therefore can be consistently integrated with emission-based exposures along the rest of the paint product life cycle for use in life cycle impact assessment (LCIA), absolute environmental sustainability assessment (AESA), chemical substitution and chemical alternatives assessment (CAA), and risk screening [52–56].

2. Methods

2.1. Overall assessment framework

We employ a high-throughput quantitative source-fate-exposure-effect assessment approach that builds on and is fully compatible with the Product Intake Fraction (PiF) framework [18,21] and with the

USEtox far-field model to assess the organic chemicals in interior paints. This approach has been successfully applied to chemicals in plastic toys and building materials and has been described in detail previously [22, 23]. The approach is illustrated in Fig. 1.

Briefly, we first quantify the mass of each chemical used in one application of interior paint, by multiplying the amount of applied paint and the mass fraction of chemical in the paint. Then we use multimedia models to estimate the emission of chemicals from interior paints during the wet and dry phases, the multi-pathway chemical transport and fate among compartments in the near- and far-field environments, and finally human exposures. The models result in exposure pathway-specific product intake fractions (PiFs), which is defined as the chemical mass taken in by exposed humans over a given exposure period per unit mass of chemical in a product [21]. The PiFs are then aggregated by exposure routes such as inhalation, ingestion and dermal. The route-specific aggregate PiFs are then multiplied by the chemical mass in the paint to calculate the human intakes in kg. The human intakes can be further divided by the number of exposed humans, the human body weight and the exposure duration to obtain the daily exposure dose expressed in mg/kg/d.

To calculate human health impacts, the estimated intakes are multiplied by human health effect factors differentiating between cancer, noncancer general and noncancer reproductive/developmental effects, which are then multiplied by effect-specific severity factors and summed across effects to obtain the human health impacts in disability-adjusted life-years (DALYs) (Eq. 1).

$$I = \sum_e \left(\sum_x (PiF_x \times mf_p \times m_p \times EF_{e,x}) \times SF_e \right) \quad (1)$$

where I is the human health impact (DALY), PiF_x is the product intake fraction via exposure route x (-), mf_p is the chemical's mass fraction in the paint product ($\text{mg}_{\text{chemical}}/\text{mg}_{\text{paint}}$), m_p is the mass of paint product

used (kg), $EF_{e,x}$ is the effect factor for effect e and exposure route x ($\text{cases}/\text{kg}_{\text{intake}}$), and SF_e is the severity factor of effect e (DALY/case).

To characterize health risks, for cancer effects, the daily exposure dose is multiplied by a route-specific cancer slope factor (CSF) to calculate a lifetime cancer risk (Eq. 2a). This risk probability can then be compared to the defined acceptable lifetime cancer risk of 10^{-5} for the general population and to the 10^{-4} for workers [24], depending on the jurisdiction. For non-cancer effects, risks are characterized by dividing the route-specific exposure dose by a route-and-effect-specific reference dose (RfD) to yield a hazard quotient (HQ) (Eq. 2b); then a hazard index (HI) is calculated by summing the HQ across the different routes (Eq. 2c). A HQ (or HI) > 1 would indicate potentially harmful chemicals that require further scrutiny. The sources for the toxicity data are described in Section 2.4.

$$ILCR = \sum_x (D_x \times CSF_x) \quad (2a)$$

$$HQ_{e,x} = D_x / RfD_{e,x} \quad (2b)$$

$$HI_e = \sum_x HQ_x \quad (2c)$$

where $ILCR$ (probability) is the incremental lifetime cancer risk, D_x is the daily exposure dose via route x ($\text{mg}/\text{kg}_{\text{BW}}/\text{d}$), CSF_x is the cancer slope factor via route x (incidence risk/ $\text{mg}/\text{kg}_{\text{BW}}/\text{d}$), $RfD_{e,x}$ is the reference dose for effect e (noncancer general “g” or noncancer reproductive/developmental “rd”) and exposure route x ($\text{mg}/\text{kg}_{\text{BW}}/\text{d}$).

Finally, we estimate the maximum acceptable contents based on our high-throughput screening results (MAC_{HTS}) for the studied chemicals in interior paints as the content of chemical mass in the paint which results in a cancer risk of 10^{-6} and a hazard index of 1, respectively. The minimum MAC between cancer and non-cancer effects is taken as the final

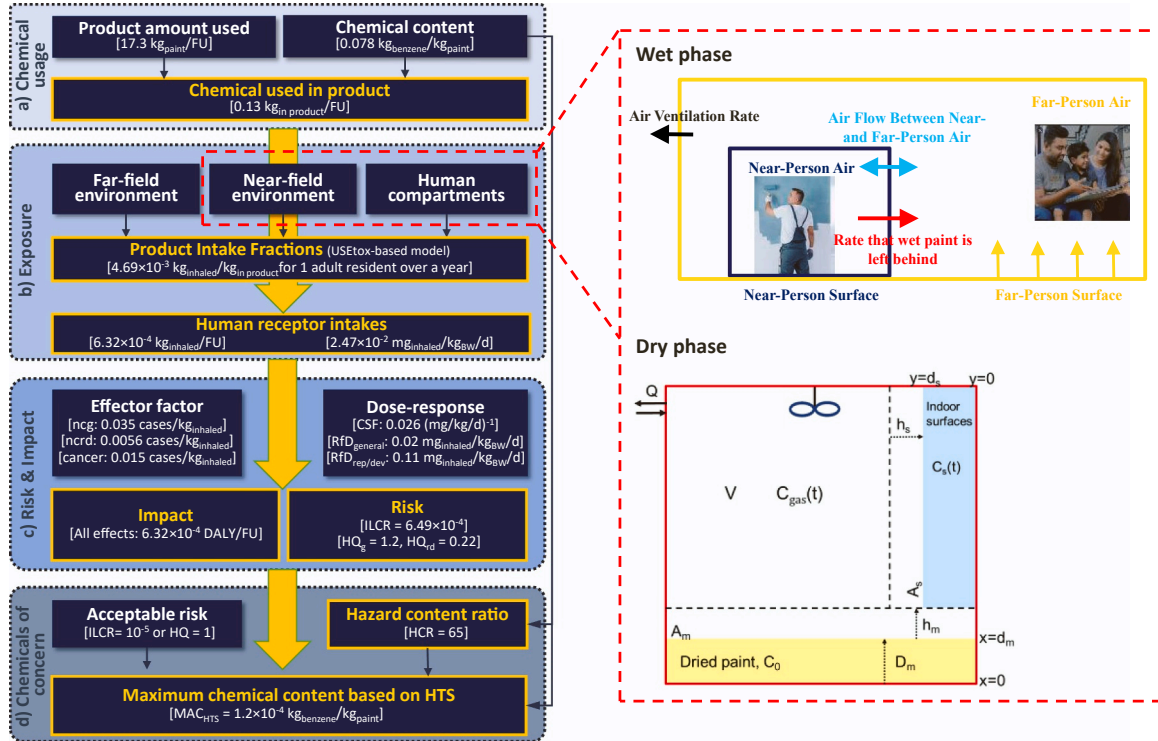


Fig. 1. Right: schematic of the two-box model for the application of interior paints (wet phase) and the diffusion-partition model for the dry phase. ncg: noncancer general effects, ncrd: noncancer reproductive or developmental effects, CSF: cancer slope factor, RfD: reference dose, ILCR: incremental lifetime cancer risk, HQ: hazard quotient, DALY: disability-adjusted life-years, HCR: hazard content ratio, MAC: maximum chemical content, HTS: high-throughput screening.

Left: diagram of the assessment framework, from mass in product to health risks and impacts, illustrated with the example of benzene in solvent-based paint with xylenes as the solvent, with the functional unit (FU) defined as a house with functional interior paints for 1 year, adapted from Huang et al., 2022 [23].

MAC, and we calculate a hazard content ratio (HCR) as the chemical's actual mass fraction in paints divided by the final MAC. (Eq. 3)

$$HCR = \max\left(\frac{HI_{ref}}{HI_{neg}}, \frac{HI_{ref}}{HI_{ncrd}}, \frac{ILCR_{ref}}{ILCR}\right) \quad (3a)$$

$$MAC_{HTS, final} = \min\left(\frac{mf_p}{HCR}, 1\right) \quad (3b)$$

where HI_{ref} is the reference hazard index which is 1, $ILCR_{ref}$ is the reference lifetime cancer risk which is 10^{-5} .

2.2. Interior paints: chemical composition and usage

The typical formulations of water-based and solvent-based interior paints were provided by experts from the Sri Lanka paint industry and are presented in Table 1. For each paint function such as binder, colorant or biocide, the experts also provided a list of potential chemicals that are

Table 1

Typical formulation of interior paints. The numbers are mass fractions presented as percentages.

	B/ White	Lightshade Colors	Mid Dark Shade colors	Very Dark Shade colors
Water-based Emulsion paint (low sheen finish)	% (w/w)	% (w/w)	% (w/w)	% (w/w)
Water	26.00	26.00	26.00	26.00
Pigment wetting agent	0.45	0.45	0.55	0.65
Pigment dispersing agent	0.25	0.25	0.30	0.40
In-can biocide (wet film biocide)	0.25	0.25	0.25	0.25
Ammonia	0.30	0.30	0.30	0.30
Cellulosic thickener	0.28	0.33	0.33	0.45
Film forming agent	1.70	1.35	1.55	1.55
Mono Ethylene glycol / mono propylene glycol	1.75	1.75	1.10	1.10
Anti foaming agent	0.38	0.38	0.38	0.38
Hydrous kaolin or Flash calcined kaolin	6.75	5.25	0	0
Calcined kaolin	6.75	6.75	3.50	0
Precipitated calcium carbonate	11.00	11.00	9.00	2.50
Calcium magnesium carbonate (dolomite)	1.75	0.50	0	0
Magnesium silicate (Talc)	2.50	2.50	0.50	7.00
Hydrous Aluminium silicate	3.50	3.50	9.00	27.50
Titanium Dioxide (Rutile grade)	19.75	11.00	4.50	0
Opaque polymer (holo beads)	8.00	6.50	4.25	0
Water based binder	17.50	12.50	19.00	30.00
prepared pigment paste	0	3.50	7.00	13.50
Solvent-based Enamel paint				
Alkyd Resin (Binder)	66.50	66.50	72.50	77.50
Titanium Dioxide (white pigment)	13.75	6.00	2.00	0
Colored pigments (inorganic)	0	1.00	6.50	8.50
Colored pigments (organic)	0	2.00	4.00	4.00
Magnesium silicate (Talc)	6.00	6.00	6.00	6.00
Fumed silica	1.00	1.00	1.00	1.00
Surface drier	0.60	0.60	0.60	0.60
auxiliary drier	0.24	0.24	0.24	0.24
Hard drier	0.50	0.50	0.50	0.50
Anti Skinning agent	0.23	0.23	0.23	0.23
Solvent	19.50	19.50	19.50	19.50

commonly used in Sri Lanka and the typical chemical mass fractions in the corresponding paint component. We then multiplied these chemical-component specific fractions by the mass fraction of the paint component itself to obtain the mass fraction of each chemical in the final paint formulation. For example, a water-based paint with very dark shade color contains 30% water-based binder such as styrene acrylic copolymer emulsion, which may contain up to 0.05% styrene monomer, so this water-based paint would contain 0.015% styrene monomer.

Since this study only focuses on neutral organic chemicals due to the applicability of USEtox models, we excluded the chemicals that are inorganic, organometallic, salts, polymers, mixtures and UVCBs (chemical substances of unknown or variable composition, complex reaction products and biological materials). The final dataset includes 65 unique chemicals in water-based paints. For solvent-based paints, there are 26 unique chemicals excluding solvents. The solvents used in paints are generally mixtures of different organic chemicals. Since there are diverse solvent compositions, for simplicity we consider the solvent to be pure solvent for modeling purposes. The following twelve organic solvents are studied: methanol, toluene, 2-propanol, n-butyl acetate, secondary butyl acetate, xylenes, acetone, methyl isobutyl ketone (MIBK), dichloromethane (DCM), cyclohexanone, ethyl acetate, and 1-butanol. Physiochemical properties of the studied chemicals and solvents are provided in Appendix B.

For interior paints usage, we assumed that the inner walls of a residential house in Sri Lanka are repainted every year, considering two usage scenarios. The first is for the painter considering exposures and risk of a daily paint application of 4.77 kg of paint on a 42 m² wall area painted in 5.6 h, assuming 2 h needed for painting an area of 15 m² [25]. We assumed the painter works for 200 working days per year and for 40 years per lifetime of 70 years. The second scenario is for the household residents, who are assumed to be present in the house during paint application and then reside in the house for the rest of the year, which is a conservative assumption but applies to DIY projects where people paint their own houses every year. Thus, we considered the exposure and risk over one year exposure, applying 17.3 kg of paint on 152 m² wall area at the beginning of the year. The year refers to a full year between two painting events.

2.3. Estimating exposures to chemicals in interior paints

Application and use of interior paints include two phases: the wet phase during which the paint is freshly applied to a substrate and dries as the solvents evaporate, and the dry phase during which the dried paint serves as a solid film on top of the substrate. Since the wet phase and dry phase are governed by different physical processes, we used two different sub-models to estimate the chemical emission and human exposure during each of these phases. The aim of these sub-models is to first estimate the direct transfer fractions of chemicals from the compartment of entry (i.e., where the chemical enters the near-field environment) to other compartments such as indoor air and human epidermis [18], to then determine indoor and outdoor multi-pathway exposures.

2.3.1. Model for chemical emission and exposure during wet phase

Four-compartment indoor model and its integration into the USEtox far-field model.

During the wet phase, the freshly applied paint acts like a liquid, so the chemical emission from the paint is mainly governed by evaporation. The painter applying the paint is assumed to have a higher exposure than the other household members, because the chemicals in the paint will first evaporate to the air that is close to the painter resulting in relatively higher concentrations in the near-person air, and then be distributed in the air of the entire house resulting in lower concentrations. To model this process, we start from a two-box mass balance model which divides the house into a near-person zone and a far-person zone. The two zones are further divided into the paint product surface (s)

and the air (a) yielding four indoor compartments: near-person surface, near-person air, far-person surface and far-person air, as illustrated in Fig. 1. The paint is first applied on the near-person surface compartment, and then evaporates to the near-person air or is taken up by direct dermal contact to the skin of the painter; the near-person air exchanges with the far-person air; as the painter moves to apply the paint to another area, the chemical in the already painted near-person surface is transferred to the far-person surface.

In USEtox 3.0, transfers between these compartments and the outdoor compartments are characterized by determining direct transfer fractions of each compartment to the neighboring compartments over the entire drying period, summarized in a matrix of direct transfer fractions ($\mathbf{TF} \in \mathbb{R}^{c \times c}$) integrating the four indoor compartments with 11 outdoor compartments plus two waste water and solid waste treatment compartments. These transfer fractions from compartment i to compartment j are themselves determined as the transfer rate constant between these compartments ($k_{j \leftarrow i}$, $1/s$) divided by the total losses from the source compartment ($k_{i, \text{total}}$, $1/s$): $TF_{j \leftarrow i} = k_{j \leftarrow i} / k_{i, \text{total}}$.

The cumulative transfer fractions matrix $\mathbf{TF}^{\text{cum}} = (\mathbf{I} - \mathbf{T})^{-1}$ is finally determined by inverting the direct transfer fractions matrix, accounting for all multi-media transfer and feedback between compartment [26].

Focusing on the indoor transfer between the 4 indoor compartments and the outdoor air compartment, the different transfer rate constants are presented below in the wet paint sub-matrix relevant to this wet paint model and the calculation of the rate constants given in Appendix A, Section A1.1. These transfer rate constants are then used to calculate the transfer fractions between these compartments as described below, which are then integrated into the full near-field/far-field \mathbf{TF} matrix in USEtox.

$$\mathbf{K}_{\text{wet paint}} = \begin{bmatrix} -k_{ns, \text{total}} & k_{na \rightarrow ns} & 0 & 0 & 0 \\ k_{ns \rightarrow na} & -k_{na, \text{total}} & 0 & k_{fa \rightarrow na} & 0 \\ k_{ns \rightarrow fs} & 0 & -k_{fs, \text{total}} & k_{fa \rightarrow fs} & 0 \\ 0 & k_{na \rightarrow fa} & k_{fs \rightarrow fa} & -k_{fa, \text{total}} & k_{oa \rightarrow fa} \\ 0 & 0 & 0 & k_{fa \rightarrow oa} & 0 \end{bmatrix}$$

Subscript ns: near-person surface, na: near-person air, fs: far-person surface, fa: far-person air, oa: outdoor air.

Net transfer fractions from the surfaces and remaining fraction for the dry phase.

Since the kinetic of these exchanges is primarily limited by the volatilization at the surface and the air exchange to the outside, we can assume that the indoor air is at quasi-steady-state between the surfaces of the room and the outside air. Section A1.2 of Appendix A shows how we can directly calculate net transfer rate constants from the surfaces.

$k_{net, ns \rightarrow na} = k_{ns \rightarrow na} \cdot (1 - k_{na \rightarrow ns} / (k_{na \rightarrow ns} + \text{AER}))$ and $k_{net, fs \rightarrow fa} = k_{fs \rightarrow fa} \cdot (1 - k_{fa \rightarrow fs} / (k_{fa \rightarrow fs} + \text{AER}))$, accounting for the feedback from the air back to the surface.

The mass balance equation for near-person surface accounting for feedback reads:

$$\begin{aligned} \frac{dm_{ns}(t)}{dt} &= -k_{net, ns, \text{tot}} \cdot m_{ns}(t) \\ &= -(k_{net, ns \rightarrow na} + k_{ns \rightarrow fs} + k_{deg, ns} + k_{ns \rightarrow skin}) \cdot m_{ns}(t) \end{aligned} \quad (4)$$

The direct transfer fractions from near-person surface to near-person air, and human skin during the wet phase are thus calculated as follows:

$$TF_{net, ns \rightarrow na} = \frac{k_{ns \rightarrow na}}{k_{net, ns, \text{tot}}} \cdot (1 - e^{-k_{net, ns, \text{tot}} \cdot t_{\text{drying}}}) \quad (5a)$$

$$TF_{net, ns \rightarrow skin} = \frac{k_{ns \rightarrow skin}}{k_{net, ns, \text{tot}}} \cdot (1 - e^{-k_{net, ns, \text{tot}} \cdot t_{\text{drying}}}) \quad (5b)$$

where t_{drying} is the duration that the applied wet paint needs to dry (s), which is described in Appendix A, Section A1.4.

In addition, a fraction of the chemical applied on the near-person

surface is transferred to the far-person surface, from which a sub-fraction will be volatilized to far-person air, and the transfer fraction from near-person surface to far-person air (via far-person surface) is given by:

$$\begin{aligned} TF_{net, ns \rightarrow fa} &= TF_{net, ns \rightarrow fs} \cdot TF_{net, fs \rightarrow fa} \\ &= \frac{k_{ns \rightarrow fs}}{k_{net, ns, \text{tot}}} \cdot (1 - e^{-k_{net, ns, \text{tot}} \cdot t_{\text{drying}}}) \cdot (1 - e^{-k_{net, fs \rightarrow fa} \cdot t_{\text{drying}}}) \end{aligned} \quad (5c)$$

The mass fraction remaining on near-person surface and far-person surface when the paint is dry will be used as the initial mass for the dry phase modeling and is given by:

$$\begin{aligned} TF_{s, \text{remain}} &= e^{-k_{net, ns, \text{tot}} \cdot t_{\text{drying}}} + TF_{net, ns \rightarrow fs} \cdot e^{-k_{net, fs \rightarrow fa} \cdot t_{\text{drying}}} \\ &= e^{-k_{net, ns, \text{tot}} \cdot t_{\text{drying}}} + \frac{k_{ns \rightarrow fs}}{k_{net, ns, \text{tot}}} \cdot (1 - e^{-k_{net, ns, \text{tot}} \cdot t_{\text{drying}}}) \cdot e^{-k_{net, fs \rightarrow fa} \cdot t_{\text{drying}}} \end{aligned} \quad (5d)$$

2.3.2. Model for chemical emission and exposure during dry phase

After the paint is dried, the chemicals remained on the surface are embedded in a solid film of paint. These chemicals can also be slowly released to the indoor air. We assume this release process is the same as for chemicals encapsulated in solid products, such as plasticizers in vinyl flooring. Since the painting process is terminated, emissions are assumed to be occurring in the far person air. Thus, we use the previously developed high-throughput suited model to estimate this chemical emission during the dry phase, called the “combined D- and K-limited model with sorption” [27,28]. Briefly, the chemical emission is assumed to be controlled by the chemical’s internal diffusion inside the solid material and its partition between the solid material and air. Chemicals in paints are classified as two types: diffusion limited (D-limited) and partition limited (K-limited), based on defined criteria in Eq. 6:

$$\begin{cases} \text{D-limited} : K_{ma} < 0.4 \cdot D_m^{-0.61} \\ \text{K-limited} : K_{ma} \geq 0.4 \cdot D_m^{-0.61} \end{cases} \quad (6)$$

where D_m is the chemical’s diffusion coefficient in the dried paint (m^2/s), and K_{ma} is the chemical’s partition coefficient between dried paint and air (unitless).

For both D-limited and K-limited cases, the chemical mass fraction emitted from dried paint to indoor air from time zero to time t ($TF_{p \rightarrow a, DP}$) can be expressed in the form of two exponentials:

$$TF_{p \rightarrow a, DP} = \frac{m_e(t)}{m_0} = a_1 \cdot e^{b_1 t} + a_2 \cdot e^{b_2 t} + a_0 \quad (7)$$

where $m_e(t)$ is the total chemical mass emitted to air from time zero to time t (μg), m_0 is the chemical mass remained in the dried paint when the dry phase starts (μg), t is time since the dry phase starts (s), a_1 , a_2 , a_0 , b_1 , b_2 are coefficients that are calculated as functions of convective mass-transfer coefficient h_m (m/s), room ventilation rate Q (m^3/s), area of the paint A_p (m^2), thickness of dried paint $L_{\text{liquid, dried}}$ (m), as well as D_m and K_{ma} .

Finally, the mass fraction of chemical transferred to indoor air during the dry phase, relative to the total initial chemical mass when the paint is being applied, is given by:

$$TF_{p \rightarrow a} = TF_{p \rightarrow a, DP} \cdot TF_{s, \text{remain}} \quad (8)$$

where $TF_{s, \text{remain}}$ is calculated by Eq. 5d.

During the dry phase, the only direct transfer fraction from the compartment of entry is to the indoor air as calculated by Eq. 8. We consider inhalation and dermal gaseous exposures for the building residents, but they occur through the indoor air (equations in Appendix A, Section A2.3). There is no direct transfer from the dried paint to human skin since the paint is applied on interior walls and we assume no dermal contact with the walls.

2.3.3. Model parameterization

In this study, we model an average residential house in non-OECD countries with a volume of 117 m³, which has two adults and one 2–3 years old child as residents. In the case of a DIY painting project, one of the two adult residents is the person applying the paint. The far-person space was considered as a single space. The duration of the wet phase is determined by the drying time as described in Appendix A, Section A1.4, and the duration of the dry phase is 1 year since in Sri Lanka the house interior is traditionally repainted every Spring. We model two air ventilation rates: 0.79 h⁻¹ for a closed building and 15.6 h⁻¹ for a naturally ventilated building with open windows. All input parameters used to parameterize the models are described in Appendix A, Section A0.

2.4. Toxicity data for risk characterization

Details of toxicity data are presented in Appendix A, Section A3. Briefly, cancer slope factors (CSFs) for adults are taken from USEtox 2.12 (<http://www.usetox.org>) and are based on the Carcinogenic Potency database (CPBD), applying an age-dependent adjustment factor (ADAF = 4) to the CSF for children [29].

For characterizing the non-cancer effects, the ingestion reference doses (RfD) and inhalation reference concentrations (RfC) are obtained from a published database providing peer-reviewed toxicity values reported in various regulatory sources [30,31]. For chemicals for which RfDs or RfCs were not available, we used the probabilistic RfDs and RfCs derived by Aurisano et al. [32,33] from experimental animal data using the WHO/IPCS framework for dose-response assessment [34] [35]. The collected RfDs and RfCs differentiate between general non-cancer and reproductive/developmental effects to account for around a factor 20 difference in severity affecting human lifetime loss [26,36]. Finally, for the substances without RfDs or RfCs available in the above sources, we designated them as “N/A”. Note that for dermal CSFs and RfDs we applied route-to-route extrapolation from oral exposure.

The human health effect factors (EFs) for cancer effects are also taken from USEtox 2.12 (<http://www.usetox.org>) and are derived from a lifetime effect dose inducing cancer in 50% of population via route *x* ($ED50_{x,lifetime}$, in kg_{intake}/lifetime). The EFs for noncancer effects are derived from an effect dose inducing non-cancer disease in 10% of population ($ED10_x$, in mg/kg/d), and there are separate ED10s for noncancer general effects and developmental/reproductive effects [32, 33].

3. Results

3.1. Models evaluation

To evaluate the proposed models, we estimated the drying time (or evaporation rates) for water and several organic solvents, and compared our results with values reported in the literature. The model predicts well the overall trend of the water evaporation process, although it slightly overestimates the evaporation rate at the beginning and underestimates the evaporation rate at the end, due to the assumption of constant liquid thickness. For 14 organic solvents, the model also accurately predicts the evaporation rates relative to butyl acetate, within a factor of 2 of the reported values [37]. Next, we compared our model predictions to measured VOC emissions from paints in chamber studies. The results show that our model can accurately predict the emissions of methyl ethyl ketoxime (MEKO) and decane from wet alkyd paints, with predicted chamber air concentrations within a factor of 2 of the observed values [11,12]. Detailed evaluation results are presented in Appendix A, Section A4. Overall, the evaluation demonstrates that our proposed models are suitable for estimating chemical emissions from interior paints during the wet phase. The models for the dry phase have been published previously and evaluated against various dataset for chemicals in building materials [27,28].

We have also compared our modeled indoor air concentrations of

VOCs with measured concentrations in Sri Lanka. The modeled concentrations of formaldehyde, xylenes and benzene are within reasonable range of the measured concentrations (Appendix A, Section A4.3), again demonstrating the suitability of our models.

3.2. Chemicals exposures and risks

Fig. 2 presents inhalation exposures as a function of toxicity levels (RfDs in inversed scale for non-cancer effects, and cancer slope factors for cancer effects) for chemicals in water-based (triangles) and solvent-based paints (circles). As the dominant exposure route, inhalation doses are presented for both the household adult resident (Fig. 2A–C) and the painter (Fig. 2D–F), differentiating various chemical functions by colors. Dermal exposure doses and risks are presented in Appendix A, Section A5. The diagonal lines in each plot represent equi-hazard quotient (HQ) or equi-incremental lifetime cancer risk (ILCR), from lowest (bottom-left corner of the graph) to highest risks (upper-right corner). Inhalation HQs are available for 23 out of the 65 unique chemicals in water-based paints, while ILCRs are only available for 10 chemicals.

As a general trend, the painter's inhalation exposure doses are 1–2 orders of magnitude higher than those for the household adult for most chemicals, which is reasonable since the painter is always exposed to freshly applied paints. Exposures to chemicals in solvent based paint (circles) tend to be higher than chemicals in the water based paint (triangles). Also binders tend to show high exposure doses but relatively low toxicity, whereas biocides in water based paints tend to present higher toxicity levels, but lower exposures.

Solvent-based paints: In Fig. 2, chemicals in solvent-based paints are presented as circles, with inhalation HQs available for 15 out of the 26 unique chemicals in these paints, while ILCRs are only available for 7 chemicals. Fig. 2 presents the results for 12 solvents and for other chemicals using xylenes as the solvent.

For the household adult resident, the inhalation HQ_g (subscript *g* stands for “general non-cancer effects”) is higher than 1 for the general non-cancer effects of 6 unique chemicals (16 data points), with 2 chemicals (10 data points) with HQ_g > 10 and 1 chemical with HQ_g > 100 (Fig. 2D). The chemical with the highest inhalation HQ_g is Pigment yellow 74 (HQ = 20,400). The risks from non-cancer reproductive or developmental effects are much lower than the general non-cancer effects, with only 1 chemical (Formaldehyde) in solvent-based paints (using xylenes as the solvent) having an inhalation HQ_{rd} > 1 (subscript *rd* stands for “reproductive or developmental effects”) (Fig. 2E). In terms of cancer effects (Figs. 2F), 5 of the 7 chemicals (18 data points) with cancer toxicity data available have estimated ILCR > 10⁻⁴ through inhalation, indicating very high concern on cancer effects, even for the household adult.

For the painter, the inhalation HQ_g is higher than 1 for the general non-cancer effects of 10 chemicals (28 data points), with 8 chemicals (20 data points) with HQ_g > 10 and 5 chemicals (15 data points) with HQ_g > 100 (Fig. 2A). The chemical with the highest inhalation HQ_g is xylenes in a solvent-based binder (alkylated urea formaldehyde resin) (HQ_g = 2570). Considering non-cancer reproductive or developmental effects, 7 chemicals (16 data points) have estimated inhalation HQ_{rd} > 1, of which 5 chemical (8 data points) with HQ_{rd} > 10 and 1 chemical (3 data points) with HQ_{rd} > 100 (Fig. 2B). Looking at cancer effects (Fig. 2C), the inhalation ILCR are > 10⁻⁵ for all chemicals in solvent-based paints, except for Pigment Red 3 (cancer risk of 8.9 × 10⁻¹¹) and n-hexane (cancer risk of 4.7 × 10⁻⁶).

Water-based paints:

Compared to solvent-based paints (circles closer to the upper right corner than triangles), resulting risks of chemicals tend to be lower for water-based paints (triangles), except for biocides that are primarily present in water-based paints.

For the household adult resident over an exposure duration of 1 year after painting, the inhalation HQ is higher than 1 for the general non-cancer effects of 10 chemicals in water-based paints, with 5 chemicals

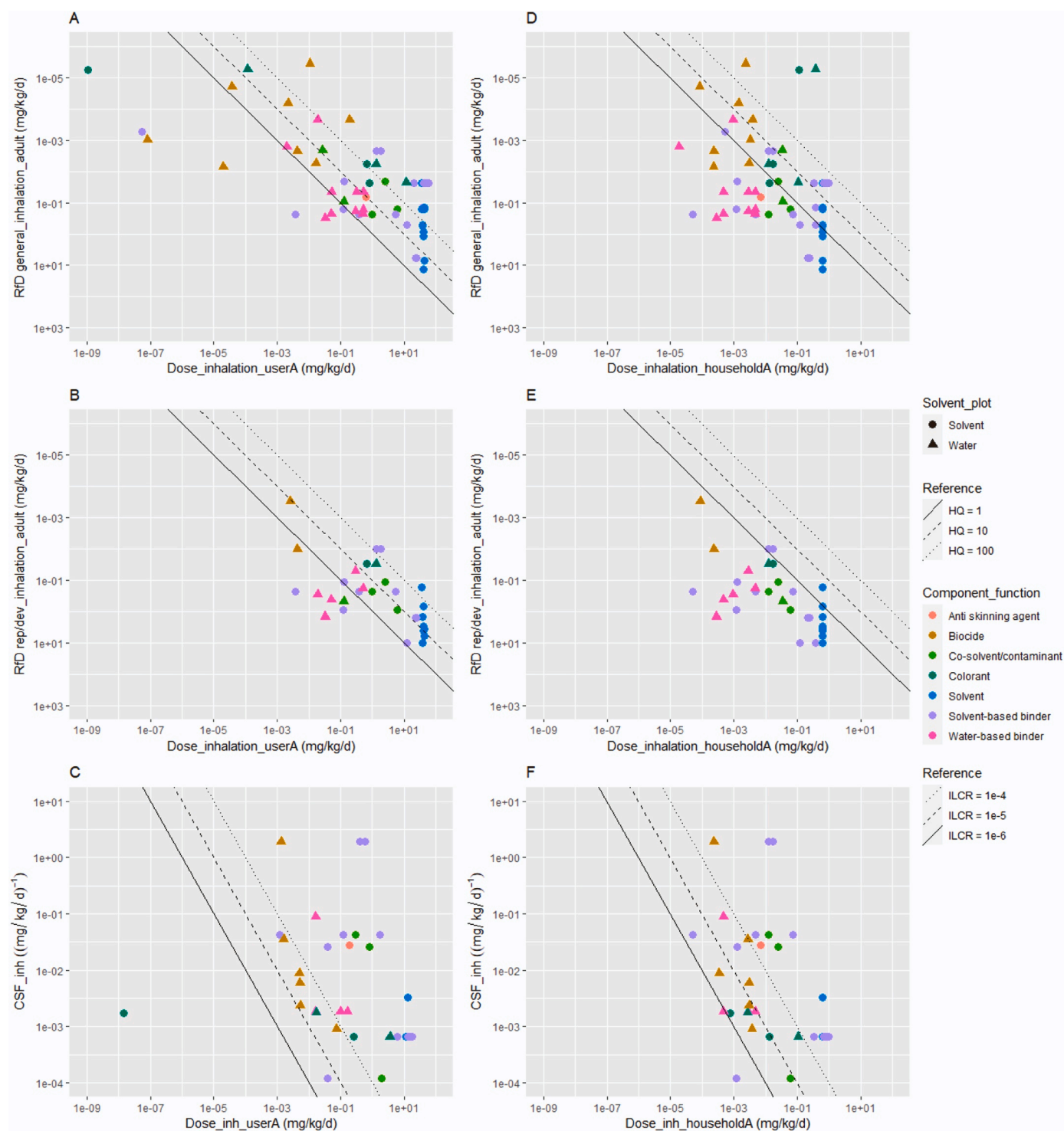


Fig. 2. Reference doses (RfDs) for non-cancer effects (A-B, D-E) and cancer slope factors (CSFs) (C, F) as a function of inhalation exposure doses for chemicals in water-based paint and solvent-based paint (xylenes as the solvent) for the painter (A-C) and the household adult resident (D-F). The exposure duration is 1 year for the household adult resident including and following one painting event and a daily exposure to a 1 day painting event for the painter. HQ: hazard quotient. ILCR: incremental lifetime cancer risk. The ILCR for the painter is adjusted for 200 working days per year and 40 working years per lifetime of 70 years.

with $HQ > 10$ and 2 chemicals with $HQ > 100$ (Fig. 2D). The chemical with the highest inhalation HQ is Pigment yellow 74 ($HQ = 68,800$). The risks associated with the non-cancer reproductive or developmental effects are much lower than the general non-cancer effects, with no chemicals in water-based paints having an inhalation $HQ > 1$ (Fig. 2E).

Looking at cancer effects of water-based paints (Figs. 2F), 5 of the 11 chemicals with cancer toxicity data available have estimated inhalation

ILCR $> 10^{-5}$. Formaldehyde as a biocide in water-based paints even has estimated inhalation cancer risk higher than 10^{-4} , indicating very high concern on cancer effects, even for the household adult resident.

For the painter exposed on a daily basis during the paint application period, doses are a factor 10 to a 100 higher than for the bystander. Inhalation HQ_g is higher than 1 for the general non-cancer effects on the painter of 17 chemicals (20 data points), with 9 chemicals with HQ_g

> 10 and 4 chemicals with $HQ_g > 100$ (Fig. 2A). The chemical with the highest inhalation HQ is 4,5-Dichloro-2-octyl-3(2 H)-isothiazolinone (DCOIT, a biocide, $HQ_g = 3110$). Interestingly, the estimated inhalation HQ of Pigment Yellow 74 is 21 for the painter, which is much lower than the HQ_g for the household adult resident. This is because Pigment Yellow 74 is not a highly volatile chemical, as only 0.35 ppm of its mass would be volatilized to the near-person air during the first day of paint application, leading to relatively low inhalation exposure for the painter.

In terms of non-cancer reproductive or developmental effects, 4 chemicals have estimated inhalation $HQ_{rd} > 1$ for the painter, of which only 1 chemical having an inhalation $HQ_{rd} > 10$ (Dibutylamine, $HQ_{rd} = 45$ (Fig. 2B).

Looking at cancer effects (Fig. 2C), all 10 chemicals in water-based paints have estimated inhalation $ILCR > 10^{-5}$ for the painter, an order of magnitude higher than for the household adult. Formaldehyde has the highest estimated inhalation cancer risk of 2.6×10^{-3} more than 2 orders of magnitude above 10^{-5} .

3.3. Maximum acceptable concentrations and chemicals of concern in paints

Solvent-based paints: Fig. 3 presents the actual chemical mass fractions, cancer and non-cancer MAC_{HTS} s and HCRs for the chemicals in solvent-based paints, using xylenes as the example solvent. The MAC_{HTS} s and HCRs correspond to the household adult resident for an exposure duration of 1 year. A chemical is defined to be of concern if the actual mass fraction exceeds one of the MAC s. This is then reflected in HCRs higher than 1 (black stars higher than the dashed horizontal line – right scale). Details of all identified chemicals of concern (CoCs) with $HCR > 1$ are provided in Appendix A, Section A6. The solvent, co-solvents and anti-skinning agent are clearly problematic in solvent-based paints (Fig. 3A), with all HCR s > 1, all due to cancer effects. Among the solvent-based binder (Fig. 3B), ethylbenzene as a solvent and formaldehyde as a residual free monomer in alkylated melamine urea formaldehyde (AMUF) resin are identified as chemicals of very high concern with HCR s > 100. Formaldehyde is of highest concern with an HCR of 32,00. In our calculations, we assume a content of 0.7% for free

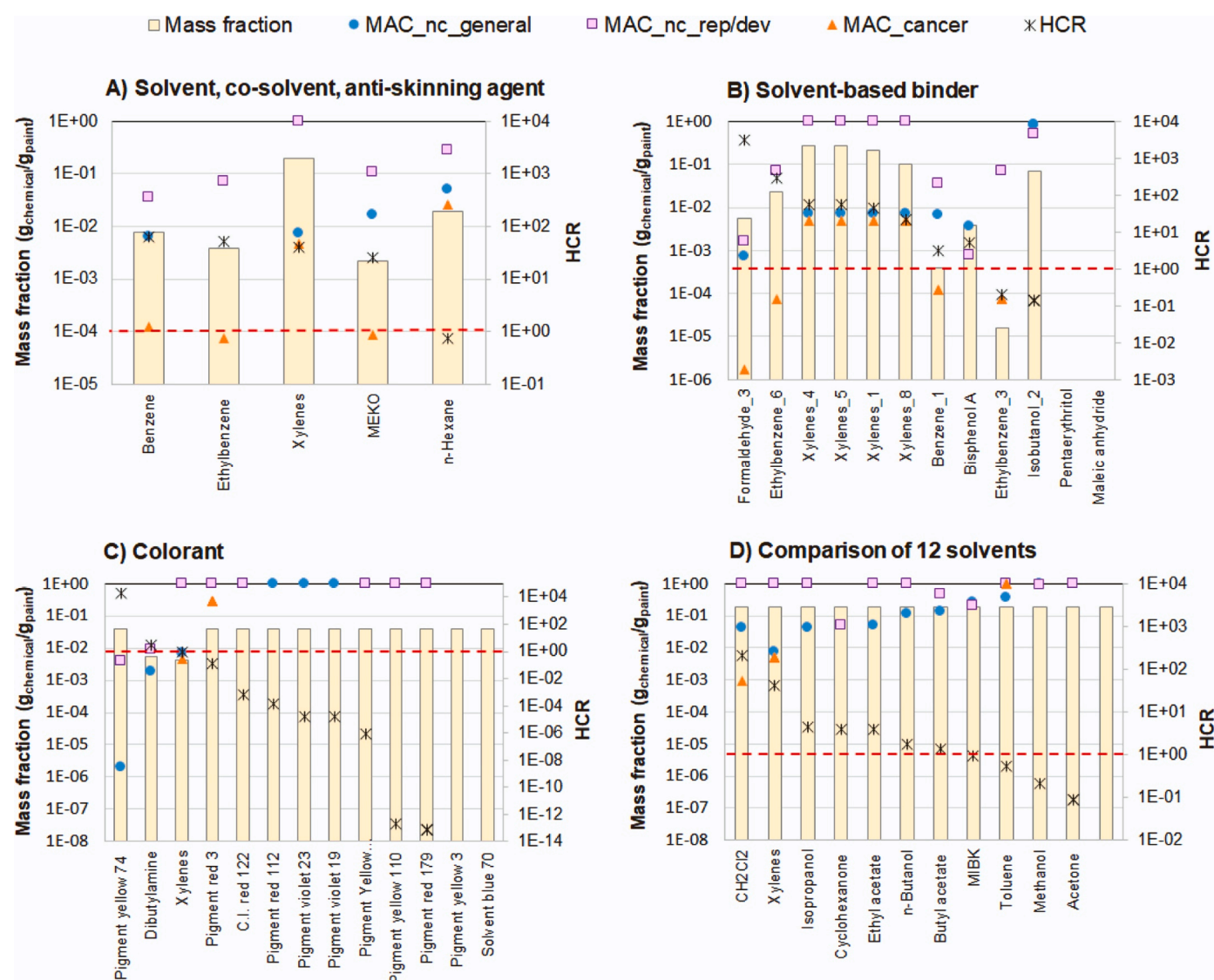


Fig. 3. Actual chemical mass fraction of chemicals in solvent-based paint (yellow bars) compared to maximum chemical content (MAC_{HTS}) for cancer (orange triangles), non-cancer general effects (blue circles) and non-cancer reproductive/developmental effects (purple squares) (left axis), and resulting Hazard Content Ratios (HCR) (black stars - right axis), for (A) solvent, co-solvent and anti-skinning agent, (B) solvent-based binder, (C) colorant, and (D) comparison of 12 solvents. The actual chemical mass fractions in paints with very dark shade colors are shown (Table 1). Results in (A)(B)(C) are calculated for xylene as the solvent. Results are for the household adult resident over 1 year. Chemicals without HCRs are due to lack of toxicity data.

formaldehyde monomer in the AMUF resin, which thus needs to be lower by 32,00 times to avoid unacceptable health risk from the paint to household residents. For colorants, pigment yellow 74 is the only one colorant of high concern identified in solvent-based paints (Fig. 3C) with a very high HCR of 20,400, which may be an overestimation due to issues of physiochemical properties and potential overestimation of inhalation toxicity as discussed below.

When looking at the solvent itself, upon comparison of 12 organic solvents (Fig. 3D) we found that most of the solvents are within the acceptable concentration zone (i.e. $HCR < 10$) when considering an exposure duration of 1 year, except for xylenes and methylene chloride that have much higher impacts. The best performing solvents in terms of human health risk include acetone, methanol, toluene and Methyl iso butyl ketone (MIBK). However, since all organic solvents are VOCs that are emitted rapidly from the paint to the indoor air, the human exposure in the beginning of paint application (or wet phase) would be much higher than the exposure afterwards (or dry phase). If we consider the exposure for the painter with continuous emissions as the first day, all 12 solvents would lead to unacceptable health risk with HCRs much larger than 10, except acetone ($HCR = 6$ for the first day) (Figure A6D in Appendix A). Methylene chloride even leads to an HCR of 4400 for the painter. This large difference in HCR across solvents is solely due to difference in toxicity of the solvent chemicals, because the exposure is the same across all solvents as they would be 100% emitted to indoor air.

Water-based paints: Fig. 4 presents the chemical mass fractions, cancer and non-cancer MAC_{HTS} s and HCRs for the chemicals in the water-based paints, that generally lead to lower HCR, with some notable exceptions. For the co-solvents, propylene glycol is slightly of concern with an HCR of 16 (Fig. 4A). Propylene glycol or ethylene glycol can be used to get freeze/thaw stability and to control the open time of the film in water-based paints. We estimated an HCR of 0.4 for Ethylene glycol and an HCR of 16 for propylene glycol. The higher HCR of propylene glycol is due to an inhalation RfC of 0.01 mg/m^3 which is a probabilistic value [32], while the inhalation RfC of ethylene glycol is 0.4 mg/m^3 which is a regulatory limit. In terms of oral toxicity, the regulatory RfD for propylene glycol is 20 mg/kg/d compared to 2 mg/kg/d for ethylene glycol, indicating that propylene glycol is less toxic than ethylene glycol via ingestion. Since the toxicity trends for propylene glycol and ethylene glycol are different via inhalation and ingestion and are based on different data type, further investigations are needed when comparing the respective risks of these substances.

Most chemicals in the binder component of the water-based paint result in acceptable human health risk, except styrene and acrylic acid (Fig. 4B). Styrene is a residual free monomer in styrene acrylic copolymer emulsion. We assume a content of 0.05% for free styrene monomer in this copolymer which results in an HCR of 4. Acrylic acid is also a residual free monomer in Anucryl 80 with 0.1% content assumed, resulting in an HCR of 4. Thus, to bring the risk down to acceptable levels for a content of 30% binder polymer in the paint, the contents of free styrene monomer and acrylic acid monomer in the binder polymer should be lower than 0.012% and 0.024%, respectively.

For colorants (Fig. 4C), Pigment yellow 74 is identified as of high concern similar to solvent-based paints, with an HCR of 68,800, due to general non-cancer effects. Xylenes and dibutylamine as solvents for colorants are also identified with HCRs of 7 and 2, respectively. The estimated risk for pigment yellow 74 is extremely high, because it is estimated that 100% of this chemical would be emitted to indoor air after 1 year. It seems unlikely that a colorant in the paint would be 100% volatilized after 1 year, which may be due to issues on its physiochemical properties and the use of the acidic based value for the Kow. If the neutral logKow is used for pigment yellow 74, it would have a more plausible value of 1.62% volatilized after 1 year, reducing the HCR for household adult to a still very high value of 500. In addition, pigment yellow 74 has a very low inhalation RfD of $5.5 \times 10^{-6} \text{ mg/kg/d}$, which is 5 orders of magnitude lower than its oral RfD. The inhalation RfD is a probabilistic value as described in Section 2.4, which may be

underestimated.

For biocides (Fig. 4D), about half of the biocide chemicals can be considered acceptable for the household adult, but 7 are identified with $HCR > 10$, of which $HCR > 100$ are found for DCOIT and pentachlorophenol (PCP). For the painter, 10 biocide chemicals are identified with $HCR > 10$, 3 of which with $HCR > 100$; DCOIT is the worst with HCR of 3110 (Appendix A, Section A6). The biocides are needed in water-based paints to prevent the growth of bacteria, which are generally not needed in solvent-based paints. Since water-based paints are generally considered to be less toxic than solvent-based paints, it is important to observe that several biocides in water-based paints may pose a significant health risk to the household residents and also the painter, which may become a disadvantage of water-based paints. The paint manufacturers need to lower the concentrations of these chemicals to be below the MAC_{HTS} s, and if that cannot fulfill the required function, explore alternative biocides that would result in lower human health risk, such as 2-Methyl-2 H-isothiazolin-3-one (MIT), 2-(Thiocyanomethylthio)benzothiazole (TCMTB), Carbendazim, Thiabendazole, and Permethrin, which have estimated HCRs below 1 for both the household adult resident and the painter.

3.4. Human health impacts

As described in Section 2.1, the human health impacts expressed in DALYs are also calculated for each organic chemical in paints. We can thus estimate the total health impact of a paint product by summing the impacts of all chemical components. We calculated the total health impacts of theoretical compositions of water-based paints and solvent-based paints (xylenes as the solvent) for the colorant, binder and biocide with both the highest and lowest impacts. We kept the solvent and co-solvent composition constant across products.

Table 2 presents the estimated total health impact of paints for all adult residents in one household over 1 year following one DIY painting event. Exposures include one adult applying the paint and then residing in the house, as well as another adult residing in the house. The total human health impact of solvent-based paints ranges from 1.5×10^{-3} to 2.1×10^{-2} DALYs, while it ranges from 4.1×10^{-4} to 9.5×10^{-3} DALYs for water-based paints. The total health impact of water-based paint is just 2–4 times lower than that of solvent-based paint. In solvent-based paints, the health impact is mainly due to the solvent/co-solvent and the solvent-based binder. In water-based paints, the health impact is mainly due to the biocide and co-solvents. These results are primarily for illustrative purposes, as the chemical combinations in Table 2 may not reflect the composition of real paint products and the total health impacts presented in Table 2 only consider the organic chemicals and are thus not comprehensive. They are nevertheless useful for comparing the health impacts between several alternatives for a specific function (e.g., biocide) while keeping the rest of the composition constant.

4. Discussion and sensitivity study

4.1. Exposure dynamics to VOC vs. SVOC

To study the dynamic of exposures for the different population groups and for different types of chemicals, we compare the inhalation exposure between the painter and the household adult resident for two example chemicals: formaldehyde as a VOC and Pigment Red 3 as an SVOC. Detailed results are presented in Appendix A, Section A7. For formaldehyde, when considering the 1-day duration which mainly corresponds to the wet phase, the daily inhalation exposure dose is similar between a painter and a household adult resident. However, over 1 year the resident's exposure average daily dose of formaldehyde would drop to about 60 times lower than the painter's exposure dose that is assumed to be painting 200 days per year, because formaldehyde is mainly released soon after the paint application.

In contrast, for Pigment Red 3, the dose is very low when considering

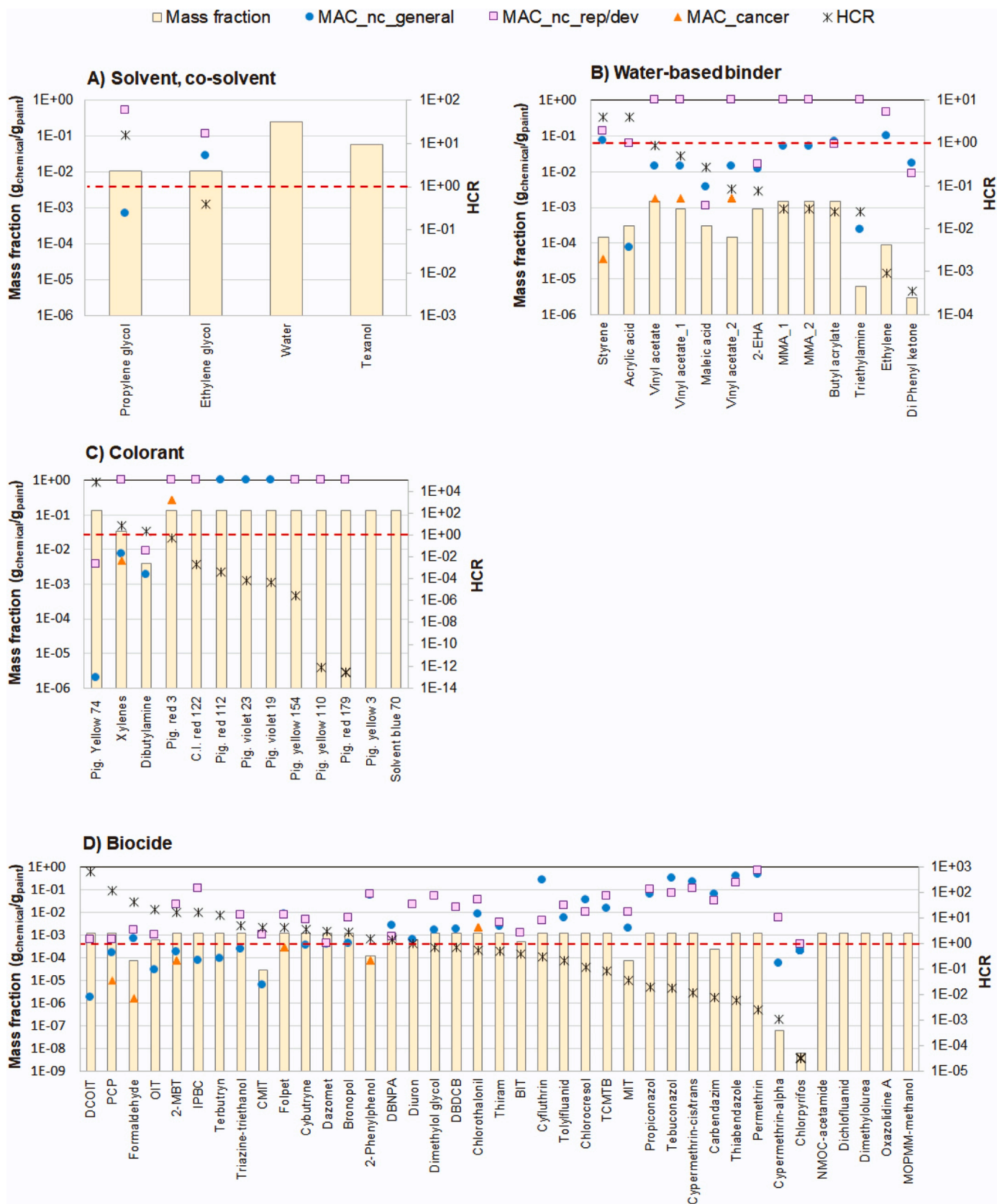


Fig. 4. Actual chemical mass fraction of chemicals in water-based paint (yellow bars) compared to maximum chemical content (MAC_{HTS}) for the household adult resident over 1 year for cancer (orange triangles), non-cancer general effects (blue circles) and non-cancer reproductive/developmental effects (purple squares) for the chemicals in water-based paint (left axis), and resulting Hazard Content Ratios (HCR) (black stars - right axis), for (A) solvent and co-solvent, (B) water-based binder, (C) colorant and (D) biocide. The actual chemical mass fractions in paints with very dark shade colors are shown (Table 1). Chemicals without HCRs are due to lack of toxicity data.

Table 2

Human health impacts of theoretical compositions of solvent-based paints and water-based paints for all adult residents over 1 year following one DIY painting event.

	CAS	Chemical	Function	Health impact (DALY)	Note
Solvent-based paint					
<i>Highest impact</i>	71-43-2	Benzene	Co-solvent/contaminant	6.4E-04	
	100-41-4	Ethylbenzene	Co-solvent/contaminant	2.0E-04	
	110-54-3	n-Hexane	Co-solvent/contaminant	2.1E-05	
	1330-20-7	Xylenes	Solvent	5.9E-04	
	111-92-2	Dibutylamine	Colorant	1.6E-04	Leafing aluminium pigment
	1330-20-7	Xylenes	Colorant	1.3E-05	Leafing aluminium pigment
	80-05-7	Bisphenol A	Solvent-based binder	1.9E-02	Solid, bisphenol-A based Araldite epoxy resin
	Sum			2.1E-02	
<i>Lowest impact</i>	71-43-2	Benzene	Co-solvent/contaminant	6.4E-04	
	100-41-4	Ethylbenzene	Co-solvent/contaminant	2.0E-04	
	110-54-3	n-Hexane	Co-solvent/contaminant	2.1E-05	
	1330-20-7	Xylenes	Solvent	5.9E-04	
	5521-31-3	Pigment red 179	Colorant	1.3E-07	Perlindo Maroon 179
	71-43-2	Benzene	Solvent-based binder	3.2E-05	Long oil urethane alkyd resin
	100-41-4	Ethylbenzene	Solvent-based binder	7.9E-07	Long oil urethane alkyd resin
	Sum			1.5E-03	
Water-based paint					
<i>Highest impact</i>	64359-81-5	DCOIT	Biocide	7.6E-03	
	111-92-2	Dibutylamine	Colorant	1.2E-04	Leafing aluminium pigment
	1330-20-7	Xylenes	Colorant	1.1E-04	Leafing aluminium pigment
	107-21-1	Ethylene glycol	Co-solvent/contaminant	2.0E-04	
	57-55-6	Propylene glycol	Co-solvent/contaminant	2.0E-04	
	77-68-9	Texanol	Co-solvent/contaminant	n/a	
	7732-18-5	Water	Solvent	n/a	
	110-16-7	Maleic acid	Water-based binder	1.0E-03	Sodium salt of polymeric carboxylic acid (Anucryl 80)
	79-10-7	Acrylic acid	Water-based binder	2.2E-04	Sodium salt of polymeric carboxylic acid (Anucryl 80)
	Sum			9.5E-03	
<i>Lowest impact</i>	67375-30-8	Cypermethrin-alpha	Biocide	2.1E-08	
	5521-31-3	Pigment red 179	Colorant	1.1E-07	Perlindo Maroon 179
	107-21-1	Ethylene glycol	Co-solvent/contaminant	2.0E-04	
	57-55-6	Propylene glycol	Co-solvent/contaminant	2.0E-04	
	77-68-9	Texanol	Co-solvent/contaminant	n/a	
	7732-18-5	Water	Solvent	n/a	
	121-44-8	Triethylamine	Water-based binder	2.4E-07	Aliphatic fatty acid modified anionic polyurethane dispersion
	Sum			4.1E-04	

the first day, 8 orders of magnitude lower than that of formaldehyde. However, when considering a 1-year duration after the paint application, the daily inhalation dose of Pigment Red 3 raises by 4 orders of magnitude for the adult resident due to emission during dry phase, whereas it remains low for the painter who is mostly exposed to wet phase paints.

4.2. Influence of ventilation rate and solvent

The influence of the air ventilation rate on the inhalation exposure is most prominent when we consider exposure during the first day of paint application. Fig. 5 thus presents the 1-day inhalation exposure to formaldehyde and pigment red 3 with a high air ventilation rate of 15.6 h^{-1} , which corresponds to a non-airtight building in non-OECD countries [38], as compared to the 0.79 h^{-1} for an airtight building.

For the low air ventilation rate, the household adult resident and the painter both get a high exposure dose, with only 10% reduction for the resident as discussed above.

As expected, the increased air ventilation rate greatly reduces the inhalation exposure of formaldehyde for both the painter and the household adult resident. However, the reduction is larger for the resident than for the painter. For formaldehyde, the high ventilation rate reduces the inhalation exposure dose by 7–9 times for the painter across 12 solvents, while the exposure is reduced by 19–21 times for the household adult, whose exposure becomes less than 50% of that of the painter. For pigment red 3, the high ventilation rate reduces the inhalation exposure dose by 2–9 times for the painter and 4–13 times for the household adult resident. This is because the near-person air breathed by the painter is in direct contact with the paint surface, get the direct

volatilization from the wet paint and therefore remains higher than the far person air breathed by the resident that is highly ventilated, so the air ventilation only indirectly affects the near-person air. These results indicate that although increasing the air ventilation (by opening the windows, doors, etc.) may effectively lower the exposure and health risk for the household residents, it is less effective in protecting the painters. Thus, it is crucial for painters to always wear personal protective equipment (PPE) such as respirators when applying the paints, regardless of the air ventilation rate.

Fig. 5 also shows that the chemical emission and the resulting inhalation exposure are different across various organic solvents. Methanol as the solvent results in the lowest human exposure of formaldehyde, followed by acetone and methylene chloride, but the difference in exposure dose between solvents does not exceed a factor of 2 (Fig. 5A-B). On the other hand, the difference between solvents for an SVOC like pigment red 3 can be up to a factor of 75 (Fig. 5C-D). This suggests that the solvent effect strongly depends on the chemical properties, so the choice of the best solvent needs to be determined based on the actual chemical composition of the paint.

4.3. Health implications for painters and residents

As described in Sections 3.2 and 3.3, our assessment shows that a considerable amount of chemicals in paints would lead to health risks exceeding acceptable levels for both painters and household residents. For noncancer effects, chemicals with $\text{HQ} > 10$ for the household adult over 1 year exposure mainly include xylenes, propylene glycol, pigment yellow 74, and several biocides. On the one hand, a number of studies have found associations between exposures to VOCs in domestic paints

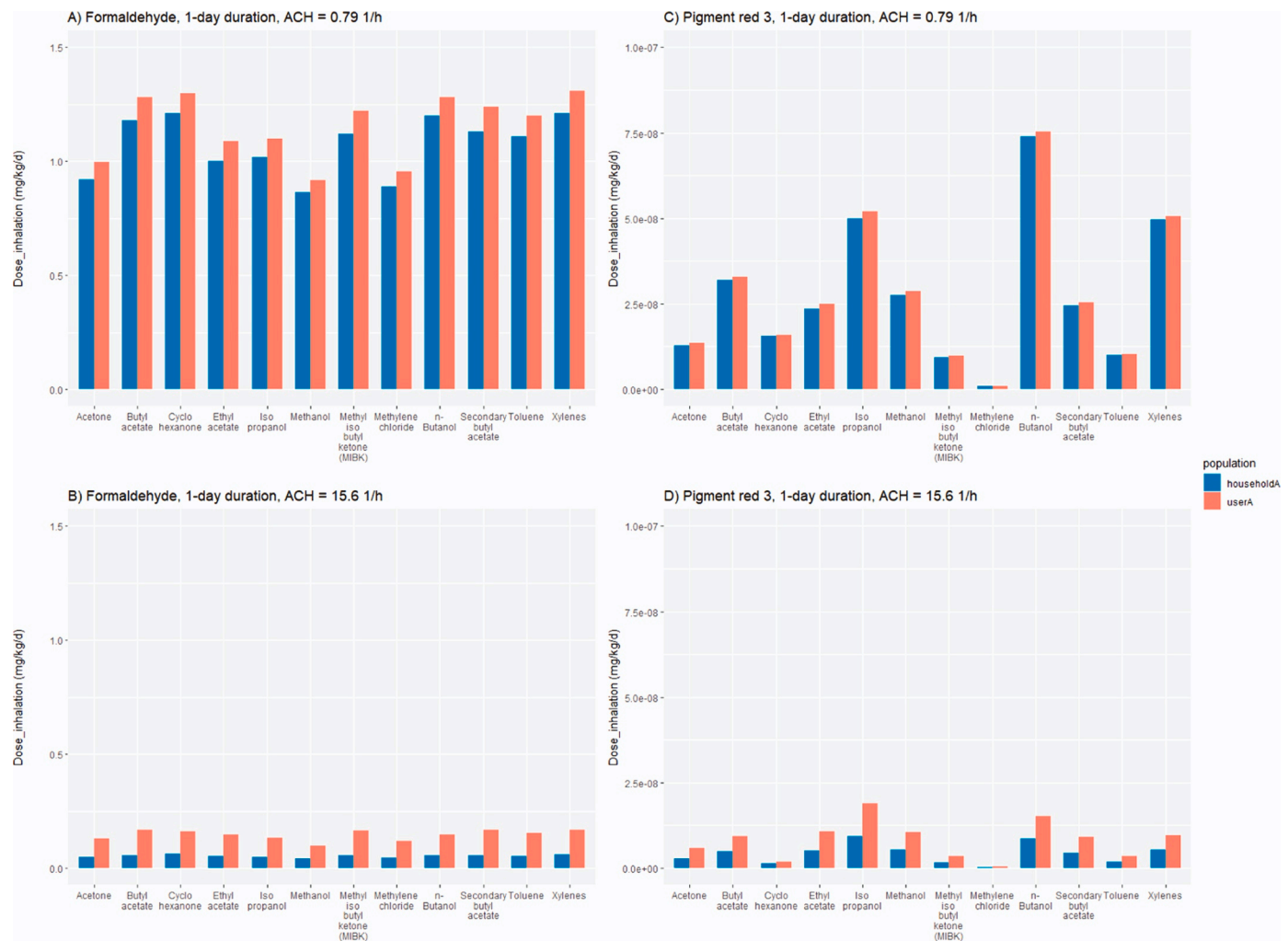


Fig. 5. Comparison of the inhalation exposure dose to formaldehyde (A)(B) and pigment red 3 (C)(D) for the 1st day of application between the painter and the household adult for (A)(C) a low air ventilation rate of 0.79 h^{-1} and (B)(D) a high air ventilation rate of 15.6 h^{-1} .

and asthma, rhinitis and other respiratory symptoms in adults and children [39–42], demonstrating the adverse effects of paints on household residents. On the other hand, there is little or no investigation of the adverse effects of biocides in water-based paints, thus the interest of the present estimates.

For cancer effects, most chemicals with cancer toxicity data exceed the cancer risk of 10^{-4} for the painter and about half of the chemicals exceed cancer risk of 10^{-5} for the household adult resident (Fig. 2). The highest cancer risk is associated with formaldehyde, which leads to a lifetime cancer risk of 3.2×10^{-2} in solvent-based paint and 4.4×10^{-4} in water-based paint, even for the household adult. For estimating the carcinogenicity of formaldehyde, we used the inhalation cancer slope factor (CSF) of $2.17 \text{ (mg/kg/d)}^{-1}$, estimated in USEtox from the harmonic mean of all positive assay for formaldehyde in the carcinogenic potency database. When derived from U.S.EPA's IRIS, the CSF of formaldehyde is $0.057 \text{ (mg/kg/d)}^{-1}$ which is 38 times lower than the USEtox CSF. Thus, the cancer risk of formaldehyde may be a high-end estimate in the present study, but cancer risk from formaldehyde in paints would still be above the 10^{-5} limit even if the IRIS toxicity data are used. Extensive studies have found that occupational exposures in painters are casually associated with the risk of lung cancer, bladder cancer, kidney and other urothelial tumors, and multiple myeloma [43–48]. Although no epidemiological studies have looked at the cancer risk of domestic paint exposure, our results suggest that the cancer risk of frequent domestic painting is also non-negligible for the household

residents.

As presented in Table 2, the human health impacts for two adult residents over 1 year following one DIY painting event range from 4.1×10^{-4} to 0.021 DALY, corresponding to 2×10^{-4} to 0.01 DALY per person. The Global Burden of Disease (GBD) 2019 did not estimate the disease burden for occupational or domestic painting. However, as a comparison, the global burden of risk factors was estimated to be 0.015 DALY per person for ambient particulate matter pollution, 2.3×10^{-4} DALY per person for residential radon exposure, 6×10^{-6} DALY per person for occupational exposure to formaldehyde, and 1.1×10^{-5} DALY per person for occupational exposure to benzene [49]. Our estimated health impacts for an annual painting event are relatively high compared to the GBD estimates for occupational exposures to formaldehyde and benzene, which might be due to uncertainties in toxicity data, but still highlights potential adverse health impacts in Sri Lanka.

Several methods can be used to reduce the cancer and noncancer risks from painting in Sri Lanka. For the painters, it is important to always wear PPE such as respirators and gloves when applying the paints. For the household residents, increasing the air ventilation rate by opening doors and windows during and shortly after painting is an efficient way to reduce exposure and health risk. Residents may also avoid living in the house for 2–3 days after painting, since the VOCs are mostly emitted quickly. Residents may choose water-based paints without formaldehyde as the biocide, or decrease the frequency of repainting to reduce risk.

4.4. Study limitations

The present study has several limitations. First, we focused on organic, non-metal chemicals in interior paints due to model applicability. However, interior paints contain inorganic and metal-containing compounds that may be harmful to humans. For example, ammonia is an inorganic chemical that the paint industry is especially concerned about, and many colorants used in interior paints are metal-containing compounds. Accounting for these chemicals will likely substantially increase the estimated overall health risks. In addition, for certain organic chemicals the hazard index or cancer risk can currently not be calculated due to lack of toxicity data. Furthermore, for all chemicals, we applied 1:1 extrapolation from ingestion route to dermal route, due to limited dermal data, whereas under this hypothesis, dermal exposure was not dominant compared to inhalation. In order to support the comprehensive high-throughput screening of health risks to chemicals in interior paints, there is therefore a need for fate and exposure models able to cover inorganic and metal compounds, as well as for experimental toxicity data for dermal exposure or prediction methods with wider applicability, making use of, for example, appropriate digitalization methods [50,51].

Second, we consider and model a pure solvent when assessing the chemicals in solvent-based paints. In reality, solvent-based paints use a combination of organic solvents, or a solvent mixture such as the commonly used white spirit that we cannot assess. A mixture of various solvents would have different physiochemical properties from pure individual solvents, which may affect the estimates of chemical emission and exposures from the paints. However, as shown in Figure 6 for our exemplary chemicals, inhalation exposures are not substantially affected by the different solvents, especially under a normal air ventilation rate. This suggests that the effect of solvent mixture on human health risk is restricted, and the assumption of pure solvents acceptable for high-throughput screening purposes. In more refined assessments of the chemicals of concern identified in the present study, it is nevertheless desirable to account for solvent mixtures.

5. Conclusions

The present study proposes a high-throughput suited modeling approach to estimate the chemical emission from interior paints, which can accurately predict the solvent drying time and chemical air concentrations. A high-throughput assessment of human exposure and health impacts for chemicals in interior paints is performed using this modeling approach, suggesting that inhalation is the dominant exposure route, followed by dermal gaseous intake. Using as prioritization criteria of a hazard content ratio > 1 , 24 chemicals in water-based paints and 10 in solvent-based paints were identified as chemicals of concern, considering a normal air ventilation rate (0.79 h^{-1}) and a household adult resident exposures. Several biocides, which are only included in water-based paints, were identified as chemicals of concern, highlighting the importance to select low risk biocides for water-based paints. For the painter, human health risk are greatly increased for VOCs when we consider repeated exposures during paint application, with higher levels of risks and additional chemicals of concern. As a result, maximum chemical contents calculated in this study vary by the exposure duration considered, which can provide different reference values for the design of sustainable interior paints to protect different population groups such as professional painters, DIY painters and household residents.

Our results also suggest that during paint application, the inhalation exposure to chemicals in interior paints for the painter is only slightly higher than that for other household residents, and increasing the air ventilation rate will reduce the exposure for other household residents to a larger extent than for the painter. Therefore, this study emphasizes the importance for painters to always wear PPE when applying the

paints to avoid unacceptable health risks from chemicals in the paints.

Environmental Implication

This study proposes a mass-balance mechanistic model to estimate organic chemical emissions and near-field exposures from interior paints, differentiating painter from household residents. It performs a High Throughput screening of paints in Sri Lanka, identifying chemicals of concern and highlighting safer alternatives. It estimates maximum chemical contents and total human health impacts of paint formulations, for the development of safer paint products. This model is integrated in USEtox 3 for use in life cycle assessment, chemical substitution and risk screening.

Interior paints contain hazardous chemicals that are harmful to humans, such as formaldehyde, xylenes, and several biocides in water paints.

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CRediT authorship contribution statement

Lei Huang: Methodology, Software, Validation, Formal analysis, Data curation, Visualization, Writing – original draft preparation. **Niccolò Aurisano:** Data curation, Writing – review & editing. **Peter Fantke:** Methodology, Software, Writing – review & editing, Funding acquisition. **Amal Dissanayake:** Data curation, Writing – review & editing. **L. G.L.M. Edirisinghe:** Resources, Writing – review & editing. **Olivier Jolliet:** Conceptualization, Methodology, Writing – review & editing, Resources, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Olivier Jolliet reports financial support was provided by United Nations Environment Programme. Olivier Jolliet reports a relationship with United Nations Environment Programme that includes: consulting or advisory. O.J. discloses his role as a member of the USEtox Center scientific advisory board and chair of the project on Global guidance for Life Cycle Impact Assessment a project supported by the Life Cycle Initiative, hosted at UN-environment.

Data Availability

Data will be made available on request.

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Appendix A

Near-Field Exposures and Human Health Impacts for Organic Chemicals in Interior Paints: A High-Throughput Screening.

Lei Huang, Nicolò Aurisano, Peter Fantke, Amal Dissanayake, L.G.L.M. Edirisinghe, and Olivier Jolliet.

A1. Input parameters

Table A0 lists all input parameters required to estimate the chemical emission from interior paints and the resulting human exposure, as long as their values or equations.

Table A1

List of all input parameters, including their symbols, values/equations, units, and references.

Parameter	Symbol	Value / Equation	Unit	Reference / Explanation
Total area that the paint is being applied	A_p	42 152	m^2 m^2	1-day exposure for the painter. 1-yr exposure for household adult. Provided by Sri Lanka experts.
Area of the near-person surface	A_{ns}	0.5	m^2	Earnest 2013[1]
Air exchange rate with outdoor air	$AER_{outdoor}$	0.79 15.6	h^{-1} h^{-1}	non-OECD countries (airtight building) non-OECD countries (non-airtight building) Rosenbaum 2015[2]
Chemical i's diffusion coefficient in water	D_{iw}	$8.2 \times 10^{-8} \cdot \frac{T}{\eta_w} \cdot \frac{1 + \left(3 \cdot \frac{MV_w}{MV_i}\right)^{\frac{2}{3}}}{\frac{1}{MV_i^3}}$	cm^2/s	Scheibel 1954[3]
Chemical i's diffusion coefficient in studied product (i.e., paint)	D_{iprod}	$8.2 \times 10^{-8} \cdot \frac{T}{\eta_{product}} \cdot \frac{1 + \left(3 \cdot \frac{MV_{solvent}}{MV_i}\right)^{\frac{2}{3}}}{\frac{1}{MV_i^3}}$	cm^2/s	Scheibel 1954[3]
Diffusion coefficient of CO ₂ in water	$D_{CO_2,w}$	1.84×10^{-5}	cm^2/s	
Diffusion coefficient of CO ₂ in the studied product (i.e., paint)	$D_{CO_2,product}$	$8.2 \times 10^{-8} \cdot \frac{T}{\eta_{product}} \cdot \frac{1 + \left(3 \cdot \frac{MV_{solvent}}{MV_{CO_2}}\right)^{\frac{2}{3}}}{\frac{1}{MV_{CO_2}^3}}$	cm^2/s	Scheibel 1954[3]
Height of the near-person air zone	H_{na}	2	m	Earnest 2013[1]
Heat capacity of air at 25 °C	HC_{air}	1006	J/(kg·K)	http://www.mhtl.uwaterloo.ca/old/onlinetools/airprop/airprop.html
Convective air transfer rate, far-person zone	$h_{a,fs}$	0.00244	m/s	Wenger 2012, 8.8 m ³ /m ² /h[4]
Convective air transfer rate around the body, near-person zone	$h_{a,ns,body}$	$\frac{k_{heat,body}}{HC_{air} \cdot \rho_{air}}$	m/s	
Air-water partition coefficient at 25 °C	K_{aw}	See Appendix B	unitless	
Air-solvent partition coefficient at 25 °C	$K_{a-solvent,i}$	$l \cdot L + s \cdot S + a \cdot A + b \cdot B + v \cdot V + e \cdot E + c$	unitless	http://www.ufz.de/lserd
Skin permeation coefficient via aqueous solution	$K_{p,aq}$	$2.78 \times 10^{-6} \times \left(\frac{0.043}{MW^{1.361}} + 10^{0.7318 \times \log K_{aw} - 0.00683 \times MW - 2.59}\right)$	m/s	Csiszar 2016[5]
Octanol-water partition coefficient at 25 °C	K_{ow}	See Appendix B	unitless	
Heat transfer rate around human body	$k_{heat,body}$	3.4	W/(m ² ·K)	De Dear et al. 1997[6]
Degradation rate in near-person surface	$k_{deg,ns}$	0	s ⁻¹	Assumed no degradation.
Degradation rate in near-person air	$k_{deg,na}$	0	s ⁻¹	Assumed no degradation.
Degradation rate in far-person surface	$k_{deg,fs}$	0	s ⁻¹	Assumed no degradation.
Degradation rate in far-person air	$k_{deg,fa}$	0	s ⁻¹	Assumed no degradation.
Wet paint to skin transfer rate	k_{ps}	$\frac{1}{L_{liquid,init}} \cdot \left(\frac{1}{K_{p,aq}} + \frac{1}{v_{iw}}\right)^{-1}$	s ⁻¹	Csiszar 2016[5]
Wet paint to air transfer rate	k_{pa}	$\frac{1}{L_{liquid,init}} \cdot \left(\frac{1}{v_a \cdot K_{aw} \times 2.78 \times 10^{-6}} + \frac{1}{v_{iw}}\right)^{-1}$	s ⁻¹	Csiszar 2016[5]
Initial thickness of liquid layer of the applied paint	$L_{liquid,init}$	9.1×10^{-5}	m	Provided by Sri Lanka experts: 11 m ² coverage per liter of paint for one coat.

(continued on next page)

Table A1 (continued)

Parameter	Symbol	Value / Equation	Unit	Reference / Explanation
Average thickness of liquid layer of the applied paint, water-based paint	$L_{liquid,avg}$	$0.75 \times L_{liquid,init}$	m	Ludwig 2005: thickness of waterborne latex would be reduced by half when water is dried.[7]
Average thickness of liquid layer of the applied paint, solvent-based paint	$L_{liquid,avg}$	$0.9 \times L_{liquid,init}$	m	We assume the final thickness of solvent-based paint after solvent is dried is 80% of the initial thickness because weight fraction of solvent is about 20% in solvent-based paint.
Molar volume of chemical i	MV_i	See Appendix B	cm ³ /mol	
Molar volume of water	MV_w	See Appendix B	cm ³ /mol	
Molar volume of CO ₂ at room temperature and pressure	MV_{CO_2}	44.7	cm ³ /mol	
Molar volume of the studied solvent	$MV_{solvent}$	See Appendix B	cm ³ /mol	
Molecular weight	MW_i	See Appendix B	g/mol	
Total mass of the applied paint	m_{paint}	$A_{app} \bullet L_{liquid,init} \bullet \rho_{paint}$	kg	
Air exchange rate between the near-person zone and the far-person zone	$Q_{na \rightarrow fa}$	200	m ³ /h	Earnest 2013[1]
Dermal contact rate with wet paint during application	$R_{contact}$	30	mg/min	ConsExpo Paint factsheet[8]
Air density at 25 °C	ρ_{air}	1.185	kg/m ³	http://www.mhlt.uwaterloo.ca/old/onlinetools/airprop/airprop.html
Density of water at 25 °C	ρ_w	0.999	g/cm ³	
Density of studied solvent at 25 °C	$\rho_{solvent}$	See Appendix B	g/cm ³	
Density of paint at room temperature	ρ_{paint}	1250	kg/m ³	ConsExpo Paint factsheet[8]
Speed of applying paint	S_{app}	480	s/m ²	ConsExpo Paint factsheet, which needs 2 h for painting an area of 15 m ² . [8]
Schmidt number of chemical i in water	Sc_{iw}	$\frac{\mu_w}{D_{iw}}$	unitless	Schwarzenbach 2003[9]
Schmidt number of chemical i in product (i.e., paint)	$Sc_{ipproduct}$	$\frac{\mu_{product}}{D_{ipproduct}}$	unitless	Analogous to the equation for Sc_{iw}
Time needed for solvent to dry	t_{drying}		s	Calculated as the time when 60% of the solvent mass is volatilized to air. See Section A1.4.
Volume of the house	V_{house}	117	m ³	non-OECD countries average, Rosenbaum 2015[2]
Volume of near-person air zone	V_{na}	1	m ³	Earnest 2013[1]
Volume of far-person air zone	V_{fa}	116	m ³	$V_{house} - V_{na}$
Air flow rate at skin surface	v_a	1000	cm/h	Default
Water flow rate at paint-skin interface	v_{iw}	$20.62 \times MW^{0.4757} \times 2.78 \times 10^{-6}$	m/s	Csiszar 2016[5]
Water-side mass transfer velocity	$\phi_{waterside,i}$	$\phi_{waterside,CO_2} \bullet \left(\frac{Sc_{iw}}{600}\right)^{\frac{2}{3}}$	m/s	Schwarzenbach 2003[9]
Waterside transfer velocity of CO ₂ at a Schmit number of 600	$\phi_{waterside,CO_2}$	6.5×10^{-6}	m/s	Schwarzenbach 2003[9]
Air-side mass transfer velocity over water	$\phi_{airside,iw}$	$K_{aw} \bullet h_{a,body,NP}$	m/s	
Water to air mass transfer velocity	$\phi_{water \rightarrow air,i}$	$\left(\frac{1}{\phi_{waterside,i}} + \frac{1}{\phi_{airside,iw}}\right)^{-1}$	m/s	Two-resistance theory
Product-side mass transfer velocity	$\phi_{productside,i}$	$\phi_{productside,CO_2} \bullet \left(\frac{Sc_{ipproduct}}{600}\right)^{\frac{2}{3}}$	m/s	Analogous to the equation for $\phi_{waterside,i}$
Product-side transfer velocity of CO ₂ at a Schmit number of 600	$\phi_{productside,CO_2}$	$\phi_{waterside,CO_2} \bullet \left(\frac{\mu_{product}}{\mu_w}\right)^{-\frac{2}{3}} \bullet \left(\frac{D_{CO_2,w}}{D_{CO_2,product}}\right)^{-\frac{2}{3}}$	m/s	See Section A1.3.
Air-side mass transfer velocity over product	$\phi_{airside,product,i}$	$K_{a-solvent,i} \bullet h_{a,fs}$	m/s	
Product to air mass transfer velocity	$\phi_{product \rightarrow air,i}$	$\left(\frac{1}{\phi_{productside,i}} + \frac{1}{\phi_{airside,product,i}}\right)^{-1}$	m/s	Two-resistance theory
Kinematic viscosity of water	μ_w	$\frac{\eta_w}{\rho_w} \bullet \frac{1}{100}$	cm ² /s	
Kinematic viscosity of product (i.e., paint)	$\mu_{product}$	$\frac{\eta_{product}}{\rho_{product}} \bullet \frac{1}{100}$	cm ² /s	
Viscosity of product (i.e., paint)	$\eta_{product}$	4 for water-based paint 8 for solvent-based paint	centipoise	We assume the water paint has higher viscosity than water itself. We assume the solvent paint is twice more viscous than water paint, and the viscosity of solvent-based paint is assumed the same across different solvents.
Abraham solvent coefficients of the studied solvent	l, s, a, b, v, e, c	See Appendix B	Various units	http://www.ufz.de/lserd
Abraham solute descriptors of chemical i	L, S, A, B, V, E	See Appendix B	Various units	http://www.ufz.de/lserd

A2. Model for chemical emission and exposure during wet phase

A2.1 Four-compartment indoor model

Table A2 presents the equations for calculating the transfer rate constants for the 4-compartment model.

Table A2

Equations for inter-compartment transfer rate constants for the 4-compartment model. Input parameters are detailed in Section A0.

Near-person surface (ns):

$$k_{ns \rightarrow na} = \frac{\varphi_{p \rightarrow a}}{L_{liquid,avg}} \quad (A1a)$$

$$k_{ns \rightarrow fs} = \frac{1}{S_{app} * A_{ns}} \quad (A1b)$$

$$k_{ns \rightarrow skin} = \frac{R_{contact}}{m_p * 6 * 10^7} * \frac{k_{ps}}{k_{ps} + k_{pa}} * (1 - e^{-(k_{ps} + k_{pa}) * t_{drying}}) \quad (A1c)$$

$$k_{ns,total} = k_{ns \rightarrow na} + k_{ns \rightarrow fs} + k_{deg,ns} + k_{ns \rightarrow skin} \quad (A1d)$$

Near-person air (na):

$$k_{na \rightarrow ns} = \frac{h_{a,ns}}{H_{na}} * \frac{1}{(1 + \frac{\varphi_{airside}}{\varphi_{productside}})} \quad (A1e)$$

$$k_{na \rightarrow fa} = \frac{Q_{na \rightarrow fa}}{V_{na}} \quad (A1f)$$

$$k_{na,total} = k_{na \rightarrow ns} + k_{na \rightarrow fa} + k_{deg,na} \quad (A1g)$$

Far-person surface (fs):

$$k_{fs \rightarrow fa} = \frac{\varphi_{p \rightarrow a}}{L_{liquid,avg}} \quad (A1h)$$

$$k_{fs,total} = k_{fs \rightarrow fa} + k_{deg,fs} \quad (A1i)$$

Far-person air (fa):

$$k_{fa \rightarrow na} = \frac{Q_{na \rightarrow fa}}{V_{fa}} \quad (A1j)$$

$$k_{fa \rightarrow fs} = h_{a,fs} * \frac{A_p}{V_{fa}} * \frac{1}{(1 + \frac{\varphi_{airside}}{\varphi_{productside}})} \quad (A1k)$$

$$k_{fa,total} = k_{fa \rightarrow fs} + k_{fa \rightarrow na} + k_{deg,fa} + AER * \frac{(V_{fa} + V_{na})}{V_{fa}} \quad (A1l)$$

$\varphi_{p \rightarrow a}$ is the mass transfer velocity from product (i.e., paint) to air (m/s) accounting for convective transfer at the liquid and air boundaries of the product surface, $L_{liquid,avg}$ is the average thickness of liquid layer of the applied paint (m), S_{app} is the time per unit area needed to apply the paint (s/m²), A_{ns} is the area of near-person surface (m²), $h_{a,na}$ is the convective air transfer coefficient around the body in near-person zone (m/s), $h_{a,fa}$ is the convective air transfer coefficient in far-person zone (m/s), H_{na} is the height of the near-person zone (m), $\varphi_{airside}$ is the airside mass transfer velocity over the product (i.e., paint) (m/s), $\varphi_{productside}$ is the product-side mass transfer velocity (m/s), $Q_{na \rightarrow fa}$ is the air exchange rate between the near-person zone and the far-person zone (m³/s), V_{na} is the volume of the near-person air zone (m³), V_{fa} is the volume of the far-person air zone (m³), A_p is the total area that the paint is being applied to (m²), AER is the air exchange rate with outdoor air (s⁻¹), $R_{contact}$ is the dermal contact rate with paint (mg/min), m_p is the mass of the applied paint product(kg), k_{ps} is the paint-skin transfer rate constant (s⁻¹), k_{pa} is the paint-air transfer rate constant (s⁻¹), t_{drying} is the duration that the applied wet paint needs to dry (s), $k_{deg,ns}$, $k_{deg,na}$, $k_{deg,fs}$, and $k_{deg,fa}$ are the degradation rate constants (s⁻¹) in the four compartments, respectively.

A2.2 Net transfer from surface to air accounting for air-to-surface feedback

To derive the net transfer rates from surface to air (both near-person and far-person) as in main text [Section 2.3.1](#), we consider a one-box model which divides the house into two compartments: indoor air and the product surface. We assume that the indoor air is at quasi-steady-state, so the mass balance of indoor air can be written as:

$$\frac{dm_a}{dt} = k_{s \rightarrow a} * m_s - (k_{a \rightarrow s} + k_{a,out}) * m_a \approx 0 \quad (A2)$$

$$\rightarrow m_a = \frac{k_{s \rightarrow a}}{k_{a \rightarrow s} + k_{a,out}} * m_s \quad (A3)$$

Inserting [Eq. A3](#) into the mass balance equation for the product surface, we get:

$$\frac{dm_s}{dt} = -k_{s \rightarrow a} * m_s + k_{a \rightarrow s} * m_a \quad (A4)$$

$$= -m_s * (k_{s \rightarrow a} - \frac{k_{a \rightarrow s} * k_{s \rightarrow a}}{(k_{a \rightarrow s} + k_{a,out})})$$

$$= -m_s * k_{s \rightarrow a} * (1 - \frac{k_{a \rightarrow s}}{(k_{a \rightarrow s} + k_{a,out})})$$

$$= -k_{net,s \rightarrow a} * m_s$$

Thus, the net transfer rate from surface to air for both near-person and far-person zones is given by:

$$k_{net,s \rightarrow a} = k_{s \rightarrow a} \bullet \left(1 - \frac{k_{a \rightarrow s}}{(k_{a \rightarrow s} + k_{a,out})}\right) \approx k_{s \rightarrow a} \bullet \left(1 - \frac{k_{a \rightarrow s}}{(k_{a \rightarrow s} + AER_{outdoor})}\right) \quad (A5)$$

A2.3 Product-side transfer velocity of CO₂

The product-side mass transfer velocity of the studied chemical *i* in the paint is calculated similar as the water-side mass transfer velocity, for which we first need to derive the product-side transfer velocity of CO₂, $\varphi_{productside,CO_2}$. According to [9], there is a relationship between the water-side transfer velocity and the Schmidt number:

$$\varphi_{waterside,i} = constant \bullet (Sc_{iw})^{-\frac{2}{3}}, \text{ with } Sc_{iw} = \frac{\mu_w}{D_{iw}} \quad (A6)$$

Thus, we assume the product-side transfer velocity follows the same rule as the water-side transfer velocity:

$$\begin{aligned} \varphi_{productside,CO_2} &= constant \bullet (Sc_{CO_2,product})^{-\frac{2}{3}} = constant \bullet \left(\frac{\mu_{product}}{D_{CO_2,product}}\right)^{-\frac{2}{3}} \\ &= constant \bullet \left(\frac{\mu_w}{D_{CO_2,w}}\right)^{-\frac{2}{3}} \bullet \left(\frac{\mu_{product}}{\mu_w}\right)^{-\frac{2}{3}} \bullet \left(\frac{D_{CO_2,w}}{D_{CO_2,product}}\right)^{-\frac{2}{3}} \\ &= [constant \bullet (Sc_{iw})^{-\frac{2}{3}}] \bullet \left(\frac{\mu_{product}}{\mu_w}\right)^{-\frac{2}{3}} \bullet \left(\frac{D_{CO_2,w}}{D_{CO_2,product}}\right)^{-\frac{2}{3}} \\ &= \varphi_{waterside,CO_2} \bullet \left(\frac{\mu_{product}}{\mu_w}\right)^{-\frac{2}{3}} \bullet \left(\frac{D_{CO_2,w}}{D_{CO_2,product}}\right)^{-\frac{2}{3}} \end{aligned} \quad (A7)$$

with the parameters explained in Table A0.

A2.4 Calculation of drying time

To estimate the time needed for the paint to be dry, we assume that 60% of the organic solvent or water is volatilized when the paint is dry and thus calculate the corresponding time. We first calculate the solvent to air transfer velocity of the solvent molecule in solvent itself, $\varphi_{p \rightarrow a,i}$ using equations presented in Table A0, setting chemical *i* as the solvent itself. For example, to estimate the drying time of xylenes, we assume the solvent as xylenes and the chemical *i* as also xylenes.

Then we calculate an adjusted surface to air transfer rate coefficient for solvent itself:

$$k_{net,fs \rightarrow fa,ss} = \frac{\varphi_{p \rightarrow a,ss}}{L_{liquid,avg}} \bullet f_{adj,ss} \quad (A8)$$

where the subscript “ss” represents solvent chemical in solvent itself. For organic solvents, $f_{adj,ss}$ is calculated using Eq. A9a, analogous to Eq. A5. In contrast, if the solvent is water, the air-to-surface feedback is mainly limited by the pre-existing water vapor in the air (i.e., relative humidity), so we assume that $f_{adj,ss}$ is determined by the relative humidity of indoor air, as in Eq. A9b.

$$f_{adj,ss,solvent} = 1 - \frac{k_{fa \rightarrow fs}}{(k_{fa \rightarrow fs} + AER_{outdoor})} \quad (a9a)$$

$$f_{adj,ss,water} = 1 - \frac{RH}{100} \quad (A9b)$$

where *RH* is the relative humidity (%).

Finally, the time needed for 60% of the solvent to be volatilized from product to air is given by:

$$t_{drying} = -\frac{\ln(1 - 0.6)}{k_{net,fs \rightarrow fa,ss}} \quad (A10)$$

A3. Model for chemical emission and exposure during dry phase

The D-limited and K-limited models in Eq. 8 of main text are described below.

A3.1 D-limited model

The chemical emission from dried paint is analogous to the chemical emission from building materials. For most volatile organic compounds (VOCs), the emission of the chemical from building material is mainly limited by the chemical diffusion inside the building material, and the chemical sorption on other indoor surfaces can be ignored [10,11]. Huang and Jolliet have developed a simplified solution for VOCs by assuming that the indoor air concentration is at quasi-steady-state between the emission from building material and the loss by ventilation [12]. This is used as the “D-limited model”. In the case of dried paint, the chemical mass fraction emitted from dried paint to indoor air from time zero to time *t* is given by [12]:

$$TF_{p \rightarrow a,DP} = \frac{m_e(t)}{m_0} = -\alpha \bullet e^{-\beta_1^2 D_m t} + (\alpha - 1) \bullet e^{-\beta_2^2 D_m t} + 1 \quad (A11)$$

where $TF_{p \rightarrow a, DP}$ is the direct chemical mass transfer fraction from dried paint to air (dimensionless), $m_e(t)$ is the total chemical mass emitted to air from time zero to time t (μg), m_0 is the total chemical mass in the dried paint at time zero (μg), t is time (s), α , β_1 , β_2 are coefficients which are calculated as functions of convective mass-transfer coefficient h_m (m/s), room ventilation rate Q (m^3/s), area of the dried paint A_m (m^2), thickness of dried paint L (m), as well as D_m and K_{ma} . Detailed equations for calculating α , β_1 , β_2 can be found in Huang et al., 2021, Supporting Info, Section S3.3 [13].

A 3.2 K-limited model with sorption

For the K-limited cases, chemical sorption onto indoor surfaces (e.g., walls and ceilings) is significant and cannot be ignored, so it is assumed that the chemical remains evenly distributed inside the dried paint and in the sorption material, i.e., no concentration gradient through the materials. The detailed equations for the K-limited model with sorption can be found in Huang et al., 2022, Supporting Info, Section S2.2.2 [14].

Briefly, the mass balance equations can be formulated in a matrix format as follows:

$$\dot{\mathbf{M}}(t) = \mathbf{A}_k \bullet \mathbf{M}(t) \quad (\text{A12})$$

where

$$\mathbf{M}(t) = \begin{bmatrix} m_{\text{paint}}(t) \\ m_{\text{sorp}}(t) \end{bmatrix}$$

$$\mathbf{A}_k = \begin{bmatrix} a & b \\ c & d \end{bmatrix}$$

$$\text{with } a = -\frac{h_m}{L \bullet K_{ma}} + \frac{h_m^2 \bullet A_m}{L \bullet K_{ma}} \bullet \frac{1}{Q \bullet (1 + K_{pa} \bullet TSP) + h_m \bullet A_m + h_s \bullet A_s},$$

$$b = \frac{h_m \bullet A_m \bullet h_s}{L_s \bullet K_s} \bullet \frac{1}{Q \bullet (1 + K_{pa} \bullet TSP) + h_m \bullet A_m + h_s \bullet A_s},$$

$$c = \frac{h_s \bullet A_s \bullet h_m}{L \bullet K_{ma}} \bullet \frac{1}{Q \bullet (1 + K_{pa} \bullet TSP) + h_m \bullet A_m + h_s \bullet A_s},$$

$$d = -\frac{h_s}{L_s \bullet K_s} + \frac{h_s^2 \bullet A_s}{L_s \bullet K_s} \bullet \frac{1}{Q \bullet (1 + K_{pa} \bullet TSP) + h_m \bullet A_m + h_s \bullet A_s}$$

where $m_{\text{paint}}(t)$ is the chemical mass in the dried paint at time t (μg), $m_{\text{sorp}}(t)$ is the chemical mass in the sorption material at time t (μg), A_s is the area of sorption surfaces (m^2), L_s is the thickness of sorption material (m), K_s is the chemical's sorption material-air partition coefficient (dimensionless), h_s is the convective mass-transfer coefficient on the sorption surfaces (m/s), K_{pa} is the chemical's particle-gas partition coefficient ($\text{m}^3/\mu\text{g}$), and TSP is the total suspended particle concentration in indoor air ($\mu\text{g}/\text{m}^3$).

The solution of Eq. A12 is given by:

$$\mathbf{M}(t) = c_1 e^{\lambda_1 t} \mathbf{u}_1 + c_2 e^{\lambda_2 t} \mathbf{u}_2 \quad (\text{A13a})$$

where λ_1 and λ_2 are the two eigenvalues of matrix \mathbf{A}_k , \mathbf{u}_1 and \mathbf{u}_2 are the respective eigenvectors of \mathbf{A}_k , and c_1 and c_2 are constants calculated as a function of the initial masses as follows:

$$\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} = [\mathbf{u}_1 \quad \mathbf{u}_2]^{-1} \mathbf{M}(0) = [\mathbf{u}_1 \quad \mathbf{u}_2]^{-1} \begin{bmatrix} m_{\text{paint}}(0) \\ m_{\text{sorp}}(0) \end{bmatrix} \quad (\text{A13b})$$

After obtaining $m_{\text{paint}}(t)$ and $m_{\text{sorp}}(t)$, the direct transfer fraction from dried paint to indoor air from time zero to time t is given by:

$$TF_{\text{paint} \rightarrow \text{air}, DP} = 1 - \frac{m_{\text{paint}}(t)}{m_0} \quad (\text{A14})$$

A3.3 Human exposure

Inhalation exposure.

There is no direct transfer from the paint to the building occupants' respiratory tract. Chemicals are first transferred from the paint to indoor air, and are then further transferred from indoor air to the respiratory tract via inhalation. The direct transfer fraction from indoor air to respiratory tract is calculated as:

$$TF_{a \rightarrow \text{resp. tract}} = \frac{C_a \bullet inhR \bullet f_{\text{indoor}} \bullet N_h}{C_a \bullet V \bullet n \bullet CF_{ds}} = \frac{inhR \bullet f_{\text{indoor}} \bullet N_h}{V \bullet n \bullet \text{day_to_second}} \quad (\text{A15})$$

where $inhR$ is the individual inhalation rate (m^3/d), f_{indoor} is the fraction of time spent indoors per day (unitless), N_h is the number of persons in the building, V is the building volume (m^3), n is the air renewal rate (s^{-1}), CF_{ds} is the conversion factor from day to second which is 86400 (s/d). All input parameters are explained in Table A0.

Dermal exposure by gaseous uptake.

Once a chemical is emitted from the paint to the indoor air, it could also be absorbed by human skin via gaseous uptake. This process only occurs for

chemicals in the gas-phase of the indoor air. The transfer fraction for dermal gaseous uptake is calculated by multiplying the average gaseous air concentration by a gaseous-skin permeation coefficient, as follows:

$$TF_{a \rightarrow \text{skin}}^{\text{dermal gaseous}} = \frac{\frac{C_a}{(1+K_{pa} \bullet TSF)} \bullet K_{p, \text{gas, total}} \bullet A_{\text{skin, gas}} \bullet f_{\text{indoor}} \bullet N_h}{C_a \bullet V \bullet n} = \frac{K_{p, \text{gas, total}} \bullet A_{\text{skin, gas}} \bullet f_{\text{indoor}} \bullet N_h}{(1+K_{pa} \bullet TSF) \bullet V \bullet n} \quad (\text{A16}).$$

where $K_{p, \text{gas, total}}$ is the total gaseous-skin permeation coefficient (m/s), and $A_{\text{skin, gas}}$ is the skin gaseous uptake area (m²). All input parameters are explained in Table A0.

Dermal exposure by direct contact.

Dermal exposure by direct contact is assumed to only occur during the paint application, which is given by the direct transfer fraction from near-person surface to human skin, as presented in Eq. A1c.

A4. Toxicity data for risk characterization

A4.1 Cancer effects

For cancer effects, we used cancer slope factors (CSFs) expressed as (mg/kg_{BW}/d)⁻¹ specific to each of the three exposure routes considered and differentiating between adults and children. For adults, we directly used the CSFs estimated from TD50 toxic dose data available from the scientific consensus model USEtox [15]. For children, we multiplied the available CSFs by an age-dependent adjustment factor (ADAF) of 4, as an age-weighted factor between 0 and 2 years (ADAF = 10) and between 2 and 14 years (ADAF = 3) [16]. In the case of CSFs not available for specific exposure routes, we applied route-to-route extrapolation to dermal exposure and between ingestion and inhalation exposure [15].

A4.2 Non-cancer effects

For non-cancer effects, the ingestion reference doses (RfD) and inhalation reference concentrations (RfC) were obtained from a published database providing peer-reviewed toxicity values reported in various regulatory sources, including, for example, the U.S. EPA's Integrated Risk Information System (IRIS) and the Superfund Regional Screening Level Tables (RSLs) [17,18]. For the chemicals for which RfDs or RfCs were not available in the considered regulatory sources, we used the probabilistic RfDs and RfCs derived by Aurisano et al. [19,20]. These probabilistic RfDs and RfCs are derived from experimental animal data reported in the U.S. EPA's Toxicity Value Database, systematically applying the World Health Organization International Programme on Chemical Safety (WHO/IPCS) framework for dose-response assessment [21,22]. The collected RfDs and RfCs differentiate between general non-cancer and reproductive/developmental effects due to the factor 20 difference in severity affecting human lifetime loss [23,24]. Finally, for the substances without RfDs or RfCs available in the above sources, we designated them as "N/A". Due to the lack of dermal RfDs we systematically applied for all substances a route-to-route extrapolation from oral RfDs.

A5. Evaluation of the proposed models

A5.1 Prediction of drying time

To evaluate the proposed models, we first compare our estimates of drying time for water and different organic solvents to literature values. Ludwig et al. [7] measured the water evaporation from waterborne latices. Fig. A1a presents the measured fraction of water left on latex film versus our predicted values using conditions specified in Ludwig 2005 (temperature of 23 °C, relatively humidity of 30%). Ludwig et al. did not specify the room volume and air ventilation rate, so we used our default values for Sri Lanka for one day of paint application (volume of 117 m³, ventilation rate of 0.79 h⁻¹, application area of 42 m²). Our predicted fraction of remaining water generally follows the trend of the measured values. The prediction slightly overestimates the water evaporation rate (i.e., underestimate the fraction of water left) at the beginning, but underestimates the evaporation rate at the end, which is expected because we used the average liquid thickness in the model and assumed the liquid thickness remained constant. Based on our definition of drying time that 60% of solvent/water is volatilized (Appendix A, Section A1.4), we estimated the drying time to be 801 s in this situation, which is close to the drying time suggested by the measured data (between 960 s and 1130 s).

For the drying time of pure organic solvents, the evaporation rates relative to butyl acetate for various pure organic solvents are reported by the industry (<http://ws.eastman.com/Wizards/eSolvents/ESolvProperty.asp?Solvent=10057&Property=-1>). We thus predicted the drying time of 14 organic solvents and converted them to relative evaporation rates. For example, we estimate the drying time of butyl acetate and toluene to be 1.1 h and 0.6 h respectively in an air-tight building, so the relative evaporation rate is 1 for butyl acetate and 1.78 for toluene. Note that to calculate the drying time of pure solvents, we used the viscosity (η_{solvent} , Table A0) of the pure solvent instead of the viscosity of 8 centipoise for solvent-based paint. Viscosities of the pure solvents can be found in Appendix B. As shown in Fig. A1b, our predicted evaporation rates (or drying time) mostly agree well with the reported values, and the predicted relative evaporation rates are almost not affected by the air ventilation rate. Our model overestimates the evaporation rates for ethyl acetate and acetone, but they are within a factor of 2. Our model also overestimates the evaporation rate for water in an airtight building, because we assumed that the water evaporation is affected by the relative humidity of indoor air instead of the ventilation rate. Overall, the results demonstrate that our model can predict the drying time of water and other organic solvents relatively accurately.

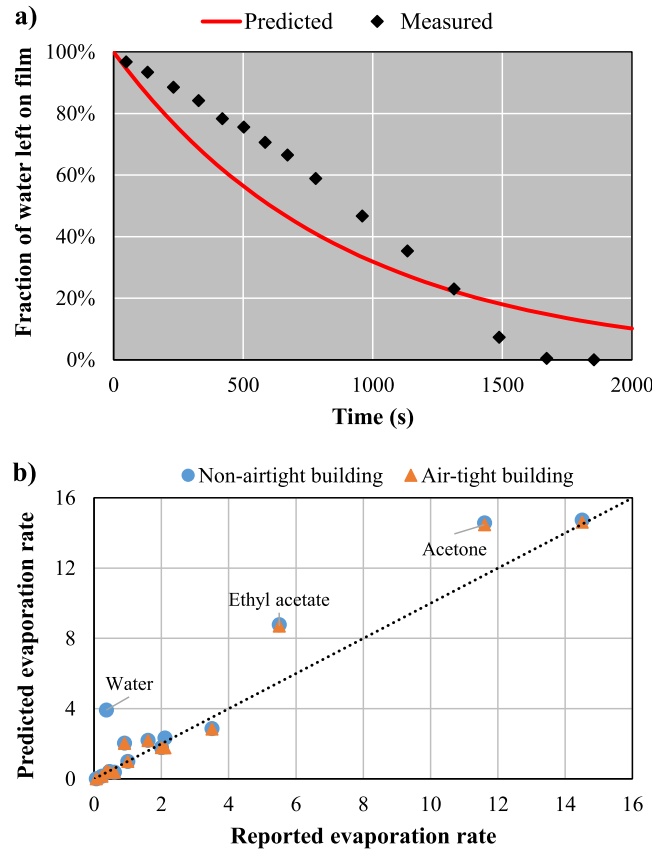


Fig. A1. Prediction of drying time using proposed models. (a) Predicted fraction of water left on a waterborne latex film as a function of time versus the measured values (Ludwig 2005). (b) Predicted evaporation rates relative to butyl acetate for water and 13 other organic solvents versus the reported values (Eastman Chemical Company, 2015 #809); the dotted line represents the 1:1 line.

A5.2 Prediction of VOC emission from paints

As a next step of model evaluation, we compare our model predictions to measured VOC emission from paints in chamber studies. In small chamber emissions tests, paints were applied on substrates which were then placed on the floor of the chamber, and the concentrations of VOCs in the chamber air were monitored over time [25,26]. To better represent the chamber test conditions which have no near-person zone, we reduce the four-compartment model (described in the main text Section 2.3.1) to a two-compartment model, which includes only the far-person air and the far-person surface compartments. The mass balance of this 2-compartment system can be described by the following equation:

$$\dot{\mathbf{M}}_2(t) = \mathbf{K}_2 \bullet \mathbf{M}_2(t) \quad (\text{A17})$$

where

$$\mathbf{M}_2(t) = \begin{bmatrix} m_{fs}(t) \\ m_{fa}(t) \end{bmatrix}$$

$$\mathbf{K}_2 = \begin{bmatrix} -k_{fs, total} & k_{fa \rightarrow fs} \\ k_{fs \rightarrow fa} & -k_{fa, total} \end{bmatrix}$$

fs: far-person surface. fa: far-person air.

The 4 transfer rate constants are calculated as follows:

$$k_{fs \rightarrow fa} = \frac{\varphi_{product \rightarrow air, i}}{L_{liquid, avg}} \quad (\text{A18a})$$

$$k_{fs, total} = k_{fs \rightarrow fa} + k_{deg, fs} \quad (\text{A18b})$$

$$k_{fa \rightarrow fs} = h_{a, fs} \bullet \frac{A_p}{V_{house}} \bullet \frac{1}{(1 + \frac{\varphi_{airside, is}}{\varphi_{productside, i}})} \quad (\text{A18c})$$

$$k_{fa, total} = k_{fa \rightarrow fs} + k_{deg, fa} + AER_{outdoor} \quad (\text{A18d})$$

with the parameters explained in Table A0.

The solution of Eq. A17 is given by:

$$\mathbf{M}_2(t) = c_1 e^{\lambda_1 t} \mathbf{u}_1 + c_2 e^{\lambda_2 t} \mathbf{u}_2$$

(A19)

where λ_1 and λ_2 are the two eigenvalues of matrix \mathbf{M}_2 , \mathbf{u}_1 and \mathbf{u}_2 are the respective eigenvectors of \mathbf{M}_2 , and c_1 and c_2 are constants calculated as a function of the initial masses as follows:

$$\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} = [\mathbf{u}_1 \quad \mathbf{u}_2]^{-1} \mathbf{M}_2(0) = [\mathbf{u}_1 \quad \mathbf{u}_2]^{-1} \begin{bmatrix} m_{fs}(0) \\ m_{fa}(0) \end{bmatrix}$$

(A20)

Finally, the chemical concentration in the chamber air is given by:

$$C_{fa}(t) = \frac{m_{fa}(t)}{V_{house}}$$

(A21)

The air concentration obtained in Eq. A21 can thus be compared to the measured air concentrations in chamber emission tests.

We compared our model predictions to two chamber studies, one for the emission of methyl ethyl ketoxime (MEKO) from alkyd paints [26] and the other for the emission of decane from alkyd paints [25]. These two chamber studies were used by the U.S.EPA to develop the Wall Pain Exposure Model (WPEM version 3.2) [27]. Table A2 presents the input parameter values for our models specific to these two studies. For the solvent system used in the model, Chang et al. [26] did not mention the solvent of the tested alkyd paints, so we assume the solvent is xylenes. Fortmann et al. [25] measured the composition of the tested alkyd paints which included several VOCs as the solvent component, and we select the VOC with the highest weight fraction as the solvent used in our models, which is decane.

Figures A2 and A3 present the modeled and measured chamber air concentrations of MEKO and decane emitted from alkyd paints. For MEKO emission, our model predictions agree well with the measured air concentrations (Figure A2). The model also captures the peak air concentrations accurately, where the predicted peak air concentrations are within 91–127% of the measured values for the three alkyd paints. However, for decane emission from alkyd paints, the model predicts a slower release than the measurements, as it predicts lower peak air concentrations but slightly higher concentrations in the long term (Figure A3). The predicted peak air concentrations of decane are within 57–75% of the measured values for the three different conditions of alkyd paint A. This could be due to an underestimate of the air-solvent partition coefficient of decane in this alkyd paint. The estimated air-solvent partition coefficient we used is 1.59×10^{-5} , and if this partition coefficient is increased by a factor of 5, our model predictions would agree perfectly with the measured air concentrations. When we estimated the air-solvent partition coefficient, we assumed the solvent is pure decane, which may not well represent the properties of the alkyd paint A used by Fortmann et al. [25], leading to underestimate of the partition coefficient. Overall, the results suggest that our model can predict the VOC emissions from wet paints relatively accurately, with the predicted air concentrations within a factor of 2 of the measured values.

Table A3
Input parameter values for the two chamber studies.

Parameter	V_{house}	A_{app}	$AER_{outdoor}$	$L_{liquid\ avg}$	$h_{a,body\ FP}$	m_{paint}
Unit	m ³	m ²	h ⁻¹	m	m/s	g
Chang 1998, Paint 1	0.053	0.0256	0.5	6.60E-05	0.00244	1.72
Chang 1998, Paint 2	0.053	0.0256	0.5	6.60E-05	0.00244	2.11
Chang 1998, Paint 3	0.053	0.0256	0.5	6.60E-05	0.00244	2.10
Fortmann 1998, Paint A, standard	0.053	0.0256	0.5	4.87E-05	0.00244	2.30
Fortmann 1998, Paint A, thick layer	0.053	0.0256	0.5	6.77E-05	0.00244	3.20
Fortmann 1998, Paint A, high ventilation	0.053	0.0256	1	4.87E-05	0.00244	2.30

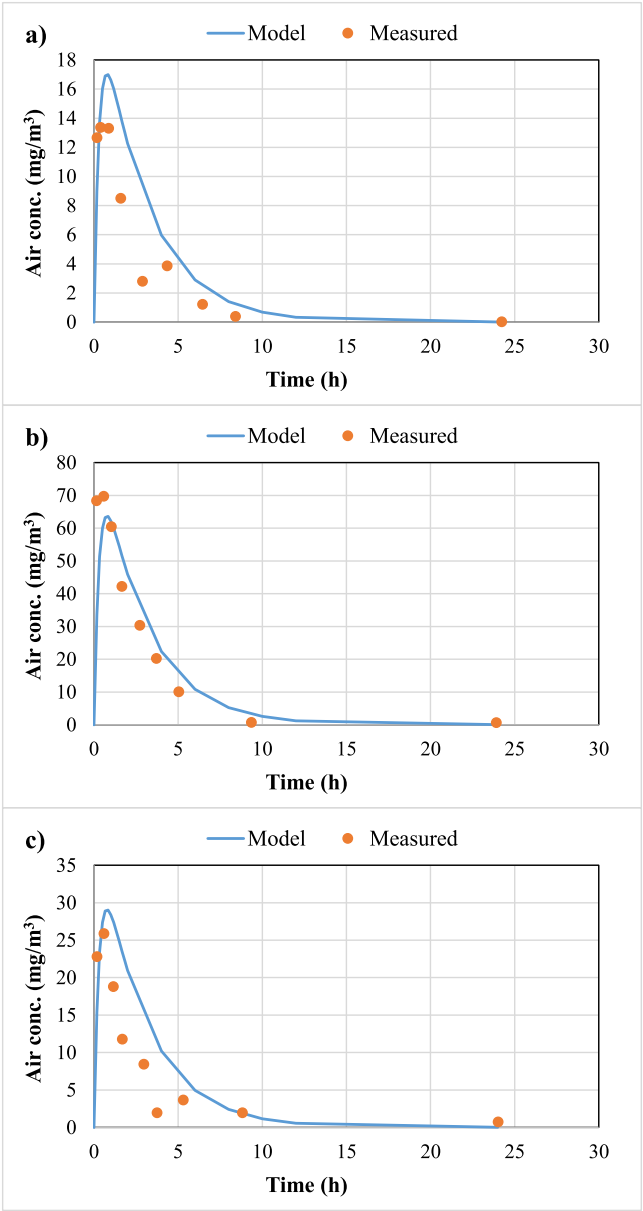


Fig. A2. Comparison between modeled and measured air concentrations of MEKO in small test chambers resulted from the MEKO emissions from a) alkyd paint 1; b) alkyd paint 2; and c) alkyd paint 3 [26].

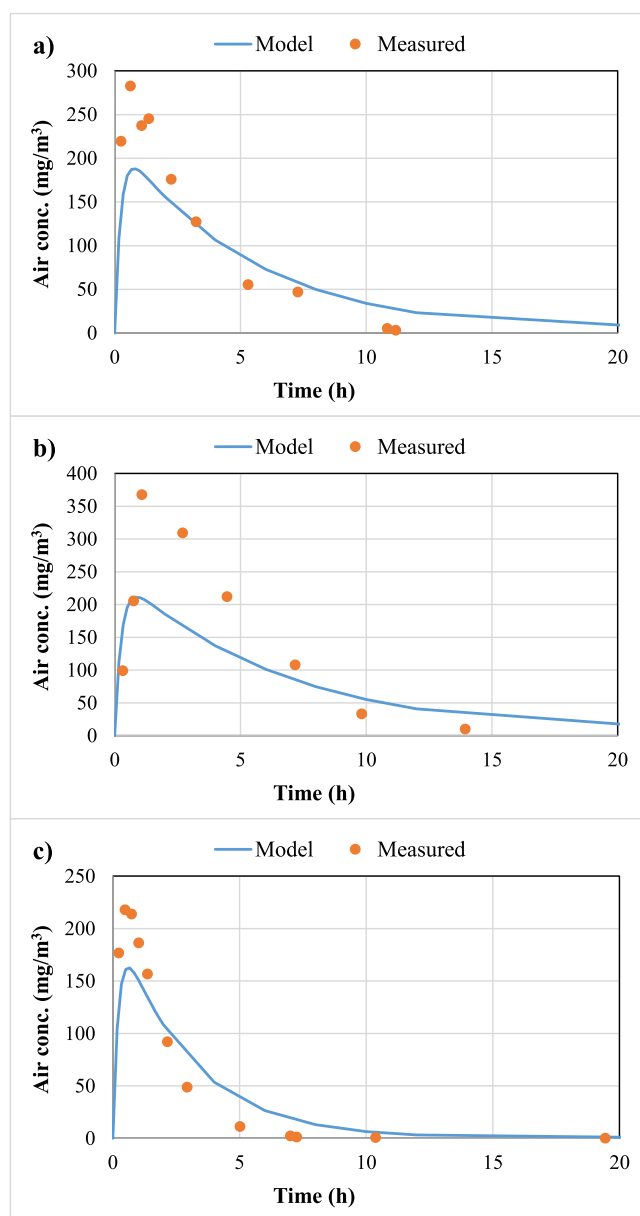


Fig. A3. Comparison between modeled and measured air concentrations of decane in small test chambers resulted from the decane emissions from a) alkyd paint A with standard conditions; b) alkyd paint A with a thick layer; and c) alkyd paint A with a high air ventilation rate [25].

A6. Human exposure and risk to chemicals in paints via dermal exposures

A6.1 Dermal gaseous uptake

Figure A4 provides a detailed view of dermal gaseous exposure and toxicity results per effect type for the chemicals in water-based and solvent-based paints. The diagonal lines in each plot represent equi-hazard quotient (HQ) or equi-incremental lifetime cancer risk (ILCR).

As compared to Fig. 2 of the main text, the risk from dermal gaseous exposure is clearly lower than that from inhalation exposure. Just one chemical has $HQ > 10$ for general non-cancer effects for the household adult resident (Figure A4-D), and no chemicals have $HQ > 100$ for the painter (Figure A4-A). Similarly, for non-cancer reproductive or developmental effects, most chemicals have $HQs < 1$ (Figure A4-B & E). In terms of cancer effects, the number of chemicals with $ILCR > 10^{-4}$ is much smaller than that for inhalation exposure (Figure A4-C & F).

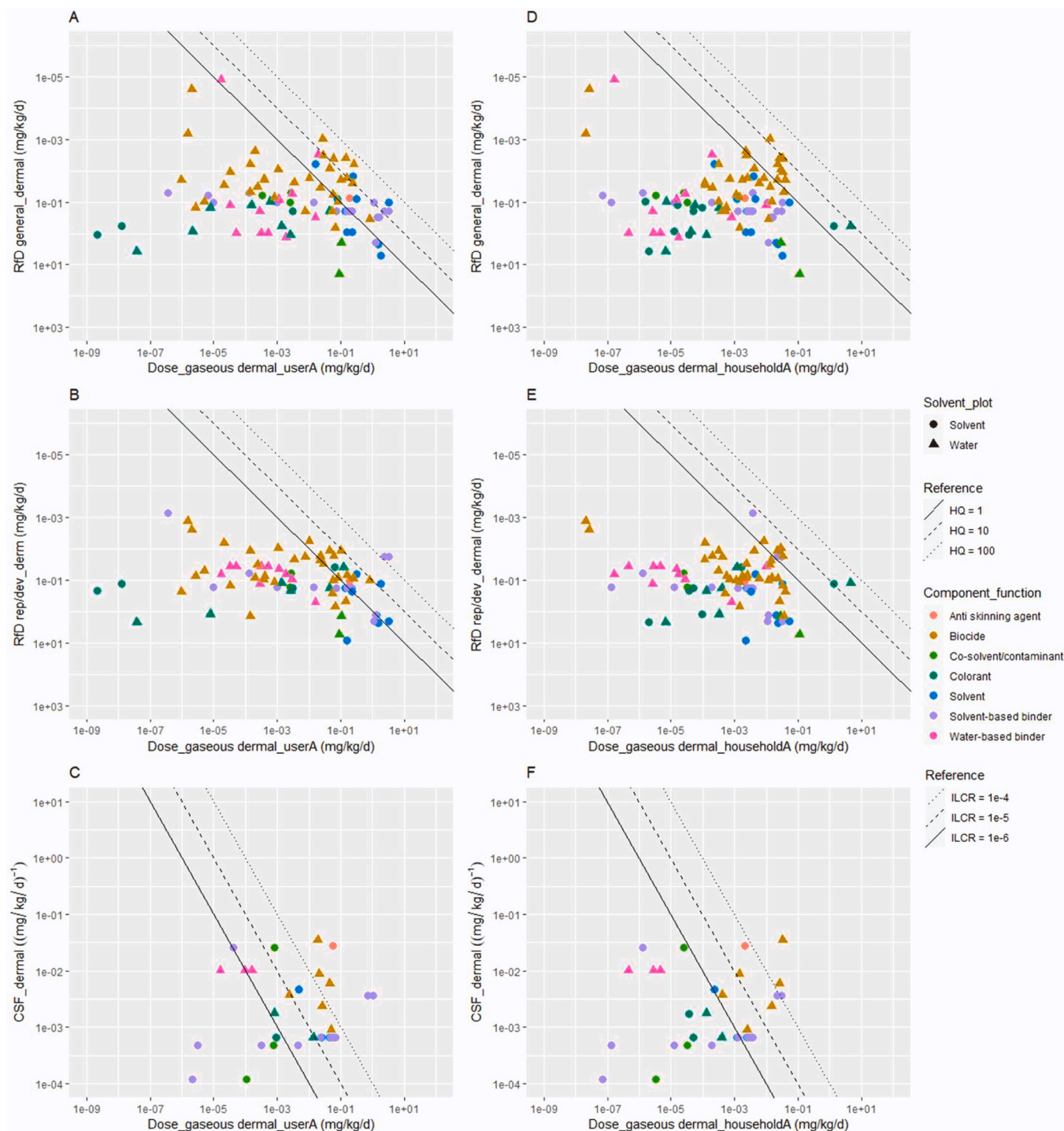


Fig. A4. Reference doses (RfDs) for non-cancer effects (A-B, D-E) and cancer slope factors (CSFs) (C, F) as a function of dermal gaseous exposure doses for chemicals in water-based paint and solvent-based paint (xylenes as the solvent) for the painter (A-C) and the household adult resident (D-F). The exposure duration is 1 year for the household adult including and following one painting event and a daily exposure to a 1 day painting event for the painter. HQ: hazard quotient. ILCR: incremental lifetime cancer risk. The ILCR for the painter is adjusted for 200 working days per year and 40 working years per lifetime of 70 years.

A6.2 Direct dermal contact

Figure A5 provides a detailed view of dermal contact exposure and toxicity results per effect type for the chemicals in water-based and solvent-based paints. Dermal contact exposure is only applicable for the painter. The diagonal lines in each plot represent equi-hazard quotient (HQ) or equi-incremental lifetime cancer risk (ILCR).

As compared to Fig. 2 of the main text and Figure A4 above, the risk from dermal contact exposure is negligible for most chemicals. Only 1 chemical has an HQ > 1 for general non-cancer effects for the painter (Figure A5-A). Also, for non-cancer reproductive or developmental effects, only 2 chemicals have HQs > 1 (Figure A5-B). In terms of cancer effects, only 2 chemicals have ILCR > 10⁻⁴ (Figure A5-C).

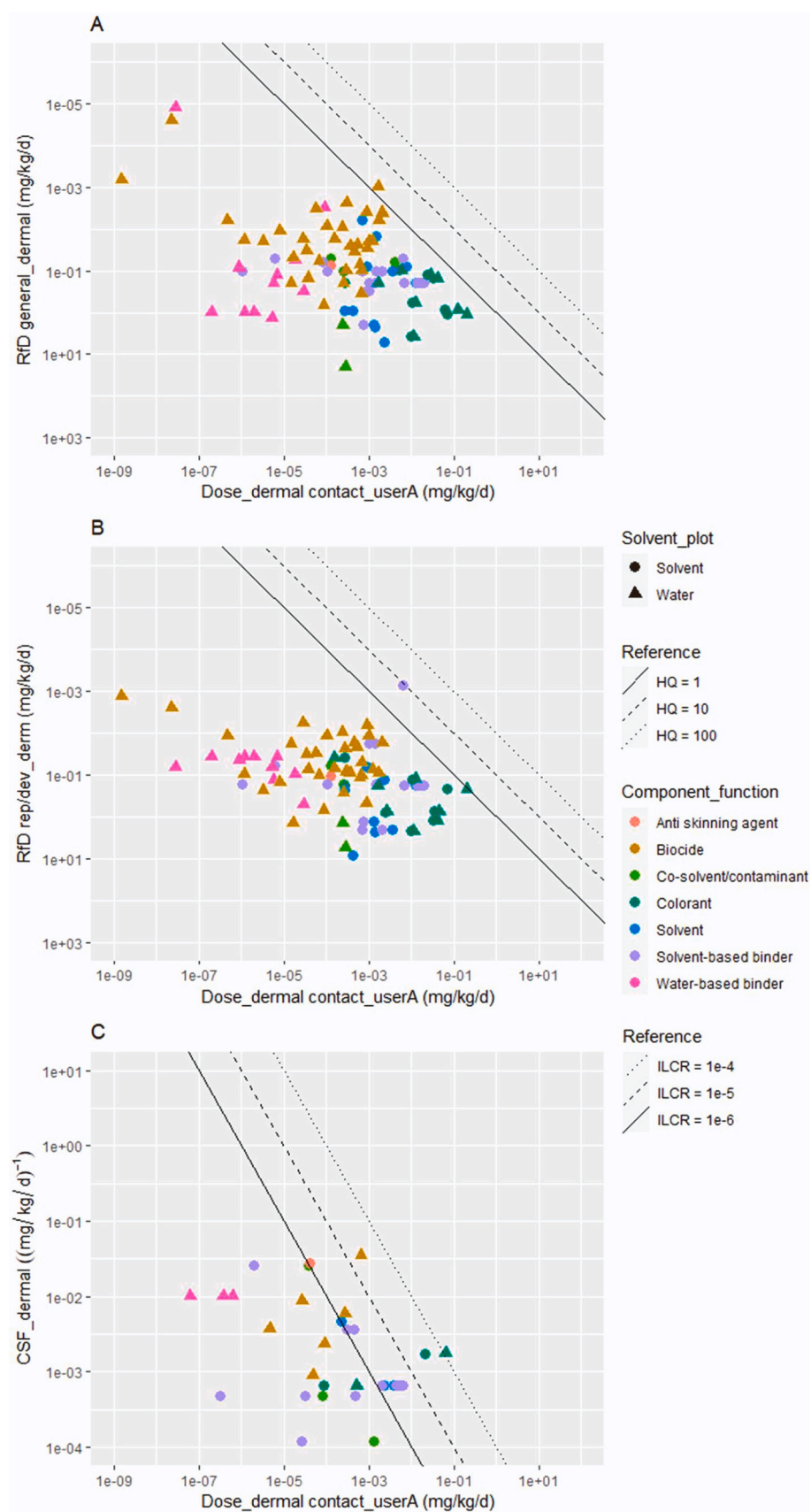


Fig. A5. Reference doses (RfDs) for non-cancer effects (A-B) and cancer slope factors (CSFs) (C) as a function of dermal contact exposure doses for chemicals in water-based paint and solvent-based paint (xylenes as the solvent) for the painter. The results reflect a daily exposure to a 1 day painting event for the painter. HQ: hazard quotient. ILCR: incremental lifetime cancer risk. The ILCR for the painter is adjusted for 200 working days per year and 40 working years per lifetime of 70 years.

A7. Maximum Acceptable Concentrations and Chemicals of Concern in paints

Figures A6 and A7 present the actual chemical mass fractions, cancer and non-cancer MAC_{HTS} and HCRs for the chemicals in solvent-based paints and water-based paints, respectively. The MAC_{HTS} and HCRs correspond to the painter for a daily exposure to freshly applied paints.

Table A3-A6 list the identified chemicals of concern (CoCs) with $HCR > 1$ in solvent-based paint and water-based paint for the household adult resident and the painter. The “Notes” column indicate the specific type of binder polymer that the target chemical is used in.

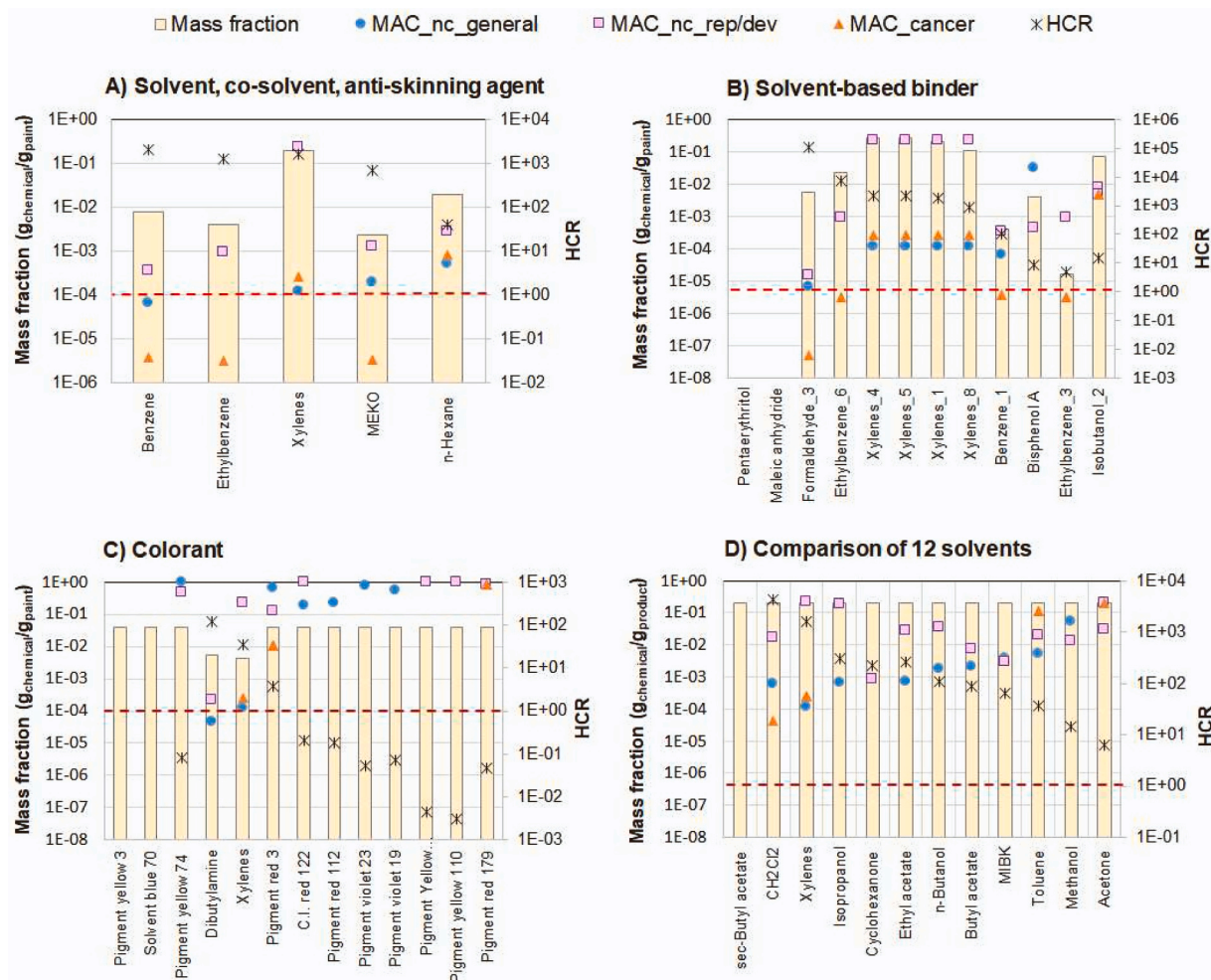


Fig. A6. Actual chemical mass fraction of chemicals in solvent-based paint (yellow bars) compared to maximum chemical content (MAC_{HTS}) for cancer (orange triangles), non-cancer general effects (blue circles) and non-cancer reproductive/developmental effects (purple squares) (left axis), and resulting Hazard Content Ratios (HCR) (black stars - right axis), for (A) solvent, co-solvent and anti-skinning agent, (B) solvent-based binder, (C) colorant, and (D) comparison of 12 solvents. The actual chemical mass fractions in paints with very dark shade colors are shown (Table 1 in main text). Results in (A)(B)(C) are calculated for xylene as the solvent. Results are for the painter performing paint application daily.

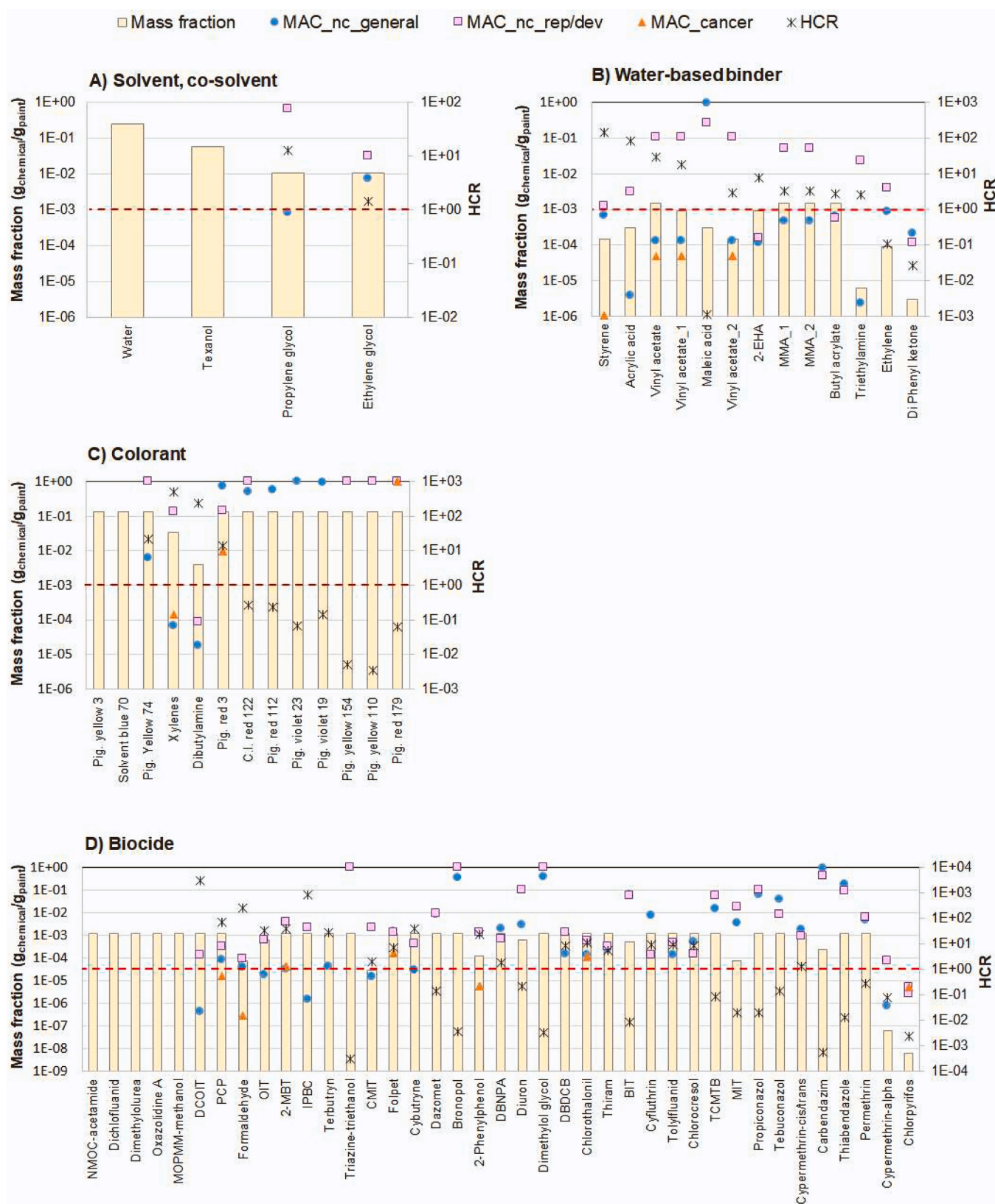


Fig. A7. Actual chemical mass fraction of chemicals in solvent-based paint (yellow bars) compared to maximum chemical content (MAC_{HTS}) for the painter performing daily paint application for cancer (orange triangles), non-cancer general effects (blue circles) and non-cancer reproductive/developmental effects (purple squares) for the chemicals in water-based paint (left axis), and resulting Hazard Content Ratios (HCR) (black stars - right axis), for (A) solvent and co-solvent, (B) water-based binder, (C) colorant and (D) biocide. The actual chemical mass fractions in paints with very dark shade colors are shown (Table 1 in main text).

Table A3

List of CoCs in solvent-based paint for the household adult resident over 1 year.

CAS	Chemical	Function	Residual monomer	MAC	MAC endpoint	Actual content in paint	HCR	Notes
6358-31-2	Pigment yellow 74	Colorant	No	1.96E-06	noncancer-general	4.00E-02	2.04E+ 04	
50-00-0	Formaldehyde	Solvent-based binder	Yes	1.69E-06	cancer	5.43E-03	3.20E+ 03	Alkylated melamine urea formaldehyde resin (CYMEL MI-97-IX)
100-41-4	Ethylbenzene	Solvent-based binder	No	7.50E-05	cancer	2.25E-02	3.00E+ 02	Alkylated melamine urea formaldehyde resin (CYMEL MI-97-IX)
71-43-2	Benzene	Co-solvent/contaminant	No	1.20E-04	cancer	7.80E-03	6.49E+ 01	
1330-20-7	Xylenes	Solvent-based binder	No	4.78E-03	cancer	2.71E-01	5.67E+ 01	modified alkyd resin with high styrene content, based on dehydrated castor and linoleic rich oils, Long oil urethane alkyd resin
1330-20-7	Xylenes	Solvent-based binder	No	4.78E-03	cancer	2.71E-01	5.67E+ 01	
100-41-4	Ethylbenzene	Co-solvent/contaminant	No	7.50E-05	cancer	3.90E-03	5.20E+ 01	
1330-20-7	Xylenes	Solvent-based binder	No	4.78E-03	cancer	2.13E-01	4.46E+ 01	Short oil non air drying alkyd resin
1330-20-7	Xylenes	Solvent	No	4.78E-03	cancer	1.95E-01	4.08E+ 01	
96-29-7	MEKO	Anti skinning agent	No	8.84E-05	cancer	2.25E-03	2.55E+ 01	
1330-20-7	Xylenes	Solvent-based binder	No	4.78E-03	cancer	1.04E-01	2.17E+ 01	Alkylated melamine urea formaldehyde resin (CYMEL MI-97-IX)
80-05-7	Bisphenol A	Solvent-based binder	Yes	7.51E-04	noncancer-rep/dev	3.88E-03	5.16E+ 00	Solid, bisphenol-A based Araldite epoxy resin of medium molecular weight
71-43-2	Benzene	Solvent-based binder	No	1.20E-04	cancer	3.88E-04	3.22E+ 00	Long oil urethane alkyd resin
111-92-2	Dibutylamine	Colorant	No	1.83E-03	noncancer-general	5.53E-03	3.02E+ 00	

Table A4

List of CoCs in water-based paint for the household adult resident over 1 year.

CAS	Chemical	Function	Residual monomer	MAC	MAC endpoint	Actual content in paint	HCR	Notes
6358-31-2	Pig. Yellow 74	Colorant	No	1.96E-06	noncancer-general	1.35E-01	6.88E+ 04	
64359-81-5	DCOIT	Biocide	No	1.85E-06	noncancer-general	1.25E-03	6.75E+ 02	
87-86-5	PCP	Biocide	No	1.01E-05	cancer	1.25E-03	1.24E+ 02	
50-00-0	Formaldehyde	Biocide	No	1.69E-06	cancer	7.50E-05	4.43E+ 01	
26530-20-1	OIT	Biocide	No	2.81E-05	noncancer-general	6.25E-04	2.22E+ 01	
149-30-4	2-MBT	Biocide	No	7.24E-05	cancer	1.25E-03	1.73E+ 01	
55406-53-6	IPBC	Biocide	No	7.29E-05	noncancer-general	1.25E-03	1.71E+ 01	
57-55-6	Propylene glycol	Co-solvent/contaminant	No	6.93E-04	noncancer-general	1.10E-02	1.59E+ 01	
886-50-0	Terbutryn	Biocide	No	9.34E-05	noncancer-general	1.20E-03	1.28E+ 01	
1330-20-7	Xylenes	Colorant	No	4.79E-03	cancer	3.38E-02	7.05E+ 00	
4719-04-4	Triazine-triethanol	Biocide	No	2.33E-04	noncancer-general	1.25E-03	5.35E+ 00	
26172-55-4	CMIT	Biocide	No	6.45E-06	noncancer-general	2.78E-05	4.30E+ 00	
133-07-3	Folpet	Biocide	No	2.98E-04	cancer	1.25E-03	4.19E+ 00	
100-42-5	Styrene	Water-based binder	Yes	3.63E-05	cancer	1.50E-04	4.14E+ 00	Styrene acrylic co polymer emulsion
79-10-7	Acrylic acid	Water-based binder	Yes	7.30E-05	noncancer-general	3.00E-04	4.11E+ 00	Sodium salt of polymeric carboxylic acid (Anucryl 80)
28159-98-0	Cybutryne	Biocide	No	3.46E-04	noncancer-general	1.25E-03	3.62E+ 00	
533-74-4	Dazomet	Biocide	No	3.94E-04	noncancer-general	1.25E-03	3.17E+ 00	

(continued on next page)

Table A4 (continued)

CAS	Chemical	Function	Residual monomer	MAC	MAC endpoint	Actual content in paint	HCR	Notes
52-51-7	Bronopol	Biocide	No	4.33E-04	noncancer-general	1.25E-03	2.89E+ 00	
111-92-2	Dibutylamine	Colorant	No	1.83E-03	noncancer-general	4.05E-03	2.22E+ 00	
90-43-7	2-Phenylphenol	Biocide	No	7.94E-05	cancer	1.25E-04	1.57E+ 00	
10222-01-2	DBNPA	Biocide	No	8.56E-04	noncancer-rep/dev	1.25E-03	1.46E+ 00	
330-54-1	Diuron	Biocide	No	6.11E-04	noncancer-general	6.25E-04	1.02E+ 00	

Table A5

List of CoCs in solvent-based paint for the painter performing paint application daily.

CAS	Chemical	Function	Residual monomer	MAC	MAC endpoint	Actual content in paint	HCR	Notes
50-00-0	Formaldehyde	Solvent-based binder	Yes	4.93E-08	cancer	5.43E-03	1.10E+ 05	Alkylated melamine urea formaldehyde resin (CYMEL MI-97-IX)
100-41-4	Ethylbenzene	Solvent-based binder	No	3.10E-06	cancer	2.25E-02	7.25E+ 03	Alkylated melamine urea formaldehyde resin (CYMEL MI-97-IX)
1330-20-7	Xylenes	Solvent-based binder	No	1.20E-04	noncancer-general	2.71E-01	2.25E+ 03	modified alkyd resin with high styrene content, based on dehydrated castor and linoleic rich oils,
1330-20-7	Xylenes	Solvent-based binder	No	1.20E-04	noncancer-general	2.71E-01	2.25E+ 03	Long oil urethane alkyd resin
71-43-2	Benzene	Co-solvent/contaminant	No	3.73E-06	cancer	7.80E-03	2.09E+ 03	
1330-20-7	Xylenes	Solvent-based binder	No	1.20E-04	noncancer-general	2.13E-01	1.77E+ 03	Short oil non air drying alkyd resin
1330-20-7	Xylenes	Solvent	No	1.20E-04	noncancer-general	1.95E-01	1.62E+ 03	
100-41-4	Ethylbenzene	Co-solvent/contaminant	No	3.10E-06	cancer	3.90E-03	1.26E+ 03	
1330-20-7	Xylenes	Solvent-based binder	No	1.20E-04	noncancer-general	1.04E-01	8.62E+ 02	Alkylated melamine urea formaldehyde resin (CYMEL MI-97-IX)
96-29-7	MEKO	Anti skinning agent	No	3.28E-06	cancer	2.25E-03	6.86E+ 02	
111-92-2	Dibutylamine	Colorant	No	4.65E-05	noncancer-general	5.53E-03	1.19E+ 02	
71-43-2	Benzene	Solvent-based binder	No	3.73E-06	cancer	3.88E-04	1.04E+ 02	Long oil urethane alkyd resin
110-54-3	n-Hexane	Co-solvent/contaminant	No	5.00E-04	noncancer-general	1.95E-02	3.90E+ 01	
1330-20-7	Xylenes	Colorant	No	1.20E-04	noncancer-general	4.25E-03	3.53E+ 01	
78-83-1	Isobutanol	Solvent-based binder	No	4.90E-03	noncancer-rep/dev	6.98E-02	1.42E+ 01	Alkylated melamine urea formaldehyde resin (CYMEL MI-97-IX)
80-05-7	Bisphenol A	Solvent-based binder	Yes	4.36E-04	noncancer-rep/dev	3.88E-03	8.89E+ 00	Solid, bisphenol-A based Araldite epoxy resin of medium molecular weight
100-41-4	Ethylbenzene	Solvent-based binder	No	3.10E-06	cancer	1.55E-05	5.00E+ 00	Long oil urethane alkyd resin
2425-85-6	Pigment red 3	Colorant	No	1.06E-02	cancer	4.00E-02	3.77E+ 00	

Table A6

List of CoCs in water-based paint for the painter performing paint application daily.

CAS	Chemical	Function	Residual monomer	MAC	MAC endpoint	Actual content in paint	HCR	Notes
64359-81-5	DCOIT	Biocide	No	4.02E-07	noncancer-general	1.25E-03	3.11E+ 03	
55406-53-6	IPBC	Biocide	No	1.45E-06	noncancer-general	1.25E-03	8.64E+ 02	
1330-20-7	Xylenes	Colorant	No	6.65E-05	noncancer-general	3.38E-02	5.07E+ 02	
50-00-0	Formaldehyde	Biocide	No	2.93E-07	cancer	7.50E-05	2.56E+ 02	

(continued on next page)

Table A6 (continued)

CAS	Chemical	Function	Residual monomer	MAC	MAC endpoint	Actual content in paint	HCR	Notes
111-92-2	Dibutylamine	Colorant	No	1.75E-05	noncancer-general	4.05E-03	2.31E+ 02	
100-42-5	Styrene	Water-based binder	Yes	1.06E-06	cancer	1.50E-04	1.42E+ 02	Styrene acrylic co polymer emulsion
79-10-7	Acrylic acid	Water-based binder	Yes	3.67E-06	noncancer-general	3.00E-04	8.18E+ 01	Sodium salt of polymeric carboxylic acid (Anucryl 80)
87-86-5	PCP	Biocide	No	1.71E-05	cancer	1.25E-03	7.33E+ 01	
28159-98-0	Cybutryne	Biocide	No	3.03E-05	noncancer-general	1.25E-03	4.13E+ 01	
149-30-4	2-MBT	Biocide	No	3.22E-05	noncancer-general	1.25E-03	3.88E+ 01	
26530-20-1	OIT	Biocide	No	1.81E-05	noncancer-general	6.25E-04	3.46E+ 01	
108-05-4	Vinyl acetate	Water-based binder	Yes	5.00E-05	cancer	1.50E-03	3.00E+ 01	Vinyl acrylic co polymer
886-50-0	Terbutryn	Biocide	No	4.23E-05	noncancer-general	1.20E-03	2.84E+ 01	
90-43-7	2-Phenylphenol	Biocide	No	5.42E-06	cancer	1.25E-04	2.31E+ 01	
6358-31-2	Pig. Yellow 74	Colorant	No	6.38E-03	noncancer-general	1.35E-01	2.12E+ 01	
108-05-4	Vinyl acetate	Water-based binder	Yes	5.00E-05	cancer	9.00E-04	1.80E+ 01	Vinyl acetate acrylic co polymer
2425-85-6	Pig. red 3	Colorant	No	9.44E-03	cancer	1.35E-01	1.43E+ 01	
57-55-6	Propylene glycol	Co-solvent/contaminant	No	8.67E-04	noncancer-general	1.10E-02	1.27E+ 01	
1897-45-6	Chlorothalonil	Biocide	No	1.11E-04	cancer	1.25E-03	1.13E+ 01	
68359-37-5	Cyfluthrin	Biocide	No	1.37E-04	noncancer-rep/dev	1.25E-03	9.13E+ 00	
731-27-1	Tolylfluanid	Biocide	No	1.38E-04	noncancer-general	1.25E-03	9.07E+ 00	
35691-65-7	DBDCB	Biocide	No	1.46E-04	noncancer-general	1.25E-03	8.53E+ 00	
59-50-7	Chlorocresol	Biocide	No	1.51E-04	noncancer-rep/dev	1.25E-03	8.26E+ 00	
103-11-7	2-EHA	Water-based binder	Yes	1.14E-04	noncancer-general	9.00E-04	7.90E+ 00	Vinyl acetate acrylic co polymer
133-07-3	Folpet	Biocide	No	1.69E-04	cancer	1.25E-03	7.39E+ 00	
137-26-8	Thiram	Biocide	No	2.33E-04	noncancer-general	1.25E-03	5.37E+ 00	
80-62-6	MMA	Water-based binder	Yes	4.64E-04	noncancer-general	1.50E-03	3.23E+ 00	Styrene acrylic co polymer emulsion
80-62-6	MMA	Water-based binder	Yes	4.64E-04	noncancer-general	1.50E-03	3.23E+ 00	Acrylic co polymer emulsion
108-05-4	Vinyl acetate	Water-based binder	Yes	5.00E-05	cancer	1.50E-04	3.00E+ 00	Vinyl acetate ethylene co polymer
141-32-2	Butyl acrylate	Water-based binder	Yes	5.49E-04	noncancer-rep/dev	1.50E-03	2.73E+ 00	Vinyl acrylic co polymer
121-44-8	Triethylamine	Water-based binder	Yes	2.27E-06	noncancer-general	6.00E-06	2.64E+ 00	Aliphatic fatty acid modified anionic polyurethane dispersion
26172-55-4	CMIT	Biocide	No	1.45E-05	noncancer-general	2.78E-05	1.92E+ 00	
10222-01-2	DBNPA	Biocide	No	6.88E-04	noncancer-rep/dev	1.25E-03	1.82E+ 00	
107-21-1	Ethylene glycol	Co-solvent/contaminant	No	7.67E-03	noncancer-general	1.10E-02	1.43E+ 00	
52315-07-8	Cypermethrin-cis/trans	Biocide	No	9.36E-04	noncancer-rep/dev	1.25E-03	1.34E+ 00	

A8. Exposure dynamics to VOC vs. SVOC

To discuss the dynamic of exposures for the different population groups and for different types of chemicals, Figure A8 compares the inhalation exposure between the painter and the household adult resident for two example chemicals: formaldehyde as a VOC and Pigment Red 3 as an SVOC, contrasting the exposure on the first day and a yearly average exposure over the entire year. For formaldehyde (Figure A8-A), when we consider the 1-day duration which mainly corresponds to the wet phase, the daily inhalation exposure dose of formaldehyde is 1.32 mg/kg/d for the painter and is only slightly higher than that for the household adult resident (1.21 mg/kg/d), because chemicals are first emitted to the near-person air and expose the painter, then are transferred to the far-person air and expose the household adult, and finally are ventilated outdoors. Since we consider an airtight building with relatively low ventilation of 0.79 per hour, the far-person air has time to equilibrate with the near-person air, so the inhalation exposure

for the household adult is not substantially different from the painter during the first day. However, over long term the resident's exposure dose of formaldehyde drops to 0.012 mg/kg/d, about 60 times lower than the painter's exposure (0.72 mg/kg/d) which is only reduced by the proportion of working days, since the painter is performing paint application every day and is thus receiving the first-day dose every day.

In contrast for Pigment Red 3 (Figure A8-B) for the first day during the painting period, the dose is very low (4.5×10^{-8} mg/kg/d), 8 orders of magnitude lower than that of formaldehyde. This is because formaldehyde is highly volatile which is mostly volatilized during the wet phase, so the exposure to formaldehyde is highest at the beginning of the paint application. Pigment Red 3 is an SVOC which is almost not volatilized during the wet phase, leading to very low inhalation exposure during the first day. When we consider a 1-year duration after the paint application, the daily inhalation exposure dose of Pigment Red 3 during the dry phase raises by 4 orders of magnitude to 7.6×10^{-4} mg/kg/d for the adult resident, whereas it remains low for the painter who is mostly exposed to wet phase paints.

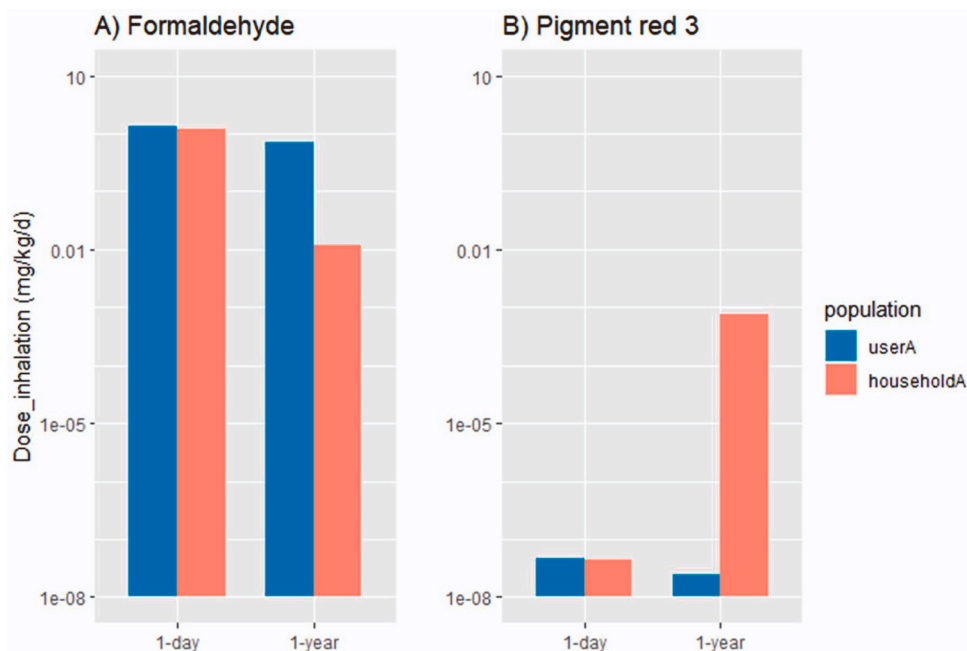


Fig. A8. Comparison of the inhalation exposure dose between the painter and the household adult resident for A) formaldehyde and B) pigment red 3 in solvent-based paint using xylenes as the solvent, considering exposure during the 1st day and average exposure during the entire year after paint application.

Appendix B. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2023.133145](https://doi.org/10.1016/j.jhazmat.2023.133145).

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