



**HAL**  
open science

## Influence of indoor chemistry on the emission of mVOCs from *Aspergillus niger* molds

C. Kalalian, L. Abis, A. Depoorter, B. Lunardelli, S. Perrier, C. George

### ► To cite this version:

C. Kalalian, L. Abis, A. Depoorter, B. Lunardelli, S. Perrier, et al.. Influence of indoor chemistry on the emission of mVOCs from *Aspergillus niger* molds. *Science of the Total Environment*, 2020, 741 (—), 10.1016/j.scitotenv.2020.140148 . hal-02956437

**HAL Id: hal-02956437**

**<https://hal.science/hal-02956437v1>**

Submitted on 1 Jul 2022

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

# Influence of indoor chemistry on the emission of mVOCs from *Aspergillus niger* molds

Carmen Kalalian<sup>1</sup>, Letizia Abis<sup>1</sup>, Antoine Depoorter<sup>1</sup>, Bastien Lunardelli<sup>1</sup>, Sébastien  
Perrier<sup>1</sup>, Christian George<sup>1</sup> \*

<sup>1</sup> Univ Lyon, Université Claude Bernard Lyon 1, CNRS, IRCELYON, F-69626, Villeurbanne, France.

## Abstract

People spend 80% of their time indoors exposed to poor air quality due to mold growth in humid air as well as human activities (painting, cooking, cleaning, smoking...). To better understand the impact of molds on indoor air quality, we studied the emission of microbial Volatile Organic Compounds (mVOCs) from *Aspergillus niger*, cultivated on malt agar extract, using a high-resolution proton transfer reaction- time of flight- mass spectrometer (PTR-TOF-MS). These emissions were studied for different cultivation time and indoor relative humidities. Our results show that the concentration of the known C<sub>4</sub>-C<sub>9</sub> mVOCs tracers of the microbial activity (like 1-octen-3-ol, 3-methylfuran, 2-pentanone, dimethyl sulfide, dimethyl disulfide, nitromethane, 1,3-octadiene...) was the highest in the early stage of growth. However, these emissions decreased substantially after a cultivation time of 10-14 days and were highly affected by the relative humidity. In addition, the emissions of certain mVOCs was sensitive to indoor light, suggesting an impact of photochemistry on the relative amounts of indoor mVOCs. Based on this study, an estimation of the mVOC concentration for a standard living room was established at different air exchange rates and their indoor lifetimes toward hydroxyl radicals and ozone were also estimated. These findings give insights on possible mVOCs levels in moisture-damaged buildings for an early detection of microbial activity and new evidences about the effect of indoor light on their emission.

**Keywords:** *Aspergillus niger*, molds, microbial Volatile Organic Compounds mVOC, indoor air quality, light effect, cultivation time, relative humidity.

---

\* **Corresponding author** : Christian George : [christian.george@ircelyon.univ-lyon1.fr](mailto:christian.george@ircelyon.univ-lyon1.fr)

## 1        **1. Introduction**

2        Humidity and mold growth are common phenomena indoors, where 25 to 38% of  
3 residences are contaminated with visible molds, based on studies carried out in Europe and  
4 North America (Moularat et al., 2008a). Such exposure to various types of molds, bacteria,  
5 microbial and chemical compounds may increase the associated health risk, causing  
6 respiratory problems like asthma and allergies (Adams et al., 2017; Gerritsen et al., 2018;  
7 Hurraß et al., 2017; Matysik et al., 2008). Nevertheless, growing molds on building materials  
8 can be a source of indoor emission of microbial Volatile Organic Compounds (mVOCs)  
9 released either by the degradation of materials through the generation of enzymes and acids or  
10 via primary and secondary metabolisms that include processes such as metabolic oxidation of  
11 glucose from several precursors (acetate, amino acids, fatty acids, keto acids), DNA synthesis,  
12 amino and fatty acids production (Korpi et al., 2009; Moularat et al., 2008b). Among these  
13 mVOCs, different ketones, aldehydes, acids, alcohols, sulfur compounds, amines, terpenes  
14 and terpene derivatives can be found, where some of them are classified as tracers of fungal  
15 activity (Moularat et al., 2008b, 2008a). These compounds are released in the gas phase and  
16 they can be used as markers for the early detection of fungi contamination in indoor  
17 environment (Moularat et al., 2008b). Especially, in residences, ventilation is one of the most  
18 effective way to reduce or eliminate the concentrations of the emitted pollutants, and to  
19 control the thermal environment and humidity in buildings (Soldatova et al., 2014). It was  
20 also found that a common low ventilation rate (0.1 to 0.3 h<sup>-1</sup>) induces elevated levels of these  
21 compounds affecting the human health (Dimitroulopoulou, 2012; Willem et al., 2013).  
22 Moreover, among the most common molds identified in the indoor environment there is the  
23 *Aspergillus sp.* (Matysik et al., 2008). Particularly, *Aspergillus sp.* (mainly *Aspergillus niger*)  
24 are highly detected in household's unfiltered air, ventilation systems, contaminated dust,  
25 water, and can even contribute to food (fruits, vegetables, cereals, meat...) degeneration and  
26 contamination causing some serious respiratory problems related to emitted allergens (Costa  
27 et al., 2016). The growth of such microbial species depends on cultivation time as well as  
28 other conditions like the substrate's type and pH, humidity, light and temperature (Korpi et  
29 al., 2009).

30        Nevertheless, previous studies focused their investigations on mVOCs emission from  
31 various cultures of fungi grown on different types of building materials (gypsum board,  
32 particleboard, fiberglass, wall paper, ceiling tiles...) as well as culture media (Adams et al.,  
33 2017; Claeson et al., 2002; Gerritsen et al., 2018; Kozicki et al., 2019; Matysik et al., 2009,

1 2008; Morath et al., 2012; Moularat et al., 2008b; Polizzi et al., 2012; Soon et al., 2012).  
2 Whereas, other studies reviewed the photocatalytic destruction of microorganisms mainly by  
3 titanium dioxide TiO<sub>2</sub> under relevant indoor conditions (irradiation, materials) (Benabbou et  
4 al., 2007; Chen et al., 2009; Markowska-Szczupak et al., 2010; Pigeot-Rémy et al., 2011;  
5 Sánchez et al., 2012; Wu, 2009). It was found that the mVOCs' emission profiles depend  
6 mainly on the type of substrate and the molds type along their growth conditions (i.e. under  
7 which humidity, light and temperature the fungi was grown) (Adams et al., 2017; Claeson et  
8 al., 2002; Gerritsen et al., 2018; Matysik et al., 2009, 2008; Morath et al., 2012; Moularat et  
9 al., 2008b; Polizzi et al., 2012; Soon et al., 2012). Additionally, typical mVOCs emitted from  
10 molds in their initial stage of growth (e.g. 2-pentanol) can be an evidence for their spreading  
11 and growing in the indoor environment. Hence, measuring these mVOCs enable the  
12 advantage of detecting recent fungal development before any visible/evident signs of  
13 contamination (Matysik et al., 2008; Moularat et al., 2008b). However, up to our knowledge,  
14 the impact of indoor relative humidity and light irradiation on mVOC emission profiles of  
15 fully-grown *Aspergillus niger* have not yet been experimentally observed nor quantified.

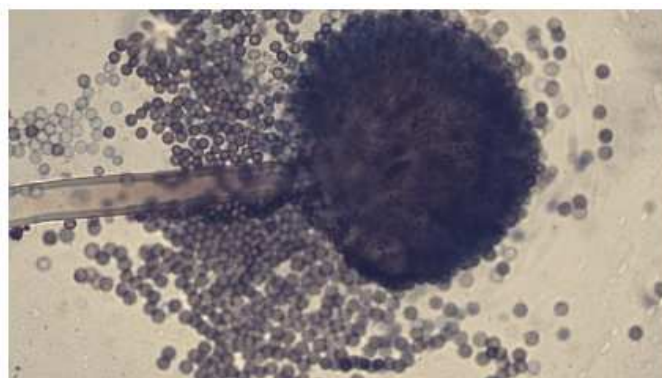
16 Thus, for a better understanding of the influence of indoor conditions (under indoor  
17 relative humidity regardless of the moistness of the substrate and typical light irradiation) on  
18 mVOC emission profiles, this work reports the production of mVOCs from *Aspergillus niger*  
19 grown on malt agar. The measurements were achieved in a dynamic mode and online  
20 monitored by means of a high-resolution proton-transfer-reaction-time of flight- mass  
21 spectrometer (PTR-TOF-MS). This study will also help discerning the mVOCs emissions  
22 levels from molds' type *Aspergillus niger* in a standard living room under different air  
23 exchange rates. This will be useful in the early detection of the microbial activity in sick  
24 buildings.

## 25 **2. Materials and method**

### 26 **2.1. Culture medium preparation**

27 Malt agar extract was selected as the culture medium since it is highly recommended for  
28 the cultivation of mold species present in indoor environments (Sunesson et al., 1995).  
29 Culture medium preparation consisted of dissolving 45g of malt agar extract in 1L of water.  
30 The solution was stirred and heated at 90-100°C until complete dissolution. The content was  
31 then poured into a flask and sterilized at 121°C for 15 min followed by pH adjustment to pH  
32 4.5 through the addition of a sterile solution of lactic acid at 10%. Finally, the solution was

1 transferred and homogenized in petri dishes (Diameter = 40 mm), which were incubated in a  
2 controlled incubation chamber under aerobic conditions, in the dark, at a temperature 25°C-  
3 30°C and at a relative humidity RH >75%. This incubator is stored in the basement of the  
4 building and allows a circulation of air from out-to inside the chamber. Hence, after 3 days of  
5 incubation, molds start growing in the petri dishes by consuming the culture media. Once the  
6 molds have completely grown (surface of petri dish completely covered), an identification by  
7 molecular biology was carried out following the protocol developed by CONIDIA's  
8 laboratory (work on ITS sequence) (Gardes and Bruns, 1996). Identification tracks are  
9 obtained through the comparison of the obtained DNA sequences with that of international  
10 databases. Additional information was obtained by performing a subculture of each strain on a  
11 specific medium. After 7 days of incubation at 25 °C, macroscopic and microscopic  
12 observations were made to refine the obtained results by molecular biology. In order to isolate  
13 and identify the molds present on the petri dish, re-culture on specific medium for the growth  
14 of yeasts and molds was performed. After 5 days of incubation, only one strain developed,  
15 and via molecular biology identification, it was found that the isolated mold in the sample  
16 belonged to the species *Aspergillus sp. type Aspergillus niger* (**Figure 1**).



17  
18 **Figure 1:** Microscopic observation (zoom x630) of *Aspergillus niger*

19

## 20 **2.2. mVOCs pattern analysis**

### 21 *Experimental set up*

22 All the experiments were performed in a borosilicate glass reactor (volume = 80 cm<sup>3</sup>)  
23 flushed by a constant flow at 110 mL/min of dry/humid synthetic air and irradiated from the  
24 top side by a 340 nm LED lamp (ThorLabs, 60mW, nominal maximum irradiance of 2.22  
25 W/m<sup>2</sup>). Humid air was obtained by mixing a dry synthetic air with a second air stream passing  
26 through a bubbler at room temperature. A humidity and temperature probe (Vaisala

1 HUMICAP® HMP110) monitored the temperature ( $T= 25 \pm 2^\circ\text{C}$ ) and the relative humidity in  
2 the reactor ( $\text{RH}=0\text{-}90\% \pm 2\%$ ). The experimental setup is presented in **supporting materials**  
3 **SM-1**. A measurement cycle consisted in measuring the mVOC concentrations at the outlet of  
4 the reactor containing the molds petri dish for 180 s, and then the LED lamp was switched on  
5 for 180s. Three irradiation cycles were made for each petri dish to test the reproducibility of  
6 the light effect. The geometric surface of molds exposed to light was around  $12.56 \text{ cm}^2$ . A  
7 blank control comprising sterile petri dishes was performed under the same conditions.  
8 Three different relative humidities of synthetic air ( $\text{RH}=0, 50, 90 \%$ ) and five different  
9 incubation times (5, 10, 14, 17 and 40 days) were tested. Under each of the experimental  
10 condition, three to four replicas of blanks and experiments were performed. The VOC  
11 concentrations detected from the blank were subtracted to the VOC concentration detected  
12 from the samples. The obtained concentrations correspond to the average of the repetitions  
13 and their corresponding uncertainties were calculated using error propagation taking into  
14 account both the standard deviation of the replicas and the uncertainty on the concentration  
15 given by the PTR-TOF-MS measurements.

#### 16 ***PTR-TOF-MS measurement***

17 A high-resolution proton transfer reaction-time of flight- mass spectrometer (PTR-  
18 TOF-MS 8000, Ionicon Analytik) was used to detect the mVOCs emitted from petri dishes  
19 containing the molds. Ionization was carried out in  $\text{H}_3\text{O}^+$  mode. PTR-MS allows the  
20 estimation of the mVOCs concentrations without an external calibration (Cappellin et al.,  
21 2012).

22 The instrument's settings used during all the experiments were chosen as follow: drift voltage  
23 of 500 V, drift temperature of 353 K and drift pressure of 2.2 mbar, resulting in an E/N ratio  
24 of about 123 Td ( $1 \text{ Td}=10^{-17} \text{ V cm}^2$ ). The reactor's outlet and the PTR-TOF-MS sample inlet  
25 tubing were constantly heated to 333 K to minimize adsorption and loss of products. The  
26 PTR-TOF-MS continuously samples 50 mL/min from the emission stream through 1.5 m of 6  
27 mm (i.d.) heated Silicon Steel tubing. The used instrument provides a mass resolution of 4500  
28  $\text{m}/\Delta\text{m}$ . Spectra were acquired at a time resolution of 1 s. A calibration gas standard (TO-14A  
29 Aromatic Mix, Restek Corporation, Bellefonte, USA)  $100 \pm 10 \text{ ppb}$  in Nitrogen was used to  
30 regularly assess the instrument performance, including mass resolution, mass accuracy,  
31 sensitivity and relative mass-dependent transmission efficiency. The sensitivity of these  
32 compounds ranged between 15 and 70 cps/ppb depending on the mass weight. However, since  
33 it was not possible to calculate the exact sensitivity for all the detected compound, we

1 assumed that the proton reaction constant was always equal to  $2 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ , and thus the  
 2 average sensitivity of 30 cps/ppb was applied for all the compounds. The ToFViewer v1.4.3  
 3 software was used for raw spectral data analysis, mass calibration, peak fitting, and area  
 4 extraction. Mass calibration was performed using internal calibration based on the two ions  
 5 having known exact mass (i) hydronium ion isotope ( $\text{H}_3^{18}\text{O}^+$   $m/z$  21.022) and (ii) protonated  
 6 acetone ( $\text{C}_3\text{H}_7\text{O}^+$   $m/z$  59.049) (Cappellin et al., 2011; Lindinger et al., 1998). The mVOC  
 7 concentrations are expressed in ppb and were calculated by the formula already described by  
 8 Cappellin et al. (2012). Since a variation in the relative humidity can affect the  $\text{H}_3\text{O}^+$  and  
 9  $\text{H}_3\text{O}^+(\text{H}_2\text{O})$  ions signals and as a consequence the  $\text{H}_3\text{O}^+$  to  $\text{H}_3\text{O}^+(\text{H}_2\text{O})$  ratio, the mVOC ion  
 10 signals were normalized to a standard reagent ion signal of  $10^6 \text{ counts s}^{-1}$  (cps) according to  
 11 the procedure given by de Gouw et al. (2003), where the  $X_R$  factor (factor reflecting both the  
 12 difference in mass-dependent detection efficiencies for  $\text{H}_3\text{O}^+$  and  $\text{H}_3\text{O}^+(\text{H}_2\text{O})$  ions and the  
 13 difference in the rate coefficients for the  $\text{H}_3\text{O}^+ + \text{R}$  and  $\text{H}_3\text{O}^+(\text{H}_2\text{O}) + \text{R}$  proton transfer  
 14 reactions) values were taken 0.1 for aromatics and 0.5 for the other compounds. Uncertainties  
 15 on the absolute concentrations can be estimated to be about 30% arising from systematic  
 16 errors.

17 The mVOC emission fluxes ( $E_{\text{mVOC}}$ ) in  $\mu\text{g m}^{-2} \text{ h}^{-1}$  were calculated as (Abis et al.,  
 18 2018; Cappellin et al., 2017; Potard, 2017):

$$E_{\text{mVOC}} = \frac{F_{\text{air}} \times ([\text{mVOC}]_{\text{molds}} - [\text{mVOC}]_{\text{blank}}) \times M_{\text{mVOC}}}{V_{\text{mol}}^{\text{air}} \times S_{\text{exposed}}} \quad (\text{Eq. 1})$$

20  
 21 where  $F_{\text{air}}$  is the air flow ( $F_{\text{air}} = 6.6 \text{ mL/h}$ ),  $[\text{mVOC}]_{\text{molds}}$  and  $[\text{mVOC}]_{\text{blank}}$  are the  
 22 concentrations of mVOC emitted from molds petri and that of sterile petri (ppb),  $M_{\text{mVOC}}$  is the  
 23 molecular mass of the corresponding mVOC (g/mol),  $V_{\text{mol}}^{\text{air}}$  is the air molar volume at standard  
 24 temperature and pressure (24.79 L/mol at  $25^\circ\text{C}$  and 1 atm), and  $S_{\text{exposed}}$  is the exposed surface  
 25 of molds to light ( $S_{\text{exposed}} = 12.56 \times 10^{-4} \text{ m}^2$ ).

26 The identification of the molecular formula of the detected VOCs was performed  
 27 using the Spectra Analyzer within the PTR viewer 3.2.8 software (Ionicon, Analytik GmbH).  
 28 For each series of experiments, a peak list was generated using the 'Auto search' tool within  
 29 the PTR Viewer alongside a further exploration for other products and fragments that might  
 30 be missing from the initial list. Using the established lists of peak, each peak's shape was

1 manually checked to define the fitting amplitude of each ion. The software offers three types  
2 of adjustment: normal, Gaussian and multipeak. The Gaussian and multipeak modes are  
3 mainly used when there are overlapping peaks of very close masses (**Figure SM-2 in the**  
4 **supporting materials**), allowing the determination of the exact limits of each peak, and hence  
5 minimizing the influence of one mass on the other during the integration of the peak.

6 Moreover, this software allows the identification of the compounds corresponding to each  
7 mass' peak by searching the possible combinations of elements leading to the closest  
8 molecular weight (within a 0.1 range). The chemical formulas and the compounds attribution  
9 correspond to the best matches for the selected m/z. The compounds assignments were done  
10 using isotopes signatures and literature reviews. It is noteworthy to mention that this  
11 technique does not provide a certain identification of the compounds as several compounds  
12 could have the same molecular formula but not the same chemical structure. Moreover, even  
13 with this procedure, some compounds could not be identified.

#### 14 *mVOCs statistical analysis*

15 The dataset before statistical analysis was made of 180 variables corresponding to the  
16 number of peaks detected in PTR-TOF-MS measurements. In order to differentiate the mVOC  
17 emission profiles between the different conditions, Principal Component Analysis (PCA,  
18 package FactormineR, Ade4 Version 1.0.153–©2009-2017 R Studio) was performed on the  
19 datasets using R software (Version 1.0.153 – ©2009–2017 RStudio). The PCA enables a  
20 statistical representation of all data without any preconception as to the type of emitted  
21 compounds and without bias. PCA generated optimum and similar graphical representations  
22 of the scatterplot representing the data matrix (Moularat et al., 2008b). Only the 60 most  
23 abundant compounds (having the highest emission rates) were presented in **Table 1**.

### 24 **3. Results and discussion**

#### 25 **3.1. Identification of the most abundant mVOC**

26 This study focused on the determination of the mVOCs emitted from surfaces covered with  
27 *Aspergillus niger* under different relative humidity and incubation time. The 60 most  
28 abundant mVOCs detected under dry air, 50% and 90% humid air for 5 days of incubation  
29 time along their corresponding emission fluxes are summarized in **Table 1** and their  
30 concentrations in **supporting materials SM-3**.



1 The mVOC production is associated to the metabolic and oxidative reactions of  
2 microorganisms. Among the most abundant mVOC, aldehydes, ketones, acids, nitrogen and  
3 sulfur containing compounds were identified.

4 Alcohols and ketones are produced through the oxidation of fatty acids and the  
5 decarboxylation of fatty acid derivatives respectively (Boots et al., 2014). In fact, C<sub>2</sub>-C<sub>4</sub>  
6 alcohols and ketones (ethanol and acetone) are produced through fundamental biochemical  
7 processes like glycolysis and Krebs cycle for almost all organisms in quantities depending on  
8 growth conditions mainly media composition and CO<sub>2</sub>/O<sub>2</sub> levels (Claeson, 2006; Wilkins et  
9 al., 2000). Besides, C<sub>8</sub> alcohols and ketones can be formed by fatty acid degradation from  
10 linoleic acid and linolenic acid (Boots et al., 2014). Longer 2-ketones (2-pentanone, 2-  
11 heptanone, 2- nonanone and 2-dodecanone) are considered as biomarkers for fungal activity.  
12 Likewise, acetoin or 3-hydroxybutanone is a derivative of the pyruvate fermentation under  
13 anaerobic conditions and is used to distinguish between lactose-fermenting and  
14 nonfermenting Enterobacteriaceae (Abis et al., 2020; Bos et al., 2013).

15 Moreover, microorganisms can produce aldehydes arising from oxidative metabolism and  
16 lipid peroxidation (Boots et al., 2014). For example, acetaldehyde is formed through oxidative  
17 carboxylation of acetolactate (byproduct of the synthesis of leucine in yeasts) (Claeson, 2006)  
18 and phenylalanine metabolism (Costa et al., 2016). 2-phenylethanol and phenylacetaldehyde  
19 are also associated to phenylalanine metabolism (Costa et al., 2016). Whereas, hexanal,  
20 heptanal and nonanal may result from the transformation of unsaturated fatty acids (Bos et al.,  
21 2013; Claeson, 2006).

22 On the other hand, low molecular weight carboxylic acids like formic acid, acetic acid,  
23 propionic acid, and butyric acid result from anaerobic metabolism linked to alcoholic  
24 fermentation of the substrate (malt agar extract in this study) (Boots et al., 2014; Bos et al.,  
25 2013). Acetic acid for example is the most abundant fatty acid emitted by yeasts (Claeson,  
26 2006), while acids like 2-methylpropanoic acid, butyric acid, 2 or 3-methylbutanoic acids,  
27 pentanoic acid and hexanoic acid can result from the lipolysis of triglycerids or amino acids  
28 metabolism (Boots et al., 2014; Costa et al., 2016).

29 Sulfur-containing compounds may result from the degradation of amino acids containing  
30 sulfur such as methionine and cysteine. As for nitrogen-containing compounds, they are of  
31 great interest due to their reactivity and potential airway irritants (Claeson et al., 2007, 2002).  
32 Their formation is due to amino-acid breakdown (Boots et al., 2014). Few studies have  
33 identified amines like pyridine, 2-methylpyridine, triethylamine as microbial metabolites

1 (Claeson et al., 2002), in addition to nitromethane that is also emitted from microbial growth  
2 and can therefore be used as a marker compound (Claeson, 2006).

3 Aromatics and oxygenated aromatics (like phenol, toluene) probably derived from aromatic  
4 amino acids or from monoterpenes via decarboxylation and oxygenation (Wilkins and Larsen,  
5 1995). Besides, hydrocarbons like isoprene may result from the methylerythritol phosphate  
6 pathway or  $\beta$ -oxidation of fatty acids (Boots et al., 2014). As for alkenes like 1-pentene, 1-  
7 heptene, they are produced through the  $\beta$ -oxidation of fatty acids, a common pathway for  
8 most volatile hydrocarbons (Bos et al., 2013). Alkenes were also observed in the study of  
9 Wilkins et al. (2000).

10 Furthermore, the common tracers' compounds of the microbial activity of *Aspergillus* were  
11 detected. Among the identified tracers under dry air, we cite: nonanal/nonanone ( $12.3\mu\text{g}/\text{m}^3$ ),  
12 1-octen-3-ol/3-octanone ( $1.3\ \mu\text{g}/\text{m}^3$ ), isoprene ( $2.1\ \mu\text{g}/\text{m}^3$ ), methylfuran ( $1.5\ \mu\text{g}/\text{m}^3$ ), 1,3-  
13 octadiene ( $1.9\ \mu\text{g}/\text{m}^3$ ), 2-pentanol or 3-hydroxybutanone ( $1.6\ \mu\text{g}/\text{m}^3$ ), 2-pentanone ( $0.5$   
14  $\mu\text{g}/\text{m}^3$ ), heptanone ( $0.3\ \mu\text{g}/\text{m}^3$ ), butanol/2-methyl-1-propanol ( $0.8\ \mu\text{g}/\text{m}^3$ ), hexanal ( $0.6$   
15  $\mu\text{g}/\text{m}^3$ ), hexenal ( $0.2\ \mu\text{g}/\text{m}^3$ ), dimethyl sulfide ( $0.1\ \mu\text{g}/\text{m}^3$ ), dimethyl disulfide ( $0.3\ \mu\text{g}/\text{m}^3$ ),  
16 formamide ( $1\ \mu\text{g}/\text{m}^3$ ), nitromethane ( $0.3\ \mu\text{g}/\text{m}^3$ ), pyridine ( $0.1\ \mu\text{g}/\text{m}^3$ ), dimethylpyridine or  
17 benzylamine ( $0.1\ \mu\text{g}/\text{m}^3$ ).

18 Most of the mVOC identified in this study are in good agreement with those reported in  
19 the literature for *Aspergillus niger* species cultivated on different culture media and  
20 constructions materials (Chen, 2002; Costa et al., 2016; Fiedler et al., 2001; Gerritsen et al.,  
21 2018; Lavine et al., 2012; Matysik et al., 2009, 2008; Meruva et al., 2004; Moularat et al.,  
22 2008a, 2008b; Shehu and Bello, 2011; Siddiquee et al., 2015). However, each growth material  
23 has a specific fingerprint, and the most common compounds emitted from *Aspergillus niger*  
24 for different growth materials (malt agar extract, DG-agar, wall paper, fiberglass, glass fiber)  
25 were: methylfuran, 1-octen-3-ol, 2-methyl-1-butanol, 2-methyl-1-propanol and these  
26 compounds may be considered as tracers of the microbial activity of *Aspergillus niger* and  
27 indicator of mold growth in indoor environment.

28

29

**Table 1 :** Most abundant mVOC detected for different relative humidities for molds incubated for 5 days.

List of most emitted mVOCs			PC A <sup>†</sup>	Dry air			50% humid air			90% humid air		
m/z	Most likely formula	Tentative identification		m/z	Average contribution (%)	mVOC Fluxes (µg/m <sup>2</sup> /h) <sup>‡</sup>	m/z	Average contribution (%)	mVOC Fluxes (µg/m <sup>2</sup> /h) <sup>‡</sup>	m/z	Average contribution (%)	mVOC Fluxes (µg/m <sup>2</sup> /h) <sup>‡</sup>
33.032	CH <sub>4</sub> O	Methanol	XX	59.047	13.7	67.9 ± 23.8	59.047	21.5	39.4±13.8	95.025	33.1	52.2±18.3
41.037	C <sub>3</sub> H <sub>4</sub>	Propadiene	XX	143.14	12.2	60.5±21.2	33.032	11.6	21.2±7.4	33.032	21.8	34.3±12.0
42.033	C <sub>2</sub> H <sub>3</sub> N	Acetonitrile		61.027	9.8	48.5±16.9	95.025	8.3	15.2±5.3	43.053	5.7	8.9±3.1
43.017	C <sub>2</sub> H <sub>2</sub> O	Hexyl acetate fragment/ketene	X/X X	33.032	4.8	23.9±8.4	143.14	6.9	12.5±4.4	42.033	4.8	7.5±2.6
43.053	C <sub>3</sub> H <sub>6</sub>	Propene		43.017	4.3	21.3±7.5	43.053	4.7	8.6±3.0	69.03	3.9	6.2±2.2
45.032	C <sub>2</sub> H <sub>4</sub> O	Acetaldehyde		45.032	3.5	17.5±6.1	61.027	3.5	6.3±2.2	113.05	2.6	4.2±1.5
46.03	CH <sub>3</sub> NO	Formamide	X/X X	47.021	2.7	13.3±4.7	45.032	2.5	4.6±1.6	93.037	2.6	4.1±1.4
47.012	CH <sub>2</sub> O <sub>2</sub>	Formic acid	X/X X	87.042	2.6	13.1±4.6	43.017	2.3	4.1±1.4	143.14	2.4	3.7±1.3
47.021	CH <sub>2</sub> O <sub>2</sub> /C <sub>2</sub> H <sub>6</sub> O	Formic acid/ethanol	X/X X	47.012	2.3	11.5±4.0	47.021	2.3	4.1±1.5	51.04	2.3	3.6±1.3
47.048	C <sub>2</sub> H <sub>6</sub> O	Ethanol	X	69.068	2.0	10.1±3.5	69.03	1.7	3.1±1.1	96.01	2.1	3.4±1.2
49.998			X	69.03	2.0	9.9±3.5	60.046	1.6	3.0±1.1	97.01	2.1	3.3±1.2
51.04	CH <sub>4</sub> O-H <sub>2</sub> O	Methanol-water cluster	X/X X	60.046	2.0	9.7±3.4	111.11	1.5	2.7±0.9	111.11	1.6	2.5±0.9
57.033	C <sub>3</sub> H <sub>4</sub> O	2-propenal (acrolein)	X/X X	111.11	1.9	9.2±3.2	69.068	1.5	2.7±0.9	69.068	1.5	2.3±0.8
57.07	C <sub>4</sub> H <sub>10</sub> O/C <sub>4</sub> H <sub>8</sub>	Butanol/butene	X	89.056	1.6	7.9±2.8	47.048	1.4	2.6±0.9	95.082	1.2	1.8±0.6
59.047	C <sub>3</sub> H <sub>6</sub> O	Acetone	XX	83.083	1.5	7.2±2.5	123.95	1.2	2.2±0.8	129.12	1.0	1.5±0.5
60.046	C <sub>2</sub> H <sub>5</sub> NO	Acetamide/ N-methyl formamide/ nitrosoethano	X/X X	129.12	1.3	6.6±2.3	87.042	1.1	2.1±0.7	41.037	0.7	1.2±0.4
61.027	C <sub>2</sub> H <sub>4</sub> O <sub>2</sub>	Acetic acid	X/X X	43.053	1.3	6.4±2.2	129.12	1.0	1.8±0.6	127.11	0.7	1.1±0.4

62.027	CH <sub>3</sub> NO <sub>2</sub>	Nitromethane	X/X X	123.95	1.2	6.0±2.1	83.083	1.0	1.8±0.6	73.064	0.6	1.0±0.3
63.024	C <sub>2</sub> H <sub>6</sub> S	Dimethylsulfide	X	42.033	1.2	5.7±2.0	93.037	0.9	1.7±0.6	57.033	0.5	0.7±0.2
69.03	C <sub>4</sub> H <sub>4</sub> O	Furan	X/X X	144.14	1.1	5.6±1.9	73.064	0.9	1.6±0.56	63.024	0.4	0.6±0.2
69.068	C <sub>5</sub> H <sub>8</sub>	Isoprene	X/X X	107.05	1.1	5.4±1.9	127.11	0.9	1.6±0.6	81.042	0.4	0.6±0.2
70.071	C <sub>4</sub> H <sub>7</sub> N	butanenitrile	XX	125.13	1.0	5.1±1.8	51.04	0.9	1.6±0.5	144.14	0.3	0.5±0.2
71.05	C <sub>4</sub> H <sub>6</sub> O	Methyl vinyl ketone (MVK)	X/X X	73.064	1.0	4.8±1.7	41.037	0.8	1.5±0.5	93.068	0.3	0.6±0.1
71.08	C <sub>5</sub> H <sub>10</sub>	Pentene	X	127.11	1.0	4.8±1.7	46.03	0.8	1.4±0.5	123.95	0.3	0.±0.1
73.027	C <sub>3</sub> H <sub>4</sub> O <sub>2</sub>	2-propenoic acid	X/X X	46.03	1.0	4.8±1.7	113.05	0.7	1.3±0.5	115.07	0.3	0.4±0.1
73.064	C <sub>4</sub> H <sub>8</sub> O	2-butanone, (MEK)/2- methylpropanal	X/X X	71.05	0.9	4.7±1.6	89.056	0.7	1.3±0.5	75.043	0.3	0.4±0.1
74.061	C <sub>3</sub> H <sub>7</sub> NO	N, N- dimethylformamide/Propa namide	X/X X	57.07	0.8	4.1±1.4	108.96	0.7	1.3±0.4	97.061	0.22	0.4±0.1
75.043	C <sub>3</sub> H <sub>6</sub> O <sub>2</sub>	Hydroxyacetone	X/X X	108.96	0.8	4.0±1.4	97.01	0.7	1.2±0.4	62.027	0.2	0.3±0.1
81.042	C <sub>6</sub> H <sub>8</sub> /C <sub>5</sub> H <sub>4</sub> O	Methycyclopentadiene /2,4-cyclopentadiene-1- one		49.998	0.8	3.8±1.3	96.01	0.7	1.2±0.4	89.056	0.2	0.3±0.1
83.083	C <sub>5</sub> H <sub>6</sub> O/C <sub>6</sub> H <sub>10</sub>	2-methylfuran/hexenol fragment or hexene	X/X X	113.05	0.7	3.6±1.3	57.033	0.6	1.2±0.4	101.06	0.2	0.3±0.1
85.063	C <sub>6</sub> H <sub>12</sub>	1-hexene	X/X X	75.043	0.7	3.2±1.1	144.14	0.6	1.2±0.4	121.06	0.2	0.3±0.1
87.042	C <sub>4</sub> H <sub>6</sub> O <sub>2</sub>	Butenoic acid	X/X X	73.027	0.6	3.2±1.1	141.13	0.6	1.2±0.4	108.08	0.2	0.2±0.1
87.076	C <sub>5</sub> H <sub>10</sub> O	2-methyl butanal/pentanone	XX	47.048	0.6	2.9±0.1	125.13	0.6	1.0±0.4	107.05	0.1	0.2±0.1
89.056	C <sub>5</sub> H <sub>12</sub> O/C <sub>4</sub> H <sub>8</sub> O <sub>2</sub>	2-pentanol /Butanoic acid/Acetoin/	X/X X	101.06	0.6	2.9±1.0	73.027	0.4	0.8±0.3	91.055	0.1	0.2±0.1
91.055	C <sub>4</sub> H <sub>10</sub> S	Butanethiol / diethyl sulphide	X/X X	57.033	0.6	2.7±0.9	101.06	0.4	0.8±0.3	45.032	0.1	0.2±0.1
93.037	C <sub>6</sub> H <sub>4</sub> O/C <sub>3</sub> H <sub>8</sub> O <sub>3</sub>		X	141.13	0.5	2.4±0.8	47.012	0.4	0.7±0.3	74.061	0.1	0.2±0.1
93.068	C <sub>7</sub> H <sub>8</sub>	Toluene		87.076	0.5	2.3±0.8	107.05	0.4	0.7±0.3	99.079	0.1	0.2±0.1

95.025	C <sub>2</sub> H <sub>6</sub> S <sub>2</sub> /C <sub>2</sub> H <sub>6</sub> O <sub>2</sub> S	Dimethyl disulfide/dimethyl sulfone	X	97.01	0.4	2.2±0.8	75.043	0.4	0.7±0.2	87.076	0.1	0.2±0.1
95.082	C <sub>6</sub> H <sub>6</sub> O/C <sub>5</sub> H <sub>6</sub> N <sub>2</sub>	Phenol/pyridinamine	X	91.055	0.4	2.0±0.7	49.998	0.4	0.6±0.2	70.071	0.1	0.2±0.1
96.01	C <sub>5</sub> H <sub>5</sub> NO/C <sub>6</sub> H <sub>9</sub> N	Pyridine-N-oxide/2,4-dimethylpyrrole	X	74.061	0.4	2.0±0.7	87.076	0.4	0.6±0.2	83.083	0.1	0.2±0.1
99.079	C <sub>6</sub> H <sub>10</sub> O	Hexenal/hexenone	X/X X	103.04	0.4	1.8±0.6	71.08	0.3	0.6±0.2	85.063	0.1	0.2±0.1
107.05	C <sub>7</sub> H <sub>6</sub> O	Benzaldehyde	X	71.08	0.4	1.8±0.6	62.027	0.3	0.5±0.2	108.96	0.1	0.2±0.1
108.08	C <sub>7</sub> H <sub>9</sub> N	Dimethyl pyridine/benzylamine		41.037	0.3	1.5±0.5	71.05	0.3	0.5±0.2	46.03	0.1	0.1±0.03
108.96			X/X X	95.025	0.3	1.5±0.5	125.96	0.3	0.5±0.2	49.998	0.1	0.1±0.03
111.11	C <sub>8</sub> H <sub>14</sub>	1,3-octadiene	X/X X	125.96	0.3	1.5±0.5	108.08	0.3	0.5±0.2	125.13	<0.1	0.1±0.02
115.07	C <sub>7</sub> H <sub>14</sub> O	Heptanone/heptanal	X/X X	51.04	0.3	1.4±0.5	63.024	0.2	0.4±0.1	141.13	<0.1	0.1±0.02
121.06	C <sub>8</sub> H <sub>8</sub> O	Acetophenone		85.063	0.3	1.4±0.5	85.063	0.2	0.4±0.1	73.027	<0.1	<0.1
123.95			X/X X	62.027	0.3	1.3±0.5	81.042	0.2	0.4±0.1	71.05	<0.1	<0.1
125.13	C <sub>9</sub> H <sub>16</sub>	Nonadiene	X/X X	115.07	0.3	1.2±0.4	95.082	0.2	0.4±0.1	103.04	<0.1	<0.1
125.96			X	99.079	0.2	0.9±0.3	99.079	0.2	0.3±0.1	47.048	<0.1	<0.1
127.11	C <sub>8</sub> H <sub>14</sub> O/ fragment C <sub>8</sub> H <sub>16</sub> O	Octenone/ fragment 1-octen-3-ol or octanone	X/X X	97.061	0.2	0.9±0.3	91.055	0.1	0.3±0.1			
129.12	C <sub>8</sub> H <sub>16</sub> O	1-octen-3-ol or octanone	X/X X	96.01	0.2	0.8±0.3	97.061	0.1	0.2±0.1			
143.14	C <sub>9</sub> H <sub>18</sub> O	Nonanal/nonanone	X	108.08	0.1	0.7±0.2	74.061	0.1	0.2±0.1			
144.14	C <sub>8</sub> H <sub>17</sub> NO	Octanamide	X	70.071	0.1	0.7±0.2	121.06	0.1	0.2±0.1			
101.06	C <sub>6</sub> H <sub>12</sub> O	Hexanal/hexenol/hexanone	X	93.037	0.1	0.7±0.2	70.071	0.1	0.2±0.1			
103.04	C <sub>5</sub> H <sub>10</sub> O <sub>2</sub>	Pentanoic acid/methylbutanoic acid	X	121.06	0.1	0.6±0.2	115.07	0.1	0.2±0.1			
113.05	C <sub>7</sub> H <sub>12</sub> O/C <sub>8</sub> H <sub>16</sub> /C <sub>6</sub> H <sub>8</sub> O <sub>2</sub>	Heptenal/heptenone/octene/dimethylfuranone	X	81.042	0.1	0.6±0.2	103.04	0.1	0.1±0.03			

141.13	C <sub>9</sub> H <sub>16</sub> O	Nonenal/nonenone	X/X X	95.082	0.1	0.5±0.2	93.068	<0.1	<0.1	
97.01	C <sub>5</sub> H <sub>4</sub> O <sub>2</sub>	Furfural	X	93.068	0.1	0.4±0.1				
97.061	C <sub>6</sub> H <sub>8</sub> O	Dimethylfuran/ethylfuran	X	63.024	0.1	0.4±0.1				

‡ Uncertainties calculated using the error propagation method;

† X: compounds observed in PCA (1-3);

XX: compounds observed in PCA 4.

1 Nevertheless, few studies have given a range of concentration and emission fluxes of  
 2 the mVOCs and these values are clearly affected by the type of growth materials and  
 3 cultivation medias which can explain the variability of the concentrations (Chen, 2002;  
 4 Fiedler et al., 2001; Matysik et al., 2009, 2008; Moularat et al., 2008a). A comparison of the  
 5 concentrations of some mVOC on different substrate are presented in **Table 2**.

6

7 **Table 2:** mVOC concentration emitted by *Aspergillus niger* cultivated on different substrate

Compounds	Concentration ( $\mu\text{g}/\text{m}^3$ )					
	Malt agar extract*	Malt agar extract <sup>a</sup>	Fiberglass <sup>b</sup>	Glass fabric <sup>b</sup>	DG-18 agar <sup>c</sup>	Gypsum board <sup>a</sup>
1-octen-3-ol	1.3		13.23	6.06	1	1-1.6
1,3-octadiene	1.9		2.09	24.9		
2 or 3-methylfuran	1.5		71.68	0.95	7.8-15.4	
2-methyl-1-propanol	0.8	0.9			25	
3-methyl-1-butanol		0.1			56.8	
2-methyl-1-butanol					3.9	
2-pentanone	0.5				1.3	
3-heptanol				7.34		
4-heptanone	0.3			33.6		
Dimethyl disulfide	0.3		1.46	3.71		0.1

8 \*: Present study, a: Chen, (2002), b: Moularat et al., (2008a), c: Matysik et al., (2009)

9

10 According to **Table 2**, the concentration of the emitted mVOCs are dependent on the  
 11 type of the substrate. For example, 2-methyl furan was detected with a concentration of 0.95  
 12  $\mu\text{g}/\text{m}^3$  from *Aspergillus niger* cultivated on Glass fabric compared to a value of 1.5  $\mu\text{g}/\text{m}^3$  in  
 13 this study for malt agar media. However, when cultivated on DG-18 agar and controlled  
 14 substrate of non-emitting fiberglass, the concentration becomes 7-70 times higher (Matysik et  
 15 al., 2009; Moularat et al., 2008a). Another example for 1,3-octadiene detected with a  
 16 concentration of 2.09  $\mu\text{g}/\text{m}^3$  when cultivated on controlled fiberglass compared to 24.9  $\mu\text{g}/\text{m}^3$

1 and to 1.9  $\mu\text{g}/\text{m}^3$  when cultivated on Glass fabric and Malt agar extract in this study,  
2 respectively. Moreover, the concentration of 2-methyl-1-propanal emitted from *Aspergillus*  
3 *niger* cultivated on malt agar extract is consistent with that obtained in this study. However, in  
4 this study more mVOCs were identified compared to the study of Chen, (2002).

5 These alterations in the emission rates values for the different growth materials make it  
6 more difficult to quantify the concentrations of the chemical tracers in the indoor environment  
7 (Moularat et al., 2008a).

8

### 9 **3.2. Effect of relative humidity and incubation time on mVOC emission profiles**

10 In order to evaluate the relative humidity and incubation time effect on the emission  
11 profiles, different PCAs were performed (**Figures 2-3**).

12 **Figure 2** shows three PCAs performed for three incubation time (5, 14 and 40 days). PCA (1-  
13 3) described 67%, 60% and 42.1% of the total dataset variability respectively (56% for the  
14 first dimension and 9% for the second one for PCA-1; 52.5% for the first dimension and 7.4%  
15 for the second one for PCA-2; 30.9% for the first dimension and 11.2% for the second for  
16 PCA-3). The PCA was specifically used to identify the mVOCs that differentiate between the  
17 different relative humidities.

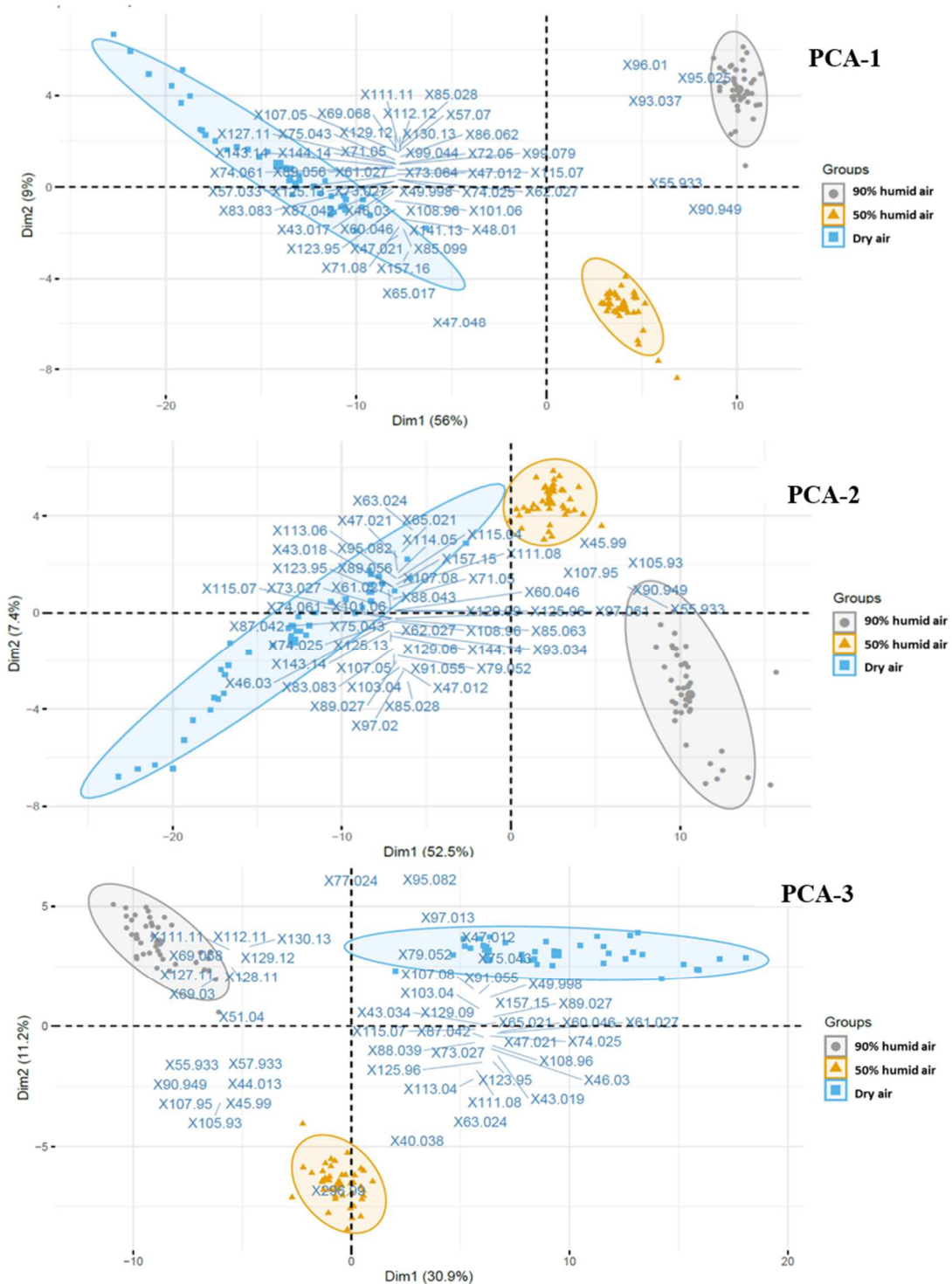
18 In addition, according to **Figure 2**, every condition has a differentiated mVOC emission  
19 profile where the ellipses grouping the different conditions are well separated. It is noticeable  
20 also that 32 to 45 of the 50 compounds explaining the variance between the different  
21 conditions are driven by the dry air condition. This means that the experiments performed  
22 under dry air condition has the most differentiated mVOC profile, which was expected since  
23 water is one of the factors that mostly affect VOC emissions.

24 Moreover, **Table 1** shows that the average contribution of each mVOC to the total  
25 emission is highly affected by the relative humidity where the contribution of some  
26 compounds remains constant ( $\text{C}_8\text{H}_{14}$ ,  $\text{C}_8\text{H}_{16}\text{O}$ ...), while others contributions increase ( $\text{C}_4\text{H}_4\text{O}$ ,  
27  $\text{C}_3\text{H}_8\text{O}$ ,  $\text{C}_7\text{H}_{12}\text{O}$ ,  $\text{C}_7\text{H}_{14}\text{O}$ ,  $\text{C}_6\text{H}_6\text{O}$ ,  $\text{C}_2\text{H}_6\text{S}$ ,  $\text{C}_2\text{H}_6\text{S}_2$ ...) or decrease ( $\text{C}_9\text{H}_{18}\text{O}$ ,  $\text{C}_5\text{H}_{12}\text{O}$ ,  
28  $\text{C}_4\text{H}_6\text{O}$ ...) with relative humidity. Hence, some VOCs can be boosted by the relative humidity  
29 due to their polarity (Wolkoff, 1998). Furthermore, mold activity is influenced by the  
30 substrate moistness and it has been demonstrated that when the humidity is higher than 75%,  
31 mold activity can increase up to 40% or more (Block, 1953). Therefore, the different VOCs  
32 profiles among dry and humid air conditions results from both the VOC chemical properties



1 and from a more active emission of microbiological fragments by molds due to the moistness  
 2 present from the humid air (Korpi et al., 2009).

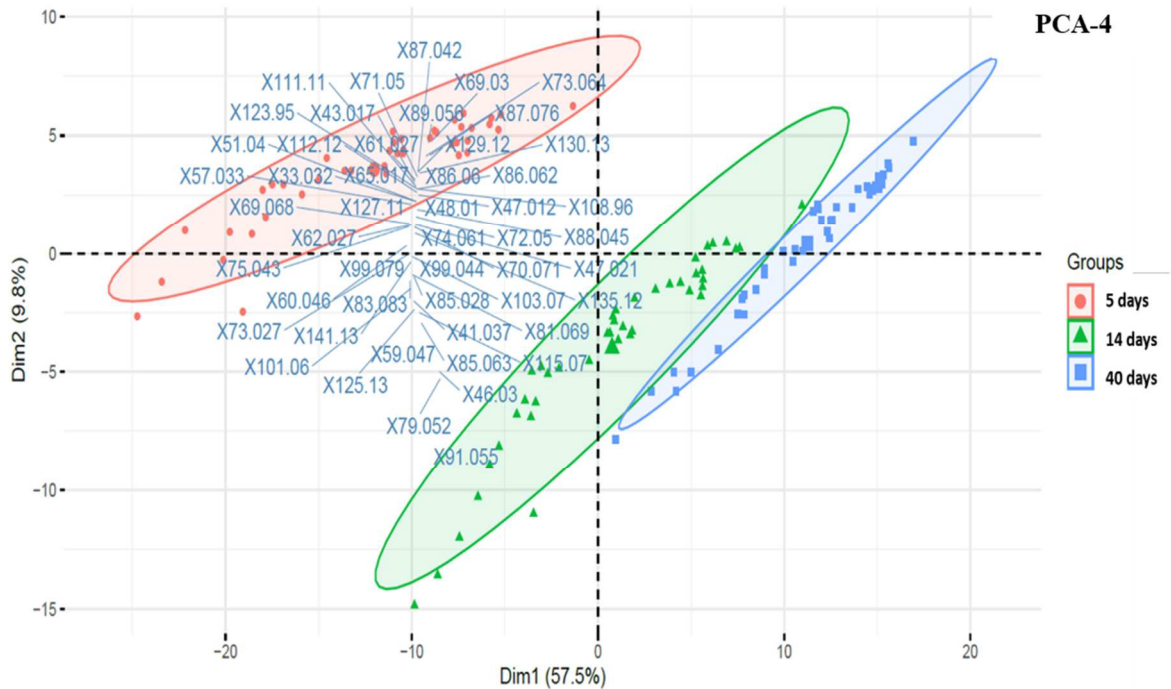
3



4

5 **Figure 2:** Relative humidity effect on mVOCs emission profiles for different incubation time  
 6 (PCA-1: 5 days, PCA-2: 14 days, PCA-3: 40 days). The m/z of the 50 compounds that are  
 7 most explaining the variance in the two first components are presented on the graph. The  
 8 percentages of the eigenvalues of the two first components are shown on each axis.

1 Another PCA (PCA-4, **Figure 3**) was performed to compare the profile of the mVOC  
 2 emissions between different incubation times under dry air condition. PCA-4 described 67.3%  
 3 of the total dataset variability (57.5% for the first dimension and 9.8% for the second). The  
 4 PCAs obtained under 50% and 90% relative humidity for different incubation time are  
 5 presented in **supporting materials SM-4**.  
 6



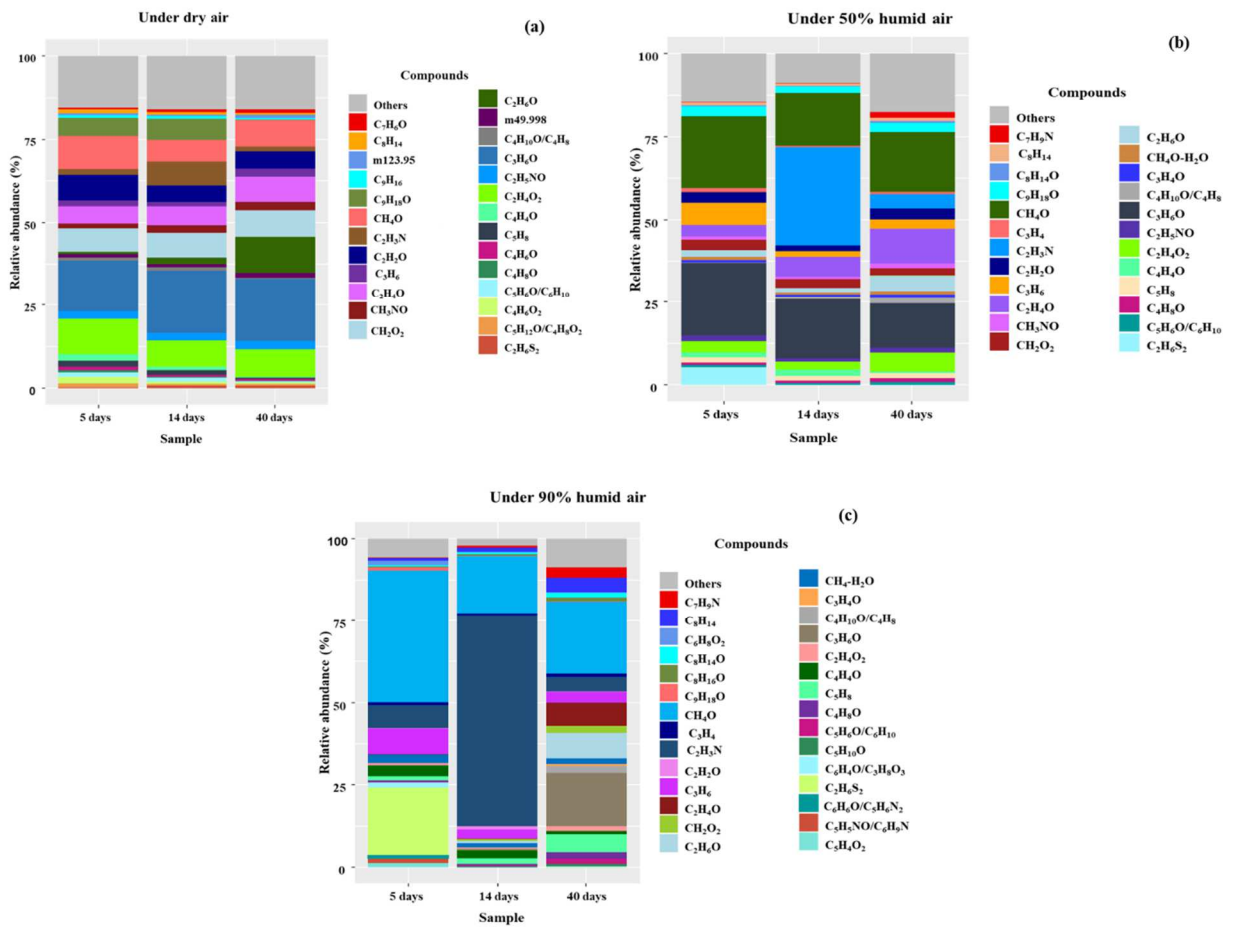
7  
 8 **Figure 3:** Incubation time effect on mVOCs emission profiles for dry air condition. The m/z  
 9 of the 50 compounds that are most explaining the variance in the two first components are  
 10 presented on the graph. The percentage of the eigenvalues of the two first components is  
 11 shown on each axe.

12  
 13 According to **Figure 3**, the ellipses grouping the different conditions are clearly  
 14 separated with a small overlapping between the 14 days and 40 days of incubation. It is also  
 15 noticeable that 32 to 45 of the 50 compounds explaining the variance between the different  
 16 conditions are driven by the 5 days incubation time, indicating that in the early stage of  
 17 growth, *Aspergillus niger* has the most different mVOC profile, reflecting the time-dependent  
 18 emission of certain mVOCs (Matysik et al., 2008). This dependency might be correlated to  
 19 the mold growth curve. It has been shown that during the first 5 days, molds' growth is  
 20 exponential (Pitt, 1993). Their metabolism is focused on the colonization of the substrate.  
 21 After 5 days, normally, they reach a plateau meaning that they might change their

1 metabolisms since it is not focused anymore on the fast colonization of the substrate  
 2 (Viitanen, 2007). This change of metabolism might be the reason behind the different VOC  
 3 profiles from 5 days and 14-40 days samples.

4 Likewise, the relative abundance of each mVOC was calculated relatively to the total  
 5 concentration detected for each condition (relative humidity and incubation time) and exposed  
 6 in **Figure 4**. This figure confirms the variability of the mVOC emission profiles observed in  
 7 PCAs analysis for each of the studied conditions of relative humidity and incubation time.

8

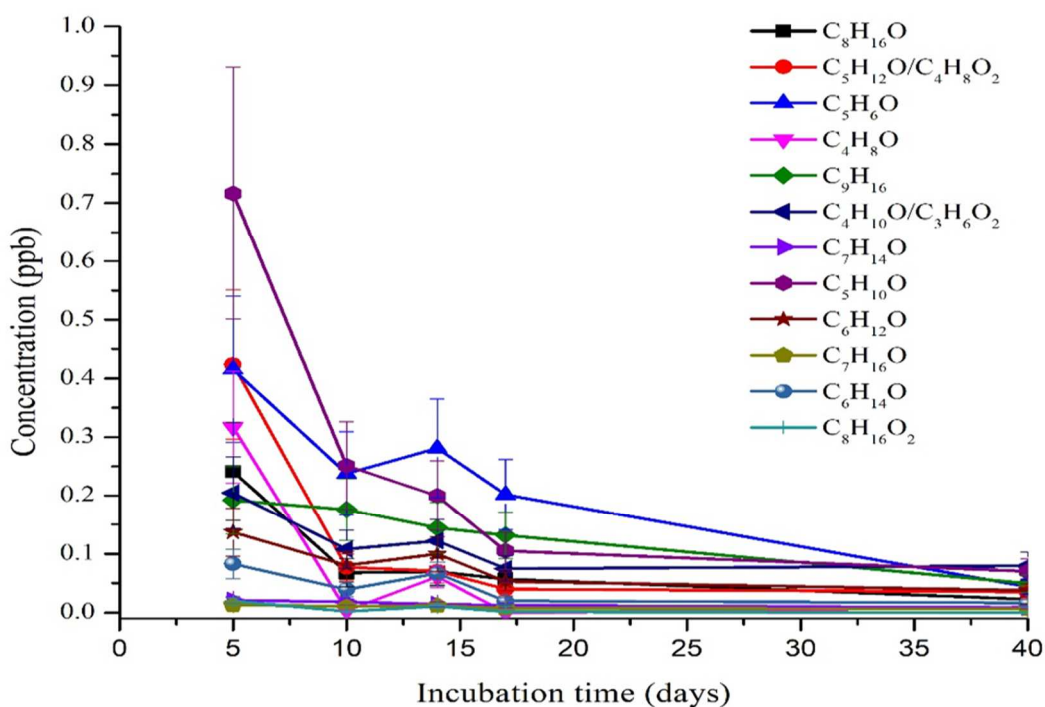


9

10 **Figure 4:** Relative abundance of the most emitted mVOCs under different relative humidity  
 11 and incubation time

12

13 Furthermore, the concentrations of 12 characteristic mVOCs emitted from *Aspergillus*  
 14 *niger* on of malt agar extract, which undergo the most dramatic change with the incubation  
 15 time are presented in **Figure 5**.



1  
 2 **Figure 5:** Concentration (ppb) of several mVOCs emitted by *Aspergillus niger* for different  
 3 incubation time under dry air.

4  
 5 According to **Figure 5**, the production of C<sub>4</sub>-C<sub>8</sub> alcohols and ketone/aldehydes was the  
 6 highest in the initial stage of growth. After 10-14 days of incubation time, the concentration of  
 7 these compounds has decreased to more than half. However, dimethyl disulfide C<sub>2</sub>H<sub>6</sub>S<sub>2</sub> seems  
 8 to increase after 14 days to reach a steady state. These results are compared to those available  
 9 in the literature for *Aspergillus niger* grown on different substrates (Matysik et al., 2008;  
 10 Moularat et al., 2008a).

11 In the study conducted by Matysik et al. on *Penicillium expansum*, *Penicillium*  
 12 *chrysogenum*, *Aspergillus fumigatus*, *Aspergillus versicolor*, *Aspergillus niger*, and  
 13 *Cladosporium cladosporoides* cultivated on dichloran glycerol agar (DG 18) and on wall  
 14 papers, different behaviors were observed depending on the cultivation media and the type of  
 15 molds (Matysik et al., 2008). For *Aspergillus niger* cultivated on wallpapers, a decrease of the  
 16 concentrations of 2-pentanol, 2-pentanone, 2-heptanone was observed over the whole period  
 17 of growth, while the emission rate of 1-octen-3-ol showed an increasing and later more  
 18 constant trend. However, for *Aspergillus niger* cultivated on DG 18, most mVOC detected

1 like 1-octen-3-ol, 3-octanol, 3-methyl-furan, 2-methyl-1-propanol, 1,3-nonadiene, decrease  
2 over the period of growth, while 3-methyl-1-butanol and 3-octanone increase (Matysik et al.,  
3 2008). Nevertheless, in the study carried out by Moularat et al. (2008a) no decrease in the  
4 emissions of mVOC was observed over the incubation periods for *Aspergillus niger* cultivated  
5 on glass fabric.

### 6 **3.3. Indoor light effect on mVOC emissions**

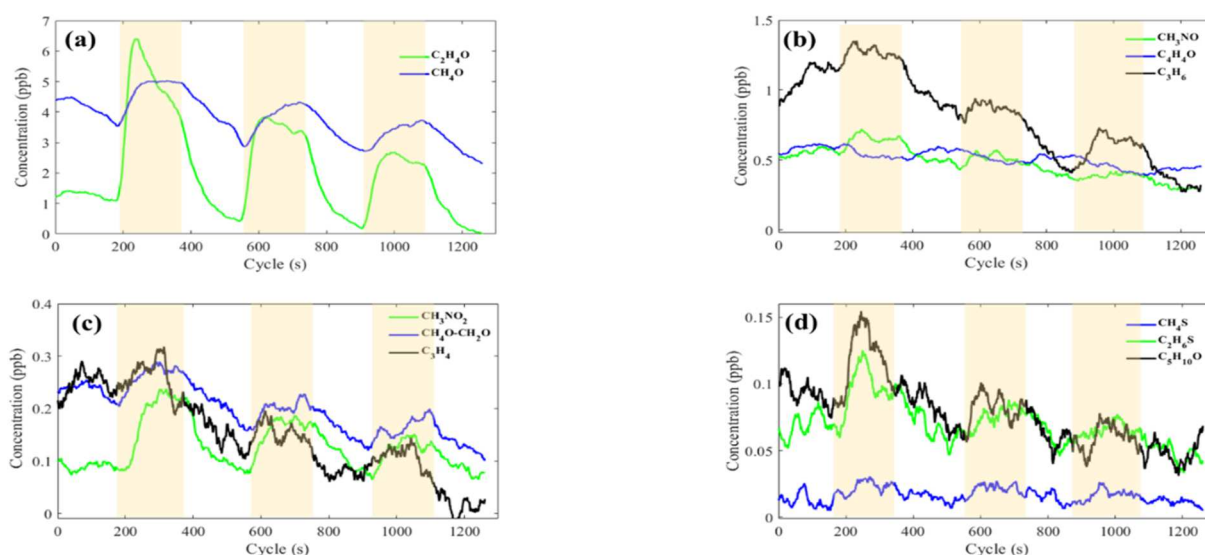
7 In order to evaluate the indoor light effect on the mVOC emission, surface covered with  
8 *Aspergillus niger* were irradiated with a 340 nm LED lamp. **Figure 6** shows an example of  
9 the temporal profile of mVOC with and without light under dry air after 5 days of incubation,  
10 where three irradiation cycles were performed.

11 According to **Figure 6**, it is seen that the concentration of some of the observed mVOC like  
12  $C_2H_4O$ ,  $CH_4O$ ,  $CH_4O-H_2O$ ,  $C_3H_4$ ,  $CH_3NO$ ,  $C_3H_6$ ,  $CH_3NO_2$ ,  $CH_4S$ ,  $C_2H_6S$ ,  $C_3H_{10}O$  is sensitive  
13 to the light, where their corresponding concentrations increase when the light is switched on,  
14 whereas the other mVOC detected remain constant. This phenomenon might be linked to the  
15 presence of aspergillin (Ray and Eakin, 1975), the black fungal spore pigment of *Aspergillus*  
16 *niger*. In fact, aspergillin is classified as melanin, providing a certain resistance to UV light  
17 due to their aptitude to absorb a wide-range of the electromagnetic spectrum, avoiding as a  
18 consequence a photo-induced damage (Nosanchuk and Casadevall, 2003). Hence, a higher  
19 pigmentation induces a higher resistance to UV radiation (Esbelin et al., 2013), and an  
20 exposure to high-intensity irradiations (UVC:100-280 nm) can prompt a deactivation of the  
21 microbial activity (Esbelin et al., 2013; Pigeot-remy et al., 2013; Schumacher and  
22 Schumacher, 2017). Nonetheless, it was found that the microbial activity's inactivation is not  
23 effective under lower-intensity irradiations (UVB: 290-320 nm and UVA: 320-400  
24 nm)(Benabbou et al., 2007). In fact, exposure to UV light (mainly UVC) induces DNA  
25 damage and the production of reactive organic species (ROS) (Schumacher and Schumacher,  
26 2017). Thus, the observed increase in the emission of certain mVOCs (**Figure 6**) are linked to  
27 a self-protection mechanism in response to stress due to the irradiation.

28 These observations suggest that surfaces covered by molds can be a source of a wide  
29 range of volatile compounds exhibiting a very complex chemistry, which is affected in part by  
30 indoor light.

31 Moreover, one should note that the UV fraction of sunlight penetrating in indoor spaces  
32 ranges from 330 up to 400 nm depending on the location, time of the year, room orientation as  
33 well as the size and type of the glass window (Gandolfo et al., 2016). Therefore the

1 wavelength of our LED corresponds to the lowest possible wavelength encountered indoor.  
 2 Also our photon flux of  $3.8 \times 10^{12}$  photons  $\text{cm}^{-2}\text{s}^{-1}$  is lower than the direct indoor UV actinic  
 3 flux (with windows closed) measured by Gandolfo et al. (i.e.,  $10^{14}$  photons  $\text{cm}^{-2}\text{s}^{-1}$ ) but  
 4 significantly higher than the one in the shadow in one room (i.e.,  $10^{10}$  photons  $\text{cm}^{-2}\text{s}^{-1}$ ).  
 5 Nevertheless, our experimental conditions can be somehow representative of realistic indoor  
 6 conditions during sunny periods. Thus, it will be interesting in future studies to evaluate the  
 7 influence of different type of indoor light sources with different intensities (Halogen,  
 8 incandescent, fluorescent lamps...) on the emission of mVOCs from *Aspergillus niger* as well  
 9 as other type of molds.



10  
 11 **Figure 6:** Temporal profile of the mVOC concentration (ppb) with and without Light under  
 12 dry air after 5 days of incubation; yellow area corresponds to light irradiation. Tentatively  
 13 identified mVOCs as follow: a) acetaldehyde ( $\text{C}_2\text{H}_4\text{O}$ ) and methanol ( $\text{CH}_4\text{O}$ ); b) formamide  
 14 ( $\text{CH}_3\text{NO}$ ), furan ( $\text{C}_4\text{H}_4\text{O}$ ) and propene ( $\text{C}_3\text{H}_6$ ); c) nitromethane ( $\text{CH}_3\text{NO}_2$ ), methanol cluster  
 15 ( $\text{CH}_4\text{O}-\text{H}_2\text{O}$ ) and propadiene ( $\text{C}_3\text{H}_4$ ); d) methanethiol ( $\text{CH}_4\text{S}$ ), dimethyl sulfide ( $\text{C}_2\text{H}_6\text{S}$ ) and  
 16 2-pentanone ( $\text{C}_5\text{H}_{10}\text{O}$ ).

17  
 18 Conclusively, this current study alongside previous ones show that the mVOCs  
 19 emission profiles depend on the cultivation time, type of substrate (malt agar extract,  
 20 wallpaper, DG 18 agar, glass fabric) and moistness of the substrate. Nevertheless, in this  
 21 study, the effect of both the air relative humidity and indoor light on the emission profiles of

1 mVOCs regardless the moistness of the substrate was also confirmed. Thus, these findings  
2 will be useful for a better understanding of the molds under indoor environmental conditions.

3

#### 4 **3.4. Estimation of mVOCs concentrations in indoor environment**

5 mVOC's indoor levels depend on the production rates, absorption to and desorption  
6 from building materials and furniture, moisture level, substrate, molds population, area of the  
7 building/room, ventilation rate as well as the chemical reactivity of the emitted VOC with the  
8 indoor oxidants. Thus the concentration of indoor mVOCs can be determined according to the  
9 description by Willem et al., (2013):

$$\frac{dC}{dt} = \frac{E_{mVOC} \times S_f}{V_r} + N_h(C_{out} - C) \quad (\text{Eq. 2})$$

10 where  $E_{mVOC}$  is the emission rate of each mVOC in  $\mu\text{g}/\text{m}^2/\text{h}$ ,  $S_f$  is the floor surface area of a  
11 room ( $\text{m}^2$ ),  $V_r$  is the room volume ( $\text{m}^3$ ),  $N_h$  is the air exchange rate ( $\text{h}^{-1}$ ),  $C_{out}$  and  $C$  are the  
12 concentrations of mVOC outdoor and indoor respectively.

13 Assuming a static set of mVOC sources over some period of time and a negligible  
14 contribution of outdoor sources to indoor concentrations, the steady state indoor concentration  
15 of mVOCs emitted from molds covering an entire surface can be written as:

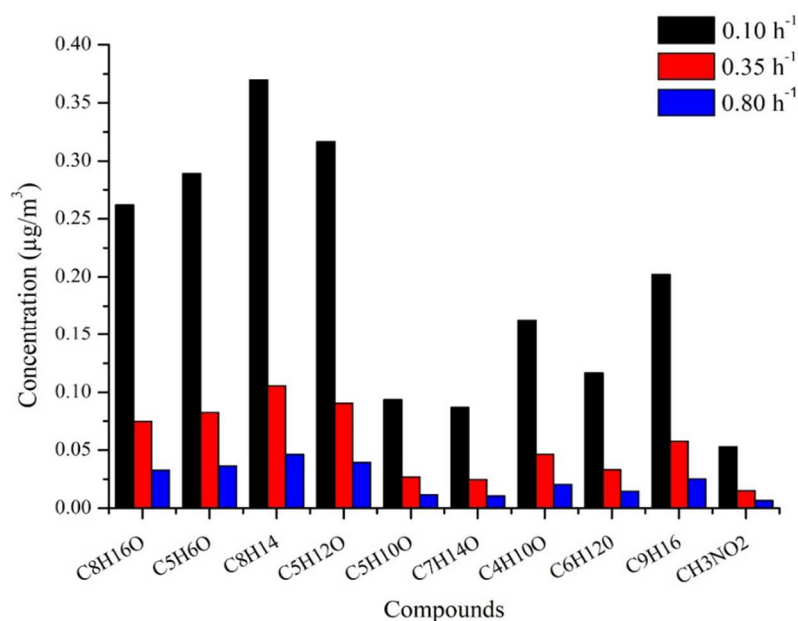
$$C = \frac{E_{mVOC} \times S_f}{N_h \times V_r} \quad (\text{Eq. 3})$$

16 **Eq. 3** can then be simplified to take into account the emission of mVOC from several  
17 damaged materials having different surfaces coverages ( $S_1, S_2, S_3 \dots$ ) and emission factors ( $E_1,$   
18  $E_2, E_3 \dots$ ) as follow (Bluyssen et al., 1997):

$$C = \frac{S_f}{N_h \times V_r} \sum_i E_{mVOC\ i} \times \frac{S_i}{S_f} \quad (\text{Eq. 4})$$

19

20 Thus, knowing that the air exchange rate within a household varies from  $0.1 \text{ h}^{-1}$  (low  
21 ventilation) up to  $0.8 \text{ h}^{-1}$  (high ventilation), an example for the indoor variation of some  
22 tracers compounds of *Aspergillus niger* activity for three air exchange rates on a scale of a  
23 standard living room ( $S_r = 25 \text{ m}^2$ ,  $V_r = 62.5 \text{ m}^3$ ) with a mold damaged surface of  $0.25 \text{ m}^2$  is  
24 shown in **Figure 7**.



1  
 2 **Figure 7:** Variation of some mVOCs concentrations for different air exchange rate 0.1, 0.35  
 3 and 0.8 h<sup>-1</sup> for 5 days of incubation time and for a molds-damaged surface of 0.25 m<sup>2</sup>

4  
 5 The above discussion has been made on a simplified basis as we did not  
 6 consider the possible chemical sinks. In fact, all the tracers presented in **Figure 7**, as well as  
 7 other mVOCs, can either be eliminated through ventilation or following chemical reactivity  
 8 with indoor oxidants like hydroxyl radicals and ozone. A calculation of their lifetimes was  
 9 performed by taken an average concentration of OH radicals and ozone equal to  $2 \times 10^6$   
 10 molecules cm<sup>-3</sup> and  $4.93 \times 10^{11}$  molecules cm<sup>-3</sup> (corresponding to 20 ppb of ozone),  
 11 respectively (Alvarez et al., 2013; Nicolas et al., 2012; Salonen et al., 2018). The calculated  
 12 lifetimes for those tracers, presented in **Figure 7**, are summarized in **Table 3**. We note that  
 13 they are all larger than the ventilation rate considered here but some mVOCs have short  
 14 lifetimes (1-5 hours) indicating that their reaction with OH radicals and O<sub>3</sub> can also be an  
 15 efficient removal process. Accordingly, from **Figure 7 and Table 3**, it is possible to conclude  
 16 that the indoor mVOCs concentrations are mainly affected by the ventilation rate but also by  
 17 the reactivity of these compounds with the indoor oxidants. A lower ventilation rate induces  
 18 higher concentration of mVOCs in indoor environment.

19  
 20



1 **Table 3:** Calculated lifetimes of some mVOCs with OH radicals and O<sub>3</sub>

Compound	$\tau_{OH}$	$\tau_{O_3}$
C <sub>8</sub> H <sub>16</sub> O: 2-octanone	12.6 h <sup>a</sup>	-
C <sub>5</sub> H <sub>6</sub> O: 3-methylfuran	1.5 h <sup>b</sup>	1.1 d <sup>b'</sup>
C <sub>5</sub> H <sub>6</sub> O: 2-methylfuran	2.3 h <sup>c</sup>	
C <sub>5</sub> H <sub>12</sub> O: 2-pentanol	11.7 h <sup>d</sup>	
C <sub>5</sub> H <sub>10</sub> O: 4-methylbutanal	5.0 h <sup>e</sup>	602 d <sup>e'</sup>
C <sub>5</sub> H <sub>10</sub> O: 2-pentanone	2.5 d <sup>f</sup>	
C <sub>7</sub> H <sub>14</sub> O: heptanal	4.7 h <sup>g</sup>	
C <sub>7</sub> H <sub>14</sub> O: 2-heptanone	21.8 h <sup>h</sup>	
C <sub>4</sub> H <sub>10</sub> O: 1-butanol	16.4 h <sup>i</sup>	
C <sub>6</sub> H <sub>12</sub> O: 3-hexen-1-ol	1.3 h <sup>j</sup>	8.8 h <sup>j</sup>
C <sub>6</sub> H <sub>12</sub> O: 2-hexanone	22.5 h <sup>h</sup>	
C <sub>6</sub> H <sub>12</sub> O: hexanal	5.0 h <sup>k</sup>	
CH <sub>3</sub> NO <sub>2</sub>	366 d $\approx$ 1 y <sup>l</sup>	1973 d $\approx$ 5.4 y <sup>l'</sup>

2 Reaction rate constants taken from: a:(Wallington and Kurylo, 1987),b:(Atkinson et al.,  
 3 1989), b' :(Alvarado et al., 1996), c:(Bierbach et al., 1992), d: (Baxley and Wells, 1998),  
 4 e:(D'Anna et al., 2001), e':(Glasius et al., 1997), f:(Alvarez-Idaboy et al., 2004),  
 5 g:(Albaladejo et al., 2002), h:(Jiménez et al., 2005), i:(Yujing and Mellouki, 2001),  
 6 j:(Atkinson et al., 1995),k:(Jiménez et al., 2007),l:(Liu et al., 1990), l':(Hastie et al., 1976).

7

8 Nevertheless, the indoor environment is complex with the presence of different VOCs  
 9 sources like building materials, furnishing, human activities, microbial activity affecting  
 10 accordingly their concentration levels (Claeson, 2006). The total count of VOC (TVOC)  
 11 concentrations are generally beneath 0.5 mg/m<sup>3</sup>, while the concentration of single compounds  
 12 is usually below 5 µg/m<sup>3</sup> and hardly exceeds 50 µg/m<sup>3</sup> (Claeson, 2006). Willem et al. (2013)  
 13 measured the indoor concentrations of VOCs emitted from homes with different building  
 14 materials and furniture and for different air exchange rates. For a low ventilation rate (0.2 h<sup>-1</sup>),  
 15 the concentrations were: 4.4-5.1 µg/m<sup>3</sup> for alcohols, 0.4-57.3 µg/m<sup>3</sup> for C4-C10 aldehydes,  
 16 1.1-2.7 µg/m<sup>3</sup> for C7-C16 alkanes, 0.2-4.9 µg/m<sup>3</sup> for aromatics, 0.2-21.1 µg/m<sup>3</sup> for terpenes.  
 17 On the other hand, other studies that focused on mVOCs measurements in dwellings with  
 18 microbial damage found that their respective individual concentrations generally range from  
 19 0.1 to 10 µg/m<sup>3</sup> (Korpi et al., 2009). It is noteworthy to mention that the detection of any  
 20 tracer like 3-methylfuran, 1-octen-3-ol and dimethyl disulfide having a concentration higher  
 21 than 0.05 µg/m<sup>3</sup> implies a microbial activity in the indoor environment (Korpi et al., 2009).

## 1        **4. Conclusion**

2            mVOCs emission from molds induce a poor indoor air quality and are used to detect  
3 the microbial activity in dwellings. Therefore, in this study, the emission of mVOCs from  
4 *Aspergillus niger* (one of the most common molds detected in indoor environment) cultivated  
5 on malt agar extract was investigated at different cultivation time for several indoor relative  
6 humidities and light irradiations. Several PCAs were performed and have shown the  
7 variability of the mVOCs emission pattern for different cultivation time and relative  
8 humidities. Some mVOCs were detected like 1-octen-3-ol, heptanol, methyl-furan, methyl-1-  
9 butanol, dimethyl sulfide, dimethyl disulfide, nitromethane, 1,3-octadiene... considered as  
10 tracers of the microbial activity. Results have shown the sharp decrease of the concentrations  
11 of tracers after 10-14 days of cultivation time, and indoor light seems to promote the  
12 concentration of specific mVOCs as a response to stress. The concentration of the tracer's  
13 compounds of the fungal activity was also estimated in a scale of a room for different air  
14 exchange rate and their corresponding indoor lifetimes toward OH radicals and ozone were  
15 also calculated. This study allows a better understanding of the influence of different  
16 parameters (molds cultivation time, indoor relative humidity and light) on the mVOCs  
17 emission profiles and levels from molds' type *Aspergillus niger*. This will be useful in the  
18 early detection of the microbial activity in sick buildings, nevertheless, for a more generalized  
19 quantitative results, tests on several types of molds grown on different surfaces should be  
20 tested under wider range of indoor conditions (including diverse light wavelengths).

## 21        **Acknowledgments**

22        This work was supported by the Alfred P. Sloan Foundation under its Chemistry of Indoor  
23        Environments program.

## 24        **Author contribution**

25        CK performed the experimental measurements and data analysis with the help of LA. BL and  
26        SP helped conceiving the experimental setup. CG supervised the work. CK, LA and AD  
27        discussed the results and edited the manuscript with the help of all authors.

## 28        **Conflict of interest**

29        The authors declare no conflict of interest.

## 1            References

- 2    Abis, L., Loubet, B., Ciuraru, R., Lafouge, F., Dequiedt, S., Houot, S., Maron, P.A.,  
3            Bourgeteau-Sadet, S., 2018. Profiles of volatile organic compound emissions from soils  
4            amended with organic waste products. *Sci. Total Environ.* 636, 1333–1343.  
5            <https://doi.org/10.1016/j.scitotenv.2018.04.232>
- 6    Abis, L., Loubet, B., Ciuraru, R., Lafouge, F., Houot, S., Nowak, V., Tripied, J., Dequiedt, S.,  
7            Maron, P.A., Sadet-Bourgeteau, S., 2020. Reduced microbial diversity induces larger  
8            volatile organic compound emissions from soils. *Sci. Rep.* 10, 6104.  
9            <https://doi.org/doi/10.15454/4W9DM2>
- 10   Adams, R.I., Lymperopoulou, D.S., Misztal, P.K., De Cassia Pessotti, R., Behie, S.W., Tian,  
11            Y., Goldstein, A.H., Lindow, S.E., Nazaroff, W.W., Taylor, J.W., Traxler, M.F., Bruns,  
12            T.D., 2017. Microbes and associated soluble and volatile chemicals on periodically wet  
13            household surfaces. *Microbiome* 5, 128. <https://doi.org/10.1186/s40168-017-0347-6>
- 14   Albaladejo, J., Ballesteros, B., Jiménez, E., Martín, P., Martínez, E., 2002. A PLP-LIF kinetic  
15            study of the atmospheric reactivity of a series of C4-C7 saturated and unsaturated  
16            aliphatic aldehydes with OH. *Atmos. Environ.* 36, 3231–3239.  
17            [https://doi.org/10.1016/S1352-2310\(02\)00323-0](https://doi.org/10.1016/S1352-2310(02)00323-0)
- 18   Alvarado, A., Atkinson, R., Arey, J., 1996. Kinetics of the gas-phase reactions of NO<sub>3</sub>  
19            radicals and O<sub>3</sub> with 3-methylfuran and the OH radical yield from the O<sub>3</sub> reaction. *Int. J.*  
20            *Chem. Kinet.* 28, 905–909. [https://doi.org/10.1002/\(SICI\)1097-](https://doi.org/10.1002/(SICI)1097-4601(1996)28:12<905::AID-KIN7>3.0.CO;2-R)  
21            [4601\(1996\)28:12<905::AID-KIN7>3.0.CO;2-R](https://doi.org/10.1002/(SICI)1097-4601(1996)28:12<905::AID-KIN7>3.0.CO;2-R)
- 22   Alvarez-Idaboy, J.R., Cruz-Torres, A., Galano, A., Ruiz-Santoyo, M.E., 2004. Structure-  
23            Reactivity Relationship in Ketones + OH Reactions: A Quantum Mechanical and TST  
24            Approach. *J. Phys. Chem. A* 108, 2740–2749. <https://doi.org/10.1021/jp036795o>
- 25   Alvarez, E.G., Amedro, D., Afif, C., Gligorovski, S., Schoemacker, C., Fittschen, C., Doussin,  
26            J.F., Wortham, H., 2013. Unexpectedly high indoor hydroxyl radical concentrations  
27            associated with nitrous acid. *Proc. Natl. Acad. Sci. U. S. A.* 110, 13294–13299.  
28            <https://doi.org/10.1073/pnas.1308310110>
- 29   Atkinson, R., Arey, J., Aschmann, S.M., Corchnoy, S.B., Shu, Y., 1995. Rate constants for the  
30            gas-phase reactions of cis-3-Hexen-1-ol, cis-3-Hexenylacetate, trans-2-Hexenal, and  
31            Linalool with OH and NO<sub>3</sub> radicals and O<sub>3</sub> at 296 ± 2 K, and OH radical formation  
32            yields from the O<sub>3</sub> reactions. *Int. J. Chem. Kinet.* 27, 941–955.  
33            <https://doi.org/10.1002/kin.550271002>

- 1 Atkinson, R., Aschmann, S.M., Tuazon, E.C., Arey, J., Zielinska, B., 1989. Formation of 3-  
2 Methylfuran from the gas-phase reaction of OH radicals with isoprene and the rate  
3 constant for its reaction with the OH radical. *Int. J. Chem. Kinet.* 21, 593–604.  
4 <https://doi.org/10.1002/kin.550210709>
- 5 Baxley, J.S., Wells, J.R., 1998. The hydroxyl radical reaction rate constant and atmospheric  
6 transformation products of 2-butanol and 2-pentanol. *Int. J. Chem. Kinet.* 30, 745–752.  
7 [https://doi.org/10.1002/\(SICI\)1097-4601\(1998\)30:10<745::AID-KIN7>3.0.CO;2-V](https://doi.org/10.1002/(SICI)1097-4601(1998)30:10<745::AID-KIN7>3.0.CO;2-V)
- 8 Benabbou, A.K., Derriche, Z., Felix, C., Lejeune, P., Guillard, C., 2007. Photocatalytic  
9 inactivation of *Escherichia coli*. Effect of concentration of TiO<sub>2</sub> and microorganism,  
10 nature, and intensity of UV irradiation. *Appl. Catal. B Environ.* 76, 257–263.  
11 <https://doi.org/10.1016/j.apcatb.2007.05.026>
- 12 Bierbach, A., Barnes, I., Becker, K.H., 1992. Rate coefficients for the gas-phase reactions of  
13 hydroxyl radicals with furan, 2-methylfuran, 2-ethylfuran and 2,5-dimethylfuran at 300 +  
14 2 K. *Atmos. Environ.* 26, 813–817.
- 15 Block, S.S., 1953. Humidity requirements for mold growth. *Appl. Microbiol.* 1, 287–293.  
16 <https://doi.org/10.1128/aem.1.6.287-293.1953>
- 17 Bluysen, P., Cochet, C., Fischer, M., Knoppel, H., Levy, L., Lundgren, B., Maroni, M.,  
18 Molhave, L., Rothweiler, H., Saarela, K., Seifer, B., 1997. Evaluation of VOC Emissions  
19 from Building Products Solid Flooring Materials.
- 20 Boots, A.W., Smolinska, A., Van Berkel, J.J.B.N., Fijten, R.R.R., Stobberingh, E.E.,  
21 Boumans, M.L.L., Moonen, E.J., Wouters, E.F.M., Dallinga, J.W., Van Schooten, F.J.,  
22 2014. Identification of microorganisms based on headspace analysis of volatile organic  
23 compounds by gas chromatography-mass spectrometry. *J. Breath Res.* 8.  
24 <https://doi.org/10.1088/1752-7155/8/2/027106>
- 25 Bos, L.D.J., Sterk, P.J., Schultz, M.J., 2013. Volatile Metabolites of Pathogens: A Systematic  
26 Review. *PLoS Pathog.* 9, 1–8. <https://doi.org/10.1371/journal.ppat.1003311>
- 27 Cappellin, L., Algarra Alarcon, A., Herdinger-Blatt, I., Sanchez, J., Biasioli, F., Martin, S.T.,  
28 Loreto, F., McKinney, K.A., 2017. Field observations of volatile organic compound  
29 (VOC) exchange in red oaks. *Atmos. Chem. Phys.* 17, 4189–4207.  
30 <https://doi.org/10.5194/acp-17-4189-2017>
- 31 Cappellin, L., Biasioli, F., Granitto, P.M., Schuhfried, E., Soukoulis, C., Costa, F., Märk,  
32 T.D., Gasperi, F., 2011. On data analysis in PTR-TOF-MS: From raw spectra to data  
33 mining. *Sensors Actuators, B Chem.* 155, 183–190.  
34 <https://doi.org/10.1016/j.snb.2010.11.044>

- 1 Cappellin, L., Karl, T., Probst, M., Ismailova, O., Winkler, P.M., Soukoulis, C., Aprea, E.,  
2 Märk, T.D., Gasperi, F., Biasioli, F., 2012. On quantitative determination of volatile  
3 organic compound concentrations using proton transfer reaction time-of-flight mass  
4 spectrometry. *Environ. Sci. Technol.* 46, 2283–2290. <https://doi.org/10.1021/es203985t>
- 5 Chen, B., 2002. Determination of Unique Microbial Volatile Organic Compounds Produced  
6 by Five *Aspergillus* Species Commonly Found in Problem Buildings Determination of  
7 unique microbial volatile organic compounds produced ... *AIHA J.* 63, 135–140.  
8 <https://doi.org/10.1080/15428110208984696>
- 9 Chen, F., Yang, X., Wu, Q., 2009. Antifungal capability of TiO<sub>2</sub> coated film on moist wood.  
10 *Build. Environ.* 44, 1088–1093. <https://doi.org/10.1016/j.buildenv.2008.07.018>
- 11 Claeson, A.-S., 2006. Volatile organic compounds from microorganisms - identification and  
12 health effects. *Public Health.* Umeå, Sweden.
- 13 Claeson, A.S., Levin, J.O., Blomquist, G., Sunesson, A.L., 2002. Volatile metabolites from  
14 microorganisms grown on humid building materials and synthetic media. *J. Environ.*  
15 *Monit.* 4, 667–672. <https://doi.org/10.1039/b202571j>
- 16 Claeson, A.S., Sandström, M., Sunesson, A.L., 2007. Volatile organic compounds (VOCs)  
17 emitted from materials collected from buildings affected by microorganisms. *J. Environ.*  
18 *Monit.* 9, 240–245. <https://doi.org/10.1039/b614766f>
- 19 Costa, C.P., Gonçalves Silva, D., Rudnitskaya, A., Almeida, A., Rocha, S.M., 2016. Shedding  
20 light on *Aspergillus Niger* volatile exometabolome. *Sci. Rep.* 6, 1–13.  
21 <https://doi.org/10.1038/srep27441>
- 22 D’Anna, B., Andresen, Gefen, Z., Nielsen, C.J., 2001. Kinetic study of OH and NO<sub>3</sub> radical  
23 reactions with 14 aliphatic aldehydes. *Phys. Chem. Chem. Phys.* 3, 3057–3063.  
24 <https://doi.org/10.1039/b103623h>
- 25 de Gouw, J.A., Goldan, P.D., Warneke, C., Kuster, W.C., Roberts, J.M., Marchewka, M.,  
26 Bertman, S.B., Pszenny, A.A.P., Keene, W.C., 2003. Validation of proton transfer  
27 reaction-mass spectrometry (PTR-MS) measurements of gas-phase organic compounds  
28 in the atmosphere during the New England Air Quality Study (NEAQS) in 2002. *J.*  
29 *Geophys. Res. Atmos.* 108, 1–18. <https://doi.org/10.1029/2003jd003863>
- 30 Dimitroulopoulou, C., 2012. Ventilation in European dwellings: A review. *Build. Environ.* 47,  
31 109–125. <https://doi.org/10.1016/j.buildenv.2011.07.016>
- 32 Esbelin, J., Mallea, S., Ram, A.F.J., 2013. Role of Pigmentation in Protecting *Aspergillus*  
33 *niger* Conidiospores Against Pulsed Light Radiation. *Photochem. Photobiol.* 89, 758–  
34 761. <https://doi.org/10.1111/php.12037>

1 Fiedler, K., Schutz, E., Geh, S., 2001. Detection of microbial volatile organic compounds  
2 (MVOCs) produced by moulds on various materials. *Int. J. Hyg. Environ. Health* 204,  
3 111–121.

4 Gandolfo, A., Gligorovski, V., Bartolomei, V., Tlili, S., Gómez Alvarez, E., Wortham, H.,  
5 Kleffmann, J., Gligorovski, S., 2016. Spectrally resolved actinic flux and photolysis  
6 frequencies of key species within an indoor environment. *Build. Environ.* 109, 50–57.  
7 <https://doi.org/10.1016/j.buildenv.2016.08.026>

8 Gardes, M., Bruns, T.D., 1996. ITS-RFLP matching for identification of fungi., in: *Methods*  
9 *in Molecular Biology* (Clifton, N.J.). pp. 177–186. [https://doi.org/10.1385/0-89603-323-](https://doi.org/10.1385/0-89603-323-6:177)  
10 [6:177](https://doi.org/10.1385/0-89603-323-6:177)

11 Gerritsen, M.G., Brinkman, P., Escobar, N., Bos, L.D., De Heer, K., Meijer, M., Janssen,  
12 H.G., De Cock, H., Wösten, H.A., Visser, C.E., Van Oers, M.H., Sterk, P.J., 2018.  
13 Profiling of volatile organic compounds produced by clinical *Aspergillus* isolates using  
14 gas chromatography-mass spectrometry. *Med. Mycol.* 56, 253–256.  
15 <https://doi.org/10.1093/mmy/myx035>

16 Glasius, M., Calogirou, A., Jensen, N.R., Hjorth, J., Nielsen, C.J., 1997. Kinetic study of gas-  
17 phase reactions of pinonaldehyde and structurally related compounds. *Int. J. Chem.*  
18 *Kinet.* 29, 527–533. [https://doi.org/10.1002/\(SICI\)1097-4601\(1997\)29:7<527::AID-](https://doi.org/10.1002/(SICI)1097-4601(1997)29:7<527::AID-KIN7>3.0.CO;2-W)  
19 [KIN7>3.0.CO;2-W](https://doi.org/10.1002/(SICI)1097-4601(1997)29:7<527::AID-KIN7>3.0.CO;2-W)

20 Hastie, D.R., Freeman, C.G., McEwan, M.J., Schiff, H.I., 1976. The reactions of ozone with  
21 methyl and ethyl nitrites. *Int. J. Chem. Kinet.* 8, 307–313.  
22 <https://doi.org/10.1002/kin.550080212>

23 Hurraß, J., Heinzow, B., Aurbach, U., Bergmann, K.C., Bufe, A., Buzina, W., Cornely, O.A.,  
24 Engelhart, S., Fischer, G., Gabrio, T., Heinz, W., Herr, C.E.W., Kleine-Tebbe, J.,  
25 Klimek, L., Köberle, M., Lichtnecker, H., Lob-Corzilius, T., Merget, R., Mülleneisen,  
26 N., Nowak, D., Rabe, U., Raulf, M., Seidl, H.P., Steiß, J.O., Szewczyk, R., Thomas, P.,  
27 Valtanen, K., Wiesmüller, G.A., 2017. Medical diagnostics for indoor mold exposure.  
28 *Int. J. Hyg. Environ. Health* 220, 305–328. <https://doi.org/10.1016/j.ijheh.2016.11.012>

29 Jiménez, E., Ballesteros, B., Martínez, E., Albaladejo, J., 2005. Tropospheric reaction of OH  
30 with selected linear ketones: Kinetic studies between 228 and 405 K. *Environ. Sci.*  
31 *Technol.* 39, 814–820. <https://doi.org/10.1021/es049333c>

32 Jiménez, E., Lanza, B., Martínez, E., Albaladejo, J., 2007. Daytime tropospheric loss of  
33 hexanal and trans-2-hexenal: OH kinetics and UV photolysis. *Atmos. Chem. Phys.* 7,  
34 1565–1574. <https://doi.org/10.5194/acp-7-1565-2007>

- 1 Korpi, A., Järnberg, J., Pasanen, A.L., 2009. Microbial volatile organic compounds. *Crit. Rev.*  
2 *Toxicol.* 39, 139–193. <https://doi.org/10.1080/10408440802291497>
- 3 Kozicki, M., Wiejak, A., Piasecki, M., Abram, A., 2019. Identification of MVOCs produced  
4 by *Coniophora puteana* and *Poria placenta* growing on WPC boards by using subtraction  
5 mass spectra. *Int. J. Environ. Res. Public Health* 16, 2499.  
6 <https://doi.org/10.3390/ijerph16142499>
- 7 Lavine, B.K., Mirjankar, N., LeBouf, R., Rossner, A., 2012. Prediction of mold contamination  
8 from microbial volatile organic compound profiles using head space gas  
9 chromatography/mass spectrometry. *Microchem. J.* 103, 119–124.  
10 <https://doi.org/10.1016/j.microc.2012.01.017>
- 11 Lindinger, W., Hansel, A., Jordan, A., 1998. On-line monitoring of volatile organic  
12 compounds at pptv levels by means of Proton-Transfer-Reaction Mass Spectrometry  
13 (PTR-MS) Medical applications, food control and environmental research. *Int. J. Mass*  
14 *Spectrom. Ion Process.* 173, 191–241. [https://doi.org/10.1016/s0168-1176\(97\)00281-4](https://doi.org/10.1016/s0168-1176(97)00281-4)
- 15 Liu, R., Huie, R.E., Kurylo, M.J., Nielsen, O.J., 1990. The gas phase reactions of hydroxyl  
16 radicals with a series of nitroalkanes over the temperature range 240–400 K. *Chem. Phys.*  
17 *Lett.* 167, 519–523. [https://doi.org/10.1016/0009-2614\(90\)85462-L](https://doi.org/10.1016/0009-2614(90)85462-L)
- 18 Markowska-Szczupak, A., Ulfig, K., Grzmił, B., Morawski, A., 2010. A preliminary study on  
19 antifungal effect of TiO<sub>2</sub>-based paints in natural indoor light. *Polish J. Chem. Technol.*  
20 12, 53–57. <https://doi.org/10.2478/v10026-010-0050-x>
- 21 Matysik, S., Herbarth, O., Mueller, A., 2009. Determination of microbial volatile organic  
22 compounds (MVOCs) by passive sampling onto charcoal sorbents. *Chemosphere* 76,  
23 114–119. <https://doi.org/10.1016/j.chemosphere.2009.02.010>
- 24 Matysik, S., Herbarth, O., Mueller, A., 2008. Determination of volatile metabolites  
25 originating from mould growth on wall paper and synthetic media. *J. Microbiol. Methods*  
26 75, 182–187. <https://doi.org/10.1016/j.mimet.2008.05.027>
- 27 Meruva, N.K., Penn, J.M., Farthing, D.E., 2004. Rapid identification of microbial VOCs from  
28 tobacco molds using closed-loop stripping and gas chromatography/time-of-flight mass  
29 spectrometry. *J. Ind. Microbiol. Biotechnol.* 31, 482–488.  
30 <https://doi.org/10.1007/s10295-004-0175-0>
- 31 Morath, S.U., Hung, R., Bennett, J.W., 2012. Fungal volatile organic compounds: A review  
32 with emphasis on their biotechnological potential. *Fungal Biol. Rev.* 26, 73–83.  
33 <https://doi.org/10.1016/j.fbr.2012.07.001>
- 34 Moularat, S., Robine, E., Ramalho, O., Oturan, M.A., 2008a. Detection of fungal development

1 in closed spaces through the determination of specific chemical targets. *Chemosphere* 72,  
2 224–232. <https://doi.org/10.1016/j.chemosphere.2008.01.057>

3 Moularat, S., Robine, E., Ramalho, O., Oturan, M.A., 2008b. Detection of fungal  
4 development in a closed environment through the identification of specific VOC:  
5 Demonstration of a specific VOC fingerprint for fungal development. *Sci. Total Environ.*  
6 407, 139–146. <https://doi.org/10.1016/j.scitotenv.2008.08.023>

7 Nicolas, M., Ramalho, O., Maupetit, F., 2012. Impact of ozone on indoor air quality: a  
8 preliminary field study. *7th Int. Conf. Heal. Build.* 2003, 247–252.

9 Nosanchuk, J.D., Casadevall, A., 2003. Microreview The contribution of melanin to microbial  
10 pathogenesis. *Cell. Microbiol.* 5, 203–223.

11 Pigeot-remy, S., Real, P., Simonet, F., Hernandez, C., Vallet, C., Lazzaroni, J.C., 2013.  
12 Applied Catalysis B: Environmental Inactivation of *Aspergillus niger* spores from  
13 indoor air by photocatalytic filters 135, 167–173.

14 Pigeot-Rémy, S., Simonet, F., Errazuriz-Cerda, E., Lazzaroni, J.C., Atlan, D., Guillard, C.,  
15 2011. Photocatalysis and disinfection of water: Identification of potential bacterial  
16 targets. *Appl. Catal. B Environ.* 104, 390–398.  
17 <https://doi.org/10.1016/j.apcatb.2011.03.001>

18 Pitt, R.E., 1993. A Descriptive Model of Mold Growth and Aflatoxin Formation as Affected  
19 by Environmental Conditions. *J. Food Prot.* 56, 139–146. [https://doi.org/10.4315/0362-](https://doi.org/10.4315/0362-028x-56.2.139)  
20 [028x-56.2.139](https://doi.org/10.4315/0362-028x-56.2.139)

21 Polizzi, V., Adams, A., De Saeger, S., Van Peteghem, C., Moretti, A., De Kimpe, N., 2012.  
22 Influence of various growth parameters on fungal growth and volatile metabolite  
23 production by indoor molds. *Sci. Total Environ.* 414, 277–286.  
24 <https://doi.org/10.1016/j.scitotenv.2011.10.035>

25 Potard, K., 2017. Les émissions de composés organiques volatiles (COVs) des sols dans les  
26 paysages agricoles: identification des sources et incidences sur la qualité de l'air.  
27 Université Rennes 1.

28 RAY, A.C., EAKIN, R.E., 1975. Studies on the Biosynthesis of Aspergillin by *Aspergillus*  
29 *niger*. *Appl. Microbiol.* 30, 909–915.

30 Salonen, H., Salthammer, T., Morawska, L., 2018. Human exposure to ozone in school and  
31 office indoor environments. *Environ. Int.* 119, 503–514.  
32 <https://doi.org/10.1016/j.envint.2018.07.012>

33 Sánchez, B., Sánchez-Muñoz, M., Muñoz-Vicente, M., Cobas, G., Portela, R., Suárez, S.,  
34 González, A.E., Rodríguez, N., Amils, R., 2012. Photocatalytic elimination of indoor air



1 biological and chemical pollution in realistic conditions. *Chemosphere* 87, 625–630.  
2 <https://doi.org/10.1016/j.chemosphere.2012.01.050>

3 Schumacher, J., Schumacher, J., 2017. How light affects the life of *Botrytis*. *Fungal Genet.*  
4 *Biol.* <https://doi.org/10.1016/j.fgb.2017.06.002>

5 Shehu, K., Bello, M.T., 2011. Effect of Environmental Factors on the Growth of *Aspergillus*  
6 *Species Associated with Stored Millet Grains in Sokoto.* *Niger. J. basic Appl. Sci.* 19,  
7 218–223.

8 Siddiquee, S., Azad, S. Al, Abu Bakar, F., Naher, L., Vijay Kumar, S., 2015. Separation and  
9 identification of hydrocarbons and other volatile compounds from cultures of *Aspergillus*  
10 *niger* by GC-MS using two different capillary columns and solvents. *J. Saudi Chem. Soc.*  
11 19, 243–256. <https://doi.org/10.1016/j.jscs.2012.02.007>

12 Soldatova, L.N., Rocca-Serra, P., Dumontier, M., Shah, N.H., 2014. Selected papers from the  
13 16th annual Bio-Ontologies Special Interest Group Meeting. *J. Biomed. Semantics* 5.  
14 <https://doi.org/10.1186/2041-1480-5-S1-I1>

15 Soon, A., Norback, D., Janson, C., Wieslander, G., Jogi, R., Gunnbjörnsdottir, M., Gislason,  
16 T., Sahlberg, B., 2012. Airborne molds and bacteria, microbial volatile organic  
17 compounds (MVOC), plasticizers and formaldehyde in dwellings in three North  
18 European cities in relation to sick building syndrome (SBS). *Sci. Total Environ.* 444,  
19 433–440. <https://doi.org/10.1016/j.scitotenv.2012.10.114>

20 Sunesson, A.L., Vaes, W.H.J., Nilsson, C.A., Blomquist, G., Andersson, B., Carlson, R.,  
21 1995. Identification of volatile metabolites from five fungal species cultivated on two  
22 media. *Appl. Environ. Microbiol.* 61, 2911–2918.

23 Viitanen, H., 2007. Improved Model to Predict Mold Growth in building materials, in: VTT  
24 Technical Research.

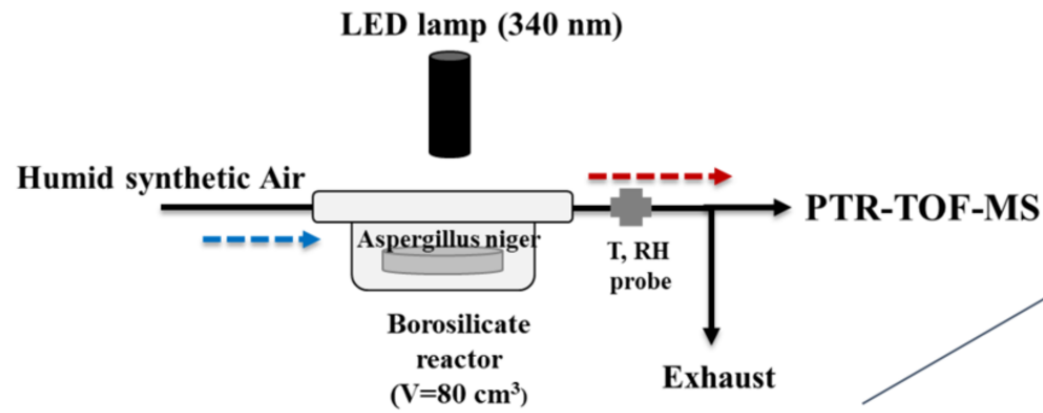
25 Wallington, T.J., Kurylo, M.J., 1987. Flash Photolysis Resonance Fluorescence Investigatlon.  
26 *J. Phys. Chem.* 91, 5050–5054.

27 Wilkins, K., Larsen, K., 1995. Variation of volatile organic compound patterns of mold  
28 species from damp buildings. *Chemosphere* 31, 3225–3236.

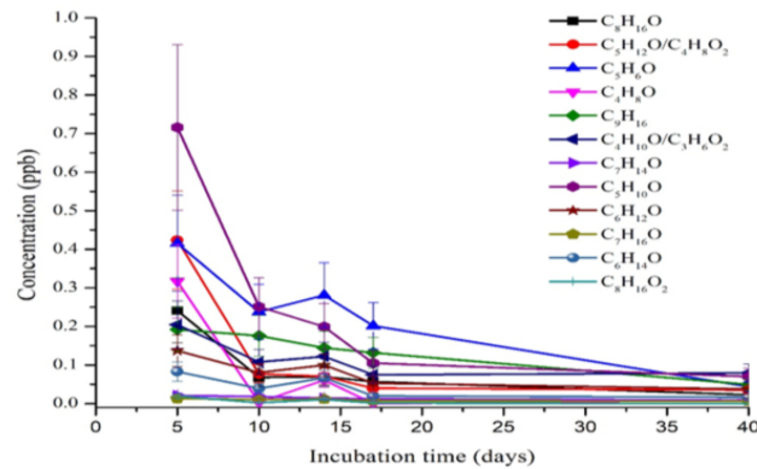
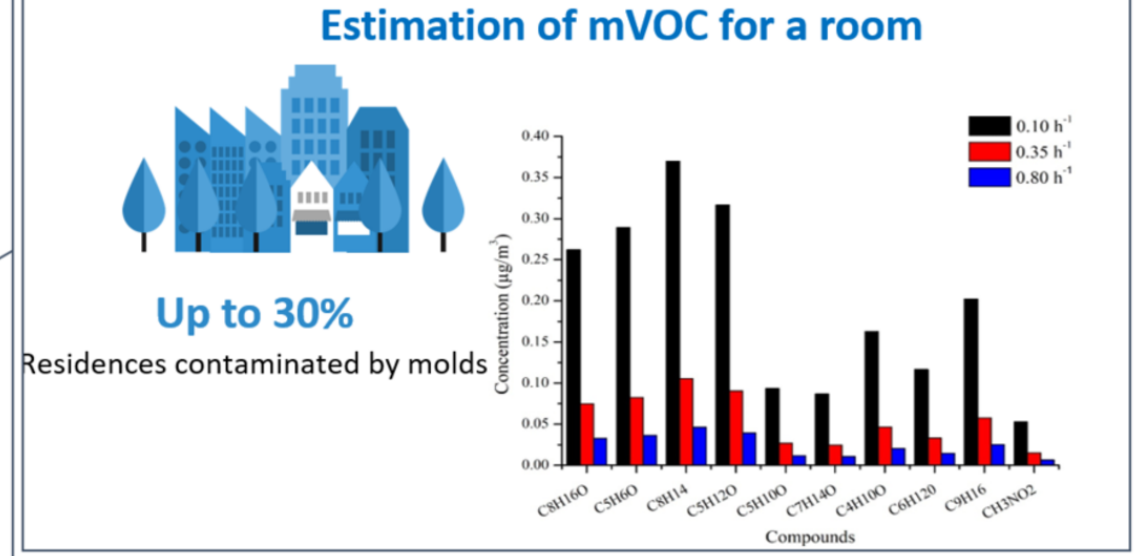
29 Wilkins, K., Larsen, K., Simkus, M., 2000. Volatile metabolites from mold growth on  
30 building materials and synthetic media. *Chemosphere* 41, 437–446.  
31 [https://doi.org/10.1016/S0045-6535\(99\)00273-8](https://doi.org/10.1016/S0045-6535(99)00273-8)

32 Willem, H., Hult, E.L., Hotchi, T., Russell, L., Maddalena, R.L., Singer, B.C., 2013.  
33 Ventilation Control of Volatile Organic Compounds in New U . S . Homes : Results of a  
34 Controlled Field Study in Nine Residential Units.

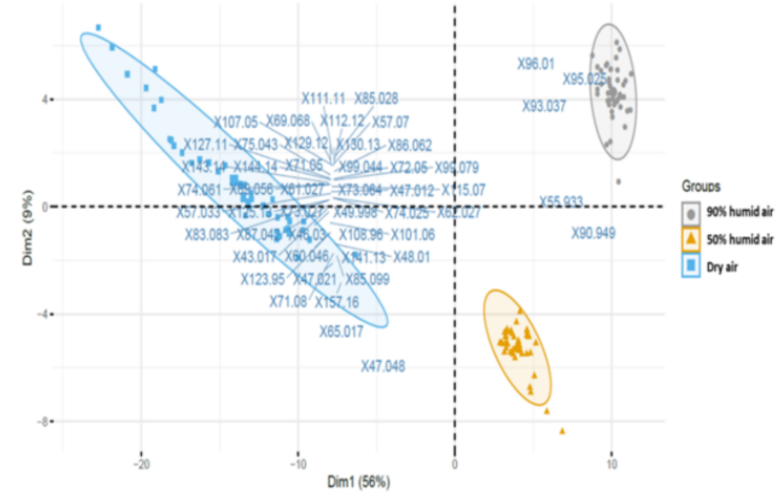
- 1 Wolkoff, P., 1998. Impact of air velocity, temperature, humidity, and air on long-term VOC  
2 emissions from building products. *Atmos. Environ.* 32, 2659–2668.  
3 [https://doi.org/10.1016/S1352-2310\(97\)00402-0](https://doi.org/10.1016/S1352-2310(97)00402-0)
- 4 Wu, Q., 2009. Photocatalytic Oxidation of Different Ultraviolet Irradiation Conditions.  
5 *Environ. Sci. Technol.* 43, 4606–4611.
- 6 Yujing, M., Mellouki, A., 2001. Temperature dependence for the rate constants of the reaction  
7 of OH radicals with selected alcohols. *Chem. Phys. Lett.* 333, 63–68.  
8 [https://doi.org/10.1016/S0009-2614\(00\)01346-4](https://doi.org/10.1016/S0009-2614(00)01346-4)  
9



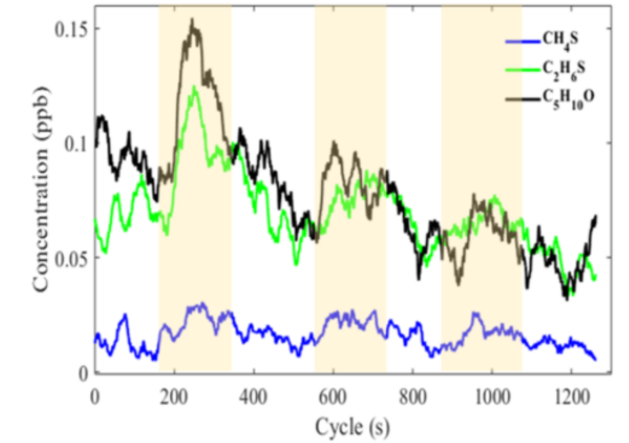
mVOCs Measurements in function of:



Incubation time



Humidity



Indoor light