



Field investigation on the removal of formaldehyde in indoor air



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ABSTRACT

The development of indoor air quality models requires to take into account the indoor removal processes (or “indoor sinks”) as accurately as the indoor concentrations. Field experiments were conducted in residential rooms to assess the indoor decay rate constant and deposition velocity of formaldehyde and to investigate the nature of these removal processes. The indoor decay rate constant and deposition velocity were respectively found to $0.34 \pm 0.07 \text{ h}^{-1}$ and $(2.53 \pm 0.51) \times 10^{-3} \text{ cm s}^{-1}$, indicating a high potential of depletion and therefore a low persistence of this compound in indoor environment. Considering only the impact of indoor sinks, a short indoor half-life of 2 h for formaldehyde was assessed. This high removal of formaldehyde indoors in comparison with other volatile organic compounds and its dependence with the absolute humidity suggest that the adsorption on the indoor surfaces is not the only loss process for formaldehyde. The transfer of formaldehyde to the aqueous films present onto the indoor surfaces and on the surface of airborne particles was proposed as a possible removal pathway in real indoor conditions and discussed.

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1. Introduction

Assessing the potential impacts of air pollutant exposure on human health requires an understanding of not only the range of indoor levels but also the contributions of sources and indoor sinks on the concentrations under typical household conditions. Different types of miniature emission chambers, like the passive flux samplers [1,2] or standard FLEC[®] emission cell coupled with an active sampling method [3] or with solid-phase microextraction [4] are now available for in situ measurement of the flux of organic compounds emitted from various kinds of indoor materials. In contrast, few data exist on the removal of organic compounds from indoor sinks. The improvement of the prediction of indoor air quality models requires a better characterization of indoor sinks and of their impact on the concentration levels in real indoor conditions [5].

Most approaches to modeling pollutant concentrations in indoor air take into account four basic physical/chemical processes that describe the behavior of chemicals in a building interior. Two of these processes increasing indoor concentration levels are the flow of outdoor chemicals into the interior environment and the rate at which pollutants are emitted by indoor sources. Two other

processes decreasing indoor contaminant levels are the flow of indoor air out of the interior environment and the net rate of removal processes (or “indoor sinks”). These latter processes include heterogeneous and gas-phase chemical reactions which may produce secondary compounds of concern; changes in gas-particle partitioning; and sorptive interactions between gases and interior material surfaces. Understanding these dynamic processes and their contribution are essential for predicting indoor concentrations.

To simply set in equation all of these processes, Dockery and Spengler [6] proposed as a first approach to use a single mass balance model. This model assumes that the building interior is a single and well-mixed chemical volume. The secondary pollutants produced, for example, in photochemical reactions are not described by this simple mass-balance approach. The mathematical expression for the change in mass concentration of indoor contaminant is:

$$dC = PaC_{\text{out}}dt + \frac{Q}{V}dt - (a + k)Cdt \quad (1)$$

where C : average indoor contaminant concentration ($\mu\text{g m}^{-3}$); C_{out} : outdoor contaminant concentration ($\mu\text{g m}^{-3}$); P : fraction of outdoor contaminants that penetrates the shell (unitless) ($1 = 100\%$ penetration); a : air exchange rate (h^{-1}), Q : indoor source strength, i.e., the mass released per unit time from all indoor sources ($\mu\text{g h}^{-1}$); and V : room volume (m^3) and k : net rate of removal

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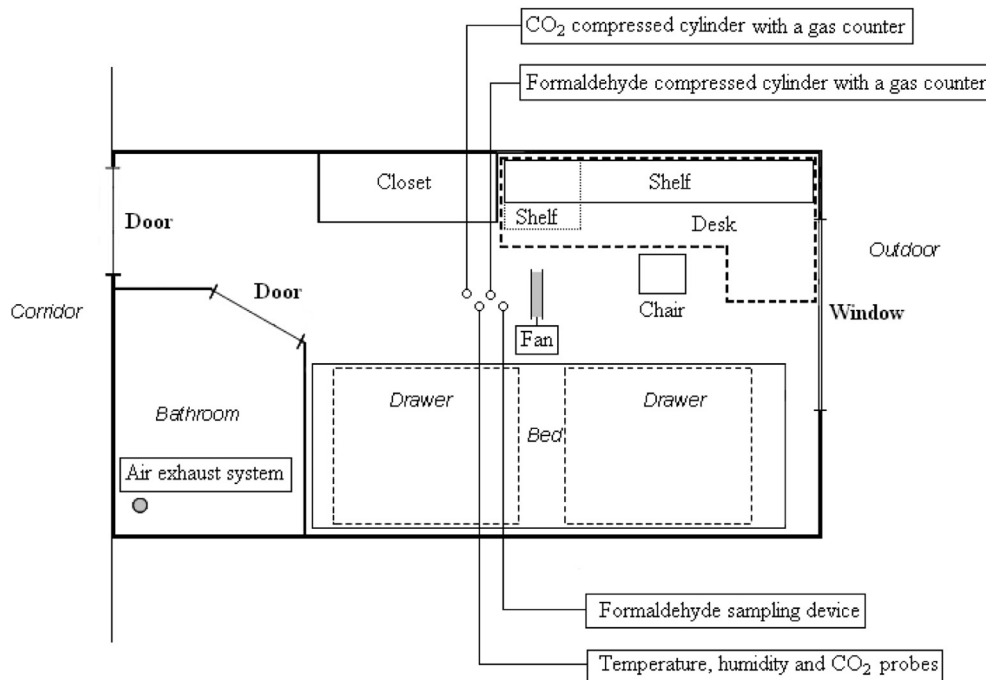


Fig. 1. Schematic of residential room (layout view) and instrumentation.

processes other than air flow, i.e., indoor decay rate constant (h^{-1}) representing the sum of surface removal rate constant and reaction rate constant (both surface removal and reaction were assumed to follow the first order kinetics).

When a steady-state regime is achieved in the system, then C , C_{out} , P , a , k and Q are constant; equation (1) can be solved for C to give:

$$C = \left[\frac{aP}{(a+k)} \right] C_{\text{out}} + \frac{Q}{V} \left[\frac{1}{(a+k)} \right] \quad (2)$$

Data about indoor concentrations and application of mass-balance models suggest that for many pollutants, indoor sinks induce decay rates comparable to or greater than the air exchange rate [7]. Generally, the most reactive compounds have the highest decay rate constants, like ozone (from 1.44 to 2.6 h^{-1}) and nitrogen dioxide (from 0.8 to 1.3 h^{-1}) assuming a ratio Area of surfaces (A) by room Volume (V) equal to 2 m^{-1} [7]. The k value of formaldehyde is poorly documented in the literature. Traynor et al. [8] reported a k value amounted to $0.40 \pm 0.24 \text{ h}^{-1}$ ($n = 5$ runs) deduced to the analysis of formaldehyde concentration decay from gas-stove emission experiments in an environmental chamber of high volume (27 m^3).

This value was largely used for indoor air quality modeling and numerous studies revealed that the formaldehyde concentration in real indoor air cannot be calculated only from indoor emission rates and air exchange rate. The concentration decay relative to indoor removal processes was taken into account to provide an accurate prediction of formaldehyde concentrations by modeling [9–11]. No data was reported to our knowledge about the assessment of this decay rate relative to indoor sinks in real indoor environments.

The objective of this study was to determine the decay rate constant for formaldehyde in several rooms by an experimental approach combining the measurement of air exchange rate and the follow-up of formaldehyde concentration decay in the rooms. The value obtained for this constant was discussed and compared with those of other indoor contaminants.

2. Experimental

2.1. Characteristics of rooms

The decay rate constant for formaldehyde was assessed in two similar unoccupied student rooms of a four-storeyed residence built in 1998. A schematic of these rooms is shown in Fig. 1. Their volume and surface area are 26 m^3 and 11 m^2 , respectively. The floor covering is linoleum. The walls are in sheet plaster that has been papered with painted fibre cloth. The ceiling is in painted sheet plaster. These rooms are furnished by a bed, a closet, a writing desk and a chair. The furnishing materials are plywood for the closet and particleboard for the writing desk, bed and chair. Each room has one window with an air entry, one front door leading out into a corridor and one inner door separating the room from the bathroom. Each room is filled with an air exhaust system located in the bathroom in continuous running during the field experiments.

These rooms have as specific characteristic a high ratio of furniture area to the sum of building surfaces (sum of floor, walls, doors and ceiling) close to 0.8 by comparison with those of other indoor environments.

2.2. Instrumentation, protocol and calculation

The measurement protocol was based on the simultaneous determination of air exchange rate (a) and total rate of removal processes including air exchange rate ($a + k$) in a room. The experiment was made in three steps:

Firstly, the injections of CO₂ and formaldehyde were carried out at the center of the room at a height of approximately 1.2 m using a compressed cylinder of CO₂ (at 99%) supplied by Air Liquide-company (Air Liquide, Loos, France) and another compressed gas cylinder containing 100 ppm of formaldehyde in nitrogen manufactured by Messer company (Messer, Mitry Mory, France). Some liters of each gas cylinder were injected using a gas counter in order to raise the CO₂ and formaldehyde concentrations five to ten times.

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