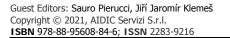


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A Model for the Evaluation of VOCs Abatement by Potted Plants in Indoor Environments

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Phytoremediation of indoor air represents an innovative, cost-effective and eco-friendly strategy to control indoor air quality that could integrate more traditional methods (e.g. ventilation and source control), especially in non-industrial environments. Since the end of the '80s, it is known that simple potted plants used as passive biofilters can remove Volatile Organic Compounds (VOCs) from indoor air even at low concentrations. In the last thirty years, many studies were carried out to quantify the removal rates of specific plant species towards different VOCs and the removal efficiencies that can be obtained introducing plants in a closed environment. Nevertheless, there are no well-assessed methodologies yet.

Considering all the factors that influence indoor air quality is crucial to comprehensively assess potted plants' removal capacity in closed environments. Therefore, basic indoor air models have been expanded and integrated with plants' abatement effect in order to develop an original indoor air model suitable for this purpose. The model consists in a mass balance where VOC concentration depends on the volume considered, on the outdoor pollutant concentration and on the ventilation rate (Air Change per Hour, ACH). At the same time potential indoor sources, and removal rate (sink) due to potted plants are taken into account in the model. The novel depletion term introduced is function of the plant's leaf area and of the VOC removal rate per unit of leaf area. The latter is species-specific and must be derived from experimental studies.

A real-existing copy shop, a non-industrial environment of particular interest, was chosen as a case study. The model was first calibrated on the basis of available measured concentrations without plants and then applied to estimate the removal efficiency obtainable with a defined leaf area of Zamioculcas Zamiifolia on ethylbenzene, xylenes and styrene concentrations (contaminants typically emitted by copiers). The estimated removal efficiencies (0.23 - 4.14% range) show that the significance of plants' removal contribution is strongly related to ventilation, but also show that the model is able to capture main features involved in idoor air quality. It can be concluded that the proposed model wants to be a step forward towards the rational use of potted plants in indoor air mitigation interventions. After defining the VOC to address and the plant species to use, it allows the evaluation of the impact of inserting a given leaf area in a defined indoor environment and the determination of the leaf area necessary to obtain a specific abatement efficiency. Moreover, it could also guide the necessary process of standardisation of the protocols in the field, especially for the experimentations in real environments.

1. Introduction

Indoor pollution is a significant public health problem still too often underestimated. Since the time spent indoors is around 90% in industrialised countries (Klepeis et al., 2001), it is evident that indoor air is a source of exposure that cannot be neglected even when the measured contaminant concentrations are low (Sundell, 2004). Among the various categories of indoor pollutants, Volatile Organic Compounds are of particular interest as they are ubiquitous, cause proven damage to health, and their concentrations are often higher than outdoors (Bruno et al., 2008).

Source control, improved ventilation, and air purification are the main strategies adopted to improve indoor air quality (EPA), but their applicability is not always straightforward. Some relevant aspects to consider are: many indoor sources are still to be identified or studied, making rigorous evaluations difficult; ventilation improvement is not always feasible in existing buildings, particularly old ones; the technologies currently

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available for air purification are not particularly effective for the low VOC concentrations commonly measured indoors (Guieysee et al., 2008).

Starting with Nasa's pioneering experiments (Wolverton et al., 1989), a broad number of species of ornamental plants have been tested in controlled environments to verify and quantify their abatement capacity concerning specific VOCs. It emerged that some plant species have superior VOC purifying abilities and can be used as effective biofilters even at low VOC concentrations (Kim et al., 2018). A determined species' ability to detoxify indoor air is related predominantly to its metabolic and morphologic characteristics, and it is influenced by the type and concentration of the pollutant/s present (Dela Cruz et al., 2014). Moreover, the leaves' VOC uptake seems to play a dominant role in the abatement for most species; in fact, the removal rates are principally defined per unit leaf area (Kim et al., 2018). The experimentations carried out so far that evaluate plant-based VOC removal in real-life settings are few, present a disparity in the protocols used (all the main factors that define indoor air quality are not always taken into consideration) and led to heterogeneous results (Wood et al., 2006; Pegas et al., 2012; Lim et al., 2009).

Using simple potted plants as passive biofilters to mitigate indoor air VOC concentrations integrating more established remediation strategies represents a simple, economical and "green" air purification technique to be evaluated primarily for domestic and non-industrial environments.

Modelling indoor air VOC concentrations is imperative for applying passive biofiltration in real-life settings. This paper presents an original indoor air model that includes the potted plants' removal contribution, allowing the assessing of passive biofilters' removal capacity in the field and the designing of plant-based VOC mitigation interventions. The model was designed starting from existing indoor air models (Moschandreas and Stark, 1978; Nazaroff, 1986; Tichenor et al., 1991).

Recalling that indoor air is a significant exposure route and considering that VOCs can lead in non-industrial occupational settings to a wide variety of adverse health effects even for low concentration exposures (Bernstein et al. 2008), it is evident that non-industrial environments are a particularly interesting target for this kind of interventions. An application of the original model for a real-existing copy shop was performed and is going to be illustrated in section 2.2.

2. Methodology

2.1 Indoor air modeling

Expanding and integrating existing indoor air models (for example, the GIOAP model, Moschandreas and Stark, 1978), a novel indoor air model has been designed. The depletion term introduced considers the abatement effect produced by specific species of plants on indoor VOC concentrations. The main model assumptions are: the volume of indoor air is considered perfectly mixed, the VOC removal is assumed to occur due to adsorption on the plant leaves only, and other physical and chemical removal processes are neglected. A schematic representation of the indoor space according to these assumptions is presented in Figure 1.

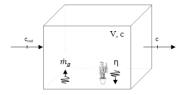


Figure 1: Schematic representation of the indoor space according to the model assumptions

The simple mass balance equation for a single contaminant used in this new model is presented in Eq(1)

$$V \frac{dc}{dt} = \dot{V} c_{out} - \dot{V} c + S - A_l \eta c$$
(1)

with initial condition $c(t_0) = c_0$ and where: *V* is the volume of the indoor space (m^3) ; *V* is the volume flow rate due to ventilation (m^3/h) ; *c* is the indoor pollutant concentration $(\mu g/m^3)$; *c_{out}* the outdoor pollutant concentration $(\mu g/m^3)$; *S* a term representing a generic indoor source for the pollutant $(\mu g/h)$. The last term on the left-hand side of Eq(1) is the novel term introduced and represents the removal rate due to potted plants $(\mu g/h)$. Specifically, the plant VOC uptake is considered a pseudo-first-order mechanism with kinetic constant given by the product of the leaf area, A_l (m²), introduced in the room and the removal rate per unit leaf area η (m/h). The latter can be seen as a deposition/adsorption characteristic velocity of the contaminant on the plant leaves, and it is species and pollutant specific.

A conventional efficiency can be defined as shown in Eq(2) to quantify the plant removal effect

$$\chi = 1 - \frac{c_p}{c} \tag{2}$$

being c_p and c the indoor concentrations in presence and absence of plants, respectively.

Regarding the model application, the removal rate per unit leaf area η is the crucial parameter to determine. The data required are obtainable from experiments in controlled environments carried out to find the removal rates of specific plant species for determined VOCs. More specifically, given the volume of the sealed chamber and leaf area adopted, the variation of the concentration over time in the chamber can be experimentally measured. The removal rate per unit leaf area can then be extrapolated from the regression of these experimental data using the model proposed (Eq(1)) for the sealed chamber and considering only the depletion term. Reuslting Eq(3) and Eq(4) are reported to show the main steps of this calculation.

$$\frac{dc}{dt} = -\eta \frac{A_l}{V} c; \quad c(0) = c_0$$
(3)
$$\frac{c(t)}{c_0} = exp\left(-\eta \frac{A_l}{V} t\right)$$
(4)

2.2 Case study

Non-industrial occupational environments represent particularly interesting targets for the application of plantbased remediation interventions. As printing devices are ubiquitous sources of pollutants that are proven to emit VOCs harmful to human health, in particular, BTEXS (Leovic et al., 1998; Lee et al. 2001; Destaillats and al., 2008), the focus was kept on copy shops. After an accurate cross-analysis of the available studies, a realexisting copy shop was selected. Ethylbenzene, xylene, and styrene were identified as contaminants of interest and the species Zamioculcas Zamiifolia was chosen for the intervention. It was decided to evaluate the effect of introducing 40 medium-sized plants into the confined environment.

Information for the calibration, validation and application of the proposed model was derived from the studies of Betha et al. (2011), Brown (1999) and Sriprapat and Thiravetyan (2013). The site selected is a copy shop (V= 206.55 m³) where the air conditioning system maintains a constant Air Changes per Hour (ACH) during working hours (7.30 a.m. - 9 p.m.) of 1.63 h⁻¹, and during closing hours, it is turned off. A set of selected VOCs was monitored for one month; samples were collected every day before opening (7.30 a.m.), immediately after opening (9 a.m.) and during peak printing hours (12 a.m.). From the results presented in the study of Betha et al. (2011) emerge that, as expected, ethylbenzene, xylene, and styrene concentrations vary with the copy machines' operativity (average values and standard deviations of their measured concentrations are shown in Table 1).

Chemical Compound	Before Printing	First Few Printing	Peak Printing period
ethylbenzene	6.51 ± 3.40	7.95 ± 4.26	10.84 ± 2.89
m,p-xylene	6.71 ± 2.25	6.96 ± 1.93	8.17 ± 0.66
o-xylene/styrene	6.32 ± 1.94	12.4 ± 2.71	11.63 ± 1.74

Table 1: Average values and standard deviations of the concentrations (μ g/m³) of the VOCs of interest measured inside the copy shop at different times of the day (Betha et al., 2011).

It has been necessary to find specific studies on copy machine BTEXS emission rates to correlate variations in measured indoor concentrations and copiers' emission rates. The experimental data obtained in a controlled room for a xerographic copier provided by Brown (1999) gave the basis for quantifying the emission rates in off, idle, and operating conditions (in the latter case, the µg of contaminant emitted per copy are supplied). Model calibration was performed making the following assumptions during predefined time intervals: steady state conditions, negligible outdoor air concentrations and constant ventilation and emission rates. The time intervals defined to perform the analysis can be described as follows: closed period (from 9 p.m. to 7.30 a.m., the copiers are off and air conditioning is off); opening period (from 7.30 a.m. to 9 a.m., the copiers are idle and air conditioning is on); first few printing (period in which the printing is starting; no calculations have been performed due to the lack of data); normal printing period (from 9 a.m. to 7.30 p.m., the copiers are operating and air conditioning is on); closing period (from 7.30 p.m. to 9 p.m., the copiers are idle and air conditioning is on); closing period (from 7.30 p.m. to 9 p.m., the copiers are idle and air conditioning is on); the average VOC emissions of printing devices (Destaillats and al., 2008), it has been considered reasonable to assume the printing devices present in the site as one single

equivalent copier. For the sake of clarity, the data available and informations missing and obtainable from model calibration are presented in Table 2.

Period	Time interval	S (µg/h)	ACH (1/h)
Closed period	9.01 p.m7.30 a.m.	known from Brown (1999)	calibration required
Opening period	7.31 a.m9.00 a.m.	known from Brown (1999)	1.63 (Betha et al., 2011)
First few printing	/	no elements for estimatior	1.63 (Betha et al., 2011)
Normal printing period	9.01 a.m7.30 p.m.	calibration required	1.63 (Betha et al., 2011)
Closing period	7.31 p.m9.00 p.m.	known from Brown (1999)	1.63 (Betha et al., 2011)

Table 2: Data available for the model calibration.

The model (Eq(1)) has been applied backwards to calculate both closed period ventilation rate and average emission rates for each contaminant (S_i , Table 3) in the printing period.

Table 3: Data derived from model calibration.

Period	$s_{ethylbenzene}$ (µg/h)	s _{m,p-xylene} (µg/h)	$s_{o-xylene/styrene} (\mu g/h)$	ACH (1/h)
Closed period	140	130	100	0.092
Opening period	280	310	210	1.63
First few printing	1	1	1	1.63
Normal printing period	4458	3837	2674	1.63
Closing period	280	310	210	1.63

The contaminant concentrations were calculated with the calibrated model to validate the model against measured values. Calculated concentrations are reported as bars in Figure 2, together with the concentrations measured in the copy shop and their relative uncertainties (Betha et al., 2011).

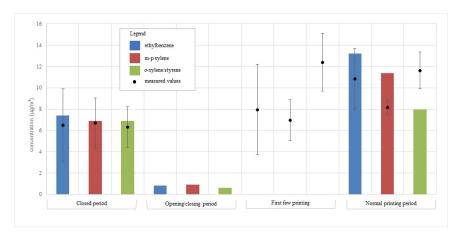


Figure 2: Concentrations values ($\mu g/m^3$) of ethylbenzene, *m*,*p*-xylene and o-xylene/styrene calculated and measured with relative uncertainty range (Betha et al., 2011).

The species Zamioculcas zamiifolia was selected for this application as it is a plant with superior VOC purifying ability (Kim et al., 2018) for the contaminants of interest. This species is particularly suitable for this application as the removal is proven to occur prevalently through the leaves; moreover, it is a very resistant and low maintenance plant. The study of Sriprapat and Thiravetyan (2013) was held as a reference as it provides all the information necessary for the calculation of η . The experiments on this species were carried out in a controlled room. For each measurement, a specimen of Zamioculcas zamiifolia with a leaf area A_l of 0.013 m² was introduced in a 15.6 litres chamber (V) with an initial amount of selected contaminant corresponding to an initial concentration c₀ of 20 ppm. As previously discussed, a plant species' removal efficiency can be derived from regression of experimental data in a sealed chamber using Eq(4) for data fitting. The results obtained for ethylbenzene and xylene/styrene are shown in Table 3.

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Table 3: Zamioculcas zamiifolia removal rates calculated for ethylbenzene and xylene/styrene.

Contaminant	η A,/ V (1/h)	η (m/h)	R^2	
ethylbenzene	0.0297	0.0357	0.945	
xylene/styrene	0.0288	0.0346	0.937	

In order to evaluate the effect of introducing 40 medium-sized plants into the confined environment, each having an estimated leaf area of 0.58 m² ($A_{I,TOT} \approx 23 \text{ m}^2$), the model has been applied to calculate the concentrations in the copy shop c_p and the percentage removal efficiency X in the defined periods.

3. Results and discussion

The removal efficiencies obtained in the copy shop for the three contaminants of interest in the presence of 40 specimens of Zamioculcas Zamiifolia during the night (closed period) and day (opening, normal printing and closing periods) are shown in Figure 3.

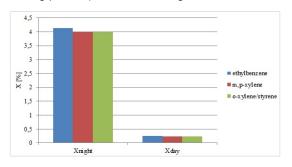


Figure 3 - Night and day removal efficiencies for ethylbenzene, m,p-xylene and o-xylene/styrene obtained by virtually introducing 40 specimens of Zamioculcas zamiifolia ($A_{I,TOT} \approx 23 \text{ m}^2$) into the copy shop.

It is immediately evident that the removal contribution due to potted plants is not significant for the selected contaminants. At night, when the air conditioning is off the efficiencies are around 4%. In all the other periods, the values obtained are around 0.23-0.24%. The main reason for this result is that ventilation plays a fundamental role in VOCs abatement and its thus contribution is much greater than the contribution due to potted plants. Moreover, the large volume of the copy shop (V= 206.55 m³) must be considered since it would require an higher number of plants.

The proposed model can also be used to find the A_i/V ratio necessary to obtain the desired removal efficiency by re-arranging Eq(2). This last application is particularly interesting but requires the definition of concentration threshold values for the VOCs typically found indoor, which are still missing in many cases. Meanwhile, the logical approach would be to lower the concentrations as much as possible by placing a reasonable large number of plants in the indoor space. To give an example and with the sole purpose of showing how the model can be applied, it can be seen that in the site analysed in the case study, under the same assumptions and in the closed period, the leaf area necessary to obtain a 50% ethylbenzene removal is 533 m².

Conclusions

The proposed indoor air model wants to be a tool to approximate the effectiveness of passive biofiltration interventions. The depletion term D, the main novelty introduced, describes the abatement effect produced by potted plants on indoor VOC concentrations. Specifically, the removal is assumed as a pseudo-first-order mechanism with the kinetic constant given by the product of the removal rate per unit leaf area (specific for each plant species and pollutant) and the leaf area introduced in the indoor environment. From the application of the model to a case study, it has been shown that the removal effect of plants is strongly related to ventilation. Introducing 40 specimens of Zamioculcas Zamiifolia in a real existing copy shop, the average removal efficiencies obtainable for the contaminants of interest are higher during closing hours than during the working hours when air conditioning is on. Moreover, as the overall contribution of ventilation is always much greater than the removal contribution of potted plants, the efficiencies are not significant. From these considerations, it is evident that smaller sites with lower air recirculation would be optimal for passive biofiltration interventions.

In conclusion, the new model can be a valuable tool to quantify the effect of introducing a defined leaf area in a closed environment, as presented in the case study and to design passive biofiltration interventions.

To set a more robust basis for the modelling of VOCs abatement by potted plants in indoor environments and, therefore, to open to the possibility of using passive biofiltration as an effective VOC remediation strategy in real settings, it is fundamental to define standardised protocols both for sealed chamber and real-life experimentations.

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