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Vanessa Hörmann

Biofiltration of indoor pollutants by ornamental plants

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Content

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List of abbreviations

ATP	Adenosine triphosphate
IAQ	Indoor air quality
$NADP^+$ / NADPH	Nicotinamide adenine dinucleotide phosphate
VOC	volatile organic compounds

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Introduction

In industrialized countries people spend 80 - 90% of their time indoors (Klepeis et al., 2001; Schweizer et al., 2006). Therefore, the health and well-being of human is primary influenced by psychological variables and indoor air quality (IAQ) (Bauer et al., 1992; Ryan and Morrow, 1992; Tham, 2016). Several studies have shown that a relationship exists between the quality of indoor environmental design and the well-being and health of human (Evans and McCoy, 1998; Hongisto et al., 2016; Ryan and Morrow, 1992). For decades there has also been evidence that IAQ, especially in regard to volatile pollutants, impacts human health (Brooks et al., 1991; Jones, 1999; Tham, 2016). Pollutants are emitted by technical office equipment, human activities as well as furniture and carpets. Furthermore, the use of synthetic building materials has increased. Although these materials often provide more comfort and lower maintenance costs, they can emit a high amount of pollutants (Jones, 1999; Yu and Kim, 2010). The most common indoor pollutants are ozone (Destaillats et al., 2008; Jones, 1999), particulate matter (Destaillats et al., 2008; Wensing et al., 2008), and volatile organic compounds (VOC) (Berrios et al., 2005; Destaillats et al., 2008; Katsoyiannis et al., 2012; Que et al., 2013).

VOC's belong to the most important chemicals occurring in indoor airs. They are characterized as organic chemicals with a boiling point between 50°C and 260°C. More than 350 VOC's have already been identified indoors, including aromatic compounds (e.g. toluene and xylene), terpenes (e.g. limonene and α-pinene), alcohols (e.g. 2-ethylhexanol and butanol), and carbonyl compounds (e.g. formaldehyde and acetaldehyde) (Salthammer and Bahadir, 2009; Sarigiannis et al., 2011). Major sources of VOC's are building materials and technical equipment (Berrios et al., 2005; Brooks et al., 1991; Destaillats et al., 2008; Jones, 1999; Wolkoff and Nielsen, 2001). Board materials like particle board, plywood, and lumber emit up to 900 µg/m³ total VOC, mostly toluene and several terpenes (Que et al., 2013). In comparison, a running PC was found to emit up to 270 µg toluene, 81.9 µg styrene, 237 µg xylene, and 188 µg ethylbenzene per hour (Berrios et al., 2005). Measurements of different photocopy devices during the copying process revealed emission rates of up to 7500 µg ozone, 29000 µg xylene, 2000 µg toluene, 14000 µg 2-ethylhexanol, and 9600 µg styrene per hour (Leovic et al., 1998, 1996). More recent studies report lower emission rates for PCs and printers. Nonetheless, total chemical emissions of up to 740 µg/h for PCs and 5000 to 8400 µg/h for copy devices are still of concern (Kowalska et al., 2015; Maddalena et al., 2011). Other VOC sources are related to human activities, like cooking or the use of products

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for cleaning and personal care. Besides the emission of primary VOC's from indoor sources, indoor gas-phase reactions lead to the formation of secondary VOC's (Luengas et al., 2015). For example, 2-ethylhexanol is a derivate of di-2-ethylhexyl phthalate (DEHP), which is a commonly used plasticizer incorporated in electrical cables, wall covering, flooring, and others. The alkaline hydrolysis of DEHP is likely a major source of 2-ethylhexanol in indoor air (Azuma et al., 2016; Nalli et al., 2006; Reiser et al., 2002). Concentrations up to 130 $\mu g/m^3$ of 2-ethylhexanol were recognized, for example, in Japanese houses (Azuma et al., 2016).

The contamination of indoor air by VOC and other pollutants is (among other factors) related to different disease symptoms of people who spend a lot of time in buildings, mainly those with mechanical heating, ventilation, and air-conditioning systems. These symptoms include headache, lethargy, dry skin, and mucous membrane symptoms related to the eyes, nose, and throat, and are summarized as sick building syndrome (SBS) or building-related illness (Burge, 2004). Studies have shown that more than 30% of office workers in Germany suffer from SBS (Bischof et al., 2004; Brasche et al., 1999). Although VOC's do not necessarily cause obvious symptoms, a chronic exposure can lead to reduced concentration and effectivity, or other health problems like asthma and cardiovascular diseases (Brasche et al., 1999; Burge, 2004; Jones, 1999; Mendell et al., 2002; Redlich et al., 1997). Further, an exposure to low concentrations of VOC's was associated with an increased risk of cancer (Vaughan et al., 1986; Wallace, 1991; Wolkoff and Nielsen, 2001).

To remove sources of pollution or to increase the ventilation rate is technically and economically difficult to achieve. The easiest way to clean the indoor air is natural ventilation. However, this is often not possible or desired, due to reasons like unfavorable weather conditions, outdoor pollution, safety standards, climate regulation, or noise pollution. Further, due to energy-saving measures, buildings have become more and more airtight, often with a minimum of air exchange, which leads to a higher concentration of pollutants indoors than outdoors (Jones, 1999; Wolkoff and Nielsen, 2001). Thus, several treatment technologies have been developed for improving IAQ. Among them are technologies based on mechanical and electrical filtration, photolysis, adsorption, and ozonation. Even though these technologies work well for particle filtration, there is no fully-satisfying method for VOC removal (Guieysse et al., 2008; Luengas et al., 2015; Vizhemehr et al., 2015). The results of several investigations demonstrated that the filter capacity of different air cleaners is partially insufficient (especially for low-molecular weight compounds like formaldehyde and

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dichloromethane) or the devices release pollutants themselves, e.g. ozone, formaldehyde, and acetaldehyde (Chen et al., 2005; Hodgson et al., 2007; Luengas et al., 2015; Tseng et al., 2005; Vizhemehr et al., 2015; Yu et al., 2011).

Beside the well-established technologies for improvement of IAQ, new developments have been introduced in recent years. Among them, innovations in air cleaning, leveraging on smart technologies and sensing systems, and regulations for the evaluation of emissions from building products (Daeumling, 2016; Tham, 2016). These recently developed strategies together with responses to climate change and energy conservation have led to a change in the indoor chemical environment. For example, heavy metal toxicants (e.g. cadmium and mercury) and carcinogens (e.g. formaldehyde and benzene) could be reduced, however, reproductive toxicants (e.g. phthalates and polychlorinated biphenyls (PCBs)) and endocrine disruptors (e.g. brominated flame-retardants and bisphenol-A) have increased (Rudel and Perovich, 2009; Weschler, 2009). The deterioration of IAQ could not be alleviated effectively and therefore, the interest in IAQ is still growing at a public, a political, and a scientific level. Considerations for its enhancement to mitigate health complains, improve quality of life and the work environment with consequential benefits to well-being and performance are mandatory (Knöppel and Wolkoff, 2013; Tham, 2016).

However, beside IAQ, different psychological variables are also important in regard to human health, especially in workplace environments (O´Leary, 1990; Ryan and Morrow, 1992). Environmental dimensions in building design, like stimulation (e.g. odor, color, and visual exposure), coherence (e.g. landmark and floorplan complexity), affordance, control (e.g. boundaries and privacy), and restoration (e.g. minimal distraction and fascination), have the potential to affect human health (Evans and McCoy, 1998). The quality of the physical environment, e.g. thermal conditions, visual and acoustic design, and the interior design are strongly related to environmental satisfaction (Hongisto et al., 2016). Thus, in addition to improved air cleaning strategies, a comfortable and satisfying indoor environment should be created to improve occupants' environmental satisfaction (as well as job satisfaction), and accompanying health.

Plants are long-standing known to assimilate and metabolize toxic compounds from the air, soil or water. Acting as an important global sink for environmental chemicals, they can be considered as "green liver" (Paterson et al., 1990; Sandermann, 1992; Schulte-Hostede et al., 1987; Terry and Banuelos, 2000). Based on this fact, research has been directed toward the capability of plants to filter toxic compounds out of indoor air. In recent decades, a large

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number of chamber experiments were conducted to evaluate the capability of ornamental plants to filter indoor pollutants, mainly VOC. First reports were published by Wolverton and co-workers as part of a NASA study (Wolverton and Wolverton, 1993 a; Wolverton et al., 1984). This was followed by a series of similar experiments, performed by other researchers worldwide. Also several field studies were conducted to examine the effects of ornamental plants on IAQ.

However, in most experiments it is not clear, whether the plant and/or the substrate with the containing microorganisms adsorbed VOC (or other air pollutants). Further, the environmental conditions relevant for plant growth and for the plant physiological status, such as humidity and CO_2 content, were often not controlled leading to non-reproducible results. Generally, less is known about the influence of plant physiology on air pollutant removal as well as about phytotoxic effects of such pollutants, which in return would affect the plant's pollutant removal efficiency. Moreover, due to a substantial heterogeneity in experimental setups (e.g. methods and test designs), the results on VOC removal by plants are quite mixed. This applies to field studies, where setup and parameters are typically less controllable, as well as to chamber experiments. Thus, there remains a need for an efficient and uniform method to determine the filter capability of pollutants by ornamental plants using test chambers.

Interestingly, despite the fact that VOC concentration in field studies was often unaffected (or even higher) in the presence of plants (Dingle et al., 2000; Kim et al., 2013, 2011; Smith and Pitt, 2011), many field studies could prove a positive effect of plants on human health. This could on one hand imply a placebo effect. On the other hand, plants might be capable to promote human well-being independently of a potential VOC removal. For instance, plants can improve human health by creating an environment that is perceived more comfortable, fascinating, and favorable (Evensen et al., 2013; Larsen et al., 1998). Several studies have shown that plants in the workplace environment can reduce health complains (Fjeld et al., 1998), or increase the level of mood, (Larsen et al., 1998), workplace satisfaction (Nieuwenhuis et al., 2014), and well-being (Lohr and Pearson-Mims, 2000).

In summery, many issues regarding a potential VOC removal by plants are still unresolved or emerged by the highly variable published studies. Therefore, there is a need for a test chamber design that is less prone to errors and ensures reproducible results in regard to the VOC removal efficiency of plants. Furthermore, knowledge concerning the correlation between VOC exposure and plant physiology is required to understand which parameters can affect the

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air pollutant removal efficiency of plants. Finally, attaining information about the overall effect of plants on human well-being can further help to give appropriate recommendations for indoor greening and thus to improve human health and well-being. In order to obtain in-depth knowledge according these issues, the current doctoral thesis was divided in the following three parts:

Chapter I	**Chapter II**	**Chapter III**
Laboratory studies *- Examination of an experimental chamber to test the VOC removal efficiency of plants* *- Influencing factors on results of chamber experiments* Hörmann et al. (2017): Suitability of test chambers for analyzing air pollutant removal by plants and assessing potential indoor air purification. Water, Air, & Soil Pollution 228: 402	Laboratory studies *- Evaluation of the pollutant removal efficiency of selected plants* *- Putative interrelation of VOC exposure and plant physiology* Hörmann et al. (2017): Assessment of filtration efficiency and physiological responses of selected plant species to indoor air pollutants (toluene and 2-ethylhexanol) under chamber conditions. Environmental Science and Pollution Research	Field study in offices *- Examination of the VOC removal efficiency of Spathiphyllum wallisii in a real-life setting* *- Analysis of people-plant relationship in regard to a possible placebo effect* Hörmann et al. (submitted): Human well-being through plants - a placebo effect? Environmental Psychology

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Chapter I - Chamber experiments to determine the pollutant removal capability of indoor plants

Experiments concerning the VOC removal capability of plants are usually examined in chamber experiments. To obtain repeatable results, test chambers should ensure an accurate VOC measurement and the control over environmental conditions that are important for plant physiology. However, such chambers are not commercially available, and the results of different investigations vary greatly and are not comparable. Therefore, the first chapter covers the following issues:

- Are so called Bioboxes (special test chambers that were developed for the examination of plant physiological responses to their environment under controlled conditions) suitable for determining the VOC uptake by plants?
- What are the major pitfalls in the experimental set up?
- What causes the high heterogeneity of results regarding VOC uptake reported in literature and which parameter have an impact on results?
- Which precautions should be taken to extrapolate the results from chamber studies to real environment?
- Finally, recommendations for chamber experiments concerning the VOC removal by plants are given.

Hörmann, V.; Brenske, K.-R. & Ulrichs, C. (2017): Suitability of test chambers for analyzing air pollutant removal by plants and assessing potential indoor air purification. Water, Air, & Soil Pollution 288:402

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Suitability of Test Chambers for Analyzing Air Pollutant Removal by Plants and Assessing Potential Indoor Air Purification

Vanessa Hörmann · Klaus-Reinhard Brenske · Christian Ulrichs

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Abstract A unique test chamber system, which enables experiments with plants under highly controlled environmental conditions, was used to examine the pollutant removal efficiency of plants. For this purpose, the removal of two different volatile organic compounds (VOC) (toluene, 2-ethylhexanol) from the air by aerial plant parts of two common indoor plant species (*Dieffenbachia maculata* and *Spathiphyllum wallisii*) was monitored. While the control over environmental conditions (temperature, relative humidity, CO_2 content, and light condition) worked very well in all experiments, control experiments with the empty chamber revealed high losses of VOC, especially 2-ethylhexanol, over the test duration of 48 h. Nonetheless, compared to the empty chamber, a significantly stronger and more rapid decline in the toluene as well as in the 2-ethylhexanol concentrations was observed when plants were present in the chamber. Interestingly, almost the same VOC removal as by aerial plant parts could be achieved by potting soil without plants. A comparative literature survey revealed substantial heterogeneity in previous results concerning the VOC removal efficiency of plants. This can be mainly attributed to a high diversity in experimental setup. The experimental setup used in the current study offers an excellent opportunity to examine also plant physiological responses to pollutant exposure (or other stressors) under highly controlled conditions. For the analysis of VOC removal under typical indoor conditions, to obtain data for the assessment of realistic VOC removal efficiencies by plants in rooms and offices, a guideline would be helpful to achieve more coherent findings in this field of research.

Keywords Ornamental plants · Indoor air quality · Air purification · Volatile organic compounds (VOC) · Toluene · 2-Ethylhexanol

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1 Introduction

People today spend 80–90% of their time indoors; thus, awareness toward indoor air quality has become increasingly important (Sarigiannis et al. 2011; Schweizer et al. 2006). Of certain interest are pollutants like volatile organic compounds (VOC) that might occur at high concentrations indoors (Jones 1999; Yu and Kim 2010). VOC are emitted by technical equipment as well as by furniture and carpets (Berrios et al. 2005; Que et al. 2013). Another source of VOC is related to human activities (Rösch et al. 2014) and indoor gas phase reactions which lead to the formation of secondary VOC (Salthammer and Bahadir 2009). Benzene, toluene, xylenes, styrene, and the group of terpenes are the

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most important VOC (Sarigiannis et al. 2011). Another compound of rising interest is 2-ethylhexanol which is formed by hydrolyses of di-2-ethylhexyl phthalate (DEHP), a major plasticizer incorporated in electrical cables, wall covering, flooring, and others (Azuma et al. 2016; Nalli et al. 2006; Reiser et al. 2002).

The contamination of indoor air by VOC (among other factors) is linked to disease symptoms including headache, lethargy, dry skin, and mucous membrane symptoms related to the eyes, nose, and throat. These symptoms are summarized as "sick building syndrome" (Burge 2004). Studies have shown that more than 30% of office workers in Germany suffer from the sick building syndrome (Bischof and Bullinger 1998; Brasche et al. 1999). Other authors associate a long-term exposure to low concentrations of VOC with an increased risk of cancer (Vaughan et al. 1986; Wallace 1991; Wolkoff and Nielsen 2001).

Several actions for abatement of indoor pollutants have been introduced. Among them are innovations in air cleaning, leveraging on smart technologies and sensing systems, and regulations for the evaluation of emissions from building products (Daeumling 2016; Tham 2016). These actions have led to a change in the indoor chemical environment. Even with air purification technology, heavy metal toxicants and carcinogens could be reduced; however, reproductive toxicants and endocrine disruptors have increased (Rudel and Perovich 2009; Weschler 2009). While technologies for particle filtration work well, there is no fully satisfying method for VOC removal (Chen et al. 2005; Guieysse et al. 2008; Luengas et al. 2015; Vizhemehr et al. 2015).

Plants are known to assimilate and metabolize toxic compounds from the air, soil, or water. Several enzymatic driven actions, like functionalization, transformation, and compartmentation, are included in the detoxification process. Xenobiotics may than be stored in vacuoles (soluble compounds) or cell walls (insoluble compounds) or further metabolized up to a deep oxidation. Acting as an important global sink for environmental chemicals, plants can be considered as "green liver" (Kvesitadze et al. 2009; Paterson et al. 1990; Sandermann 1992; Schulte-Hostede et al. 1987; Terry and Banuelos 2000). Based on this fact, research has been directed toward the capability of plants to filter toxic compounds out of indoor air. In recent decades, a large number of chamber experiments were conducted to evaluate the capability of ornamental plants to filter indoor pollutants, mainly VOC. First reports were published by authors taking part in a NASA study (Wolverton and Wolverton 1993; Wolverton et al. 1984). This was followed by a series of similar experiments, investigated by other researchers worldwide. However, if and to what extent plants really have an impact on indoor air quality is still under discussion in the scientific community (Dela Cruz et al. 2014; Llewellyn and Dixon 2011; Schmitz et al. 2000). The reports of different investigations show a broad variability in results. This gives rise to the questions of what causes this variability and what can we learn from these results considering the air purification capability of plants indoors.

Therefore, the aim of this study was to identify major challenges that have to be overcome in test chamber experiments considering the air purification capability of plants. Beside the impact of the test chamber design, the influence of different treatments and environmental conditions were taken into account. In order to identify major difficulties that may occur, we conducted a set of selected chamber experiments. The suitability of the test chambers and which conclusions can be drawn are discussed. The feature of our test chamber, in comparison to many other studies, was the possibility to control and record environmental conditions (including relative humidity and CO_2 content). The plant species *Dieffenbachia maculata* and *Spathiphyllum wallisii* were chosen because they are commonly used in interiorscapes, are widely distributed, and were already tested against their filter capability in several other studies. Toluene is a VOC which is frequently detected indoors (Sarigiannis et al. 2011) and has also been tested by other authors in similar experiments (Wood et al. 2006; Yang et al. 2009; Yoo et al. 2006). The alcohol 2-ethylhexanol is a (potential) indoor pollutant of rising interest (Azuma et al. 2016; Nalli et al. 2006; Reiser et al. 2002) and was not yet tested against removal by plants. Both VOC are chemically different, e.g., a high vapor pressure (29.1 hPa at 20 °C), no polar functional groups, and an aromatic structure for toluene versus low vapor pressure (0.48 hPa at 20 °C), a polar hydroxyl group, and an aliphatic acyclic structure for 2-ethylhexanol. The behavior and fate of these VOC with their different chemical properties was examined in our test chamber with and without plants to get an impression of the chamber's suitability regarding air purification experiments. For this, high VOC concentrations were used that are not representative of usual indoor air pollution levels. Furthermore, a literature survey was

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conducted to demonstrate the high heterogeneity of results of chamber experiments on VOC removal by plants, examining different aspects like light conditions or VOC concentration. It is discussed to what extent the experimental setup, including test chamber design, treatments, and environmental conditions, may affect results. Further, suggestions for general guidelines for VOC removal experiments are given.

2 Material and Methods

2.1 Test Chambers

Two individual gas-tight chambers measuring 80 × 60 × 50 cm (height, width, depth) were purchased from the company GMS (Gaswechsel-Messsysteme GmbH, Berlin, Germany). Each of these so-called Bioboxes (Fig. 1) with a total volume of 240 L consists of two parts: a metal base (15 × 60 × 50 cm) and a Plexiglas hood (65 × 60 × 50 cm) on top. The base contains sensors for temperature, relative humidity, and CO_2 and a heat exchanger. The base of each Biobox is further equipped with stainless steel fittings to allow injection of VOC and to take air samples. In addition, fans were installed in the base to provide a complete mixing of the VOC-loaded air within the chamber and to overcome the boundary layer resistance of the leaves. The hood that has a volume of 195 L is connected to the base via a gas-tight Neoprene gasket. It has an opening at the front that allows the loading of plants into the system. This opening can be closed by attaching a panel, also with a gas-tight Neoprene gasket. A metal panel (50 × 50 cm) separates the base from the hood. Plants (or pots with soil) were placed on top of the metal panel. Left and right of the panel, a gap facilitates the air movement between hood and base.

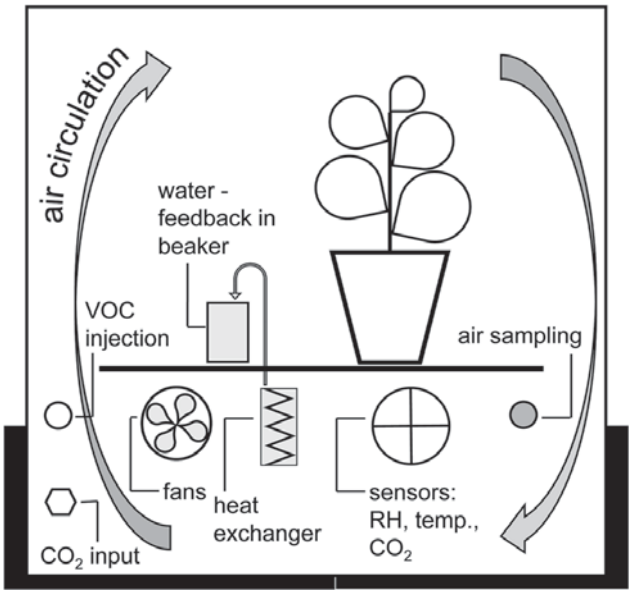

Fig. 1 Schematic illustration of the Bioboxes used in this study. RH, relative humidity; temp., temperature

Each Biobox is placed in a plant growth chamber (Adaptis A 1000, Co. Conviron, Winnipeg, Canada) which controls light intensity, light duration, and temperature. The Biobox sensors pass their signals on to a computer which controls the air humidity and the CO_2 concentration. During the experiments, the air humidity was kept constant by lowering the surface temperature of the heat exchanger so that surplus water condensed. The condensed water was fed via a tube to a beaker inside the Biobox. The CO_2 content was measured continuously by infrared spectroscopy. If the concentration dropped below the set point, CO_2 was automatically reinjected into the system via a tube from an external CO_2 gas cylinder.

Both Bioboxes were used in parallel for all experiments/treatments, including the empty chamber controls.

2.2 VOC Injection and Air Sampling

VOC used in this study were toluene (≥ 99.5% toluene for synthesis, Co. Carl Roth GmbH & Co. KG, Karlsruhe, Germany), of which 5.5 μL were injected (≈ 20 mg m^{-3}; 5.3 ppm) and 2-ethylhexanol (99% 2-ethyl-1-hexanol, Co. Alfa Aesar, Karlsruhe, Germany), of which 4.2 μL were injected (≈ 14.6 mg m^{-3}; 2.9 ppm). A 10-μL syringe with built-in Chaney Adapter (Model 701 NCH SYR, Cemented NDL, 26s ga, Co. Hamilton, Bonaduz, Switzerland) was used for all VOC injections through the left-side port in the Biobox base. This equipment ensured that the defined volume of a certain VOC could be injected very precisely in every replicate.

The right-side fitting in the base of the Biobox was used to take the air samples. Therefore, a sorption tube (C1-CXXX-5003, Tenax TA. C6-C30. Inert-coated., Co. Markes, Frankfurt, Germany) spiked with internal standards (100 ng of each, cyclooctane and cyclododecane) was connected to a gas-tight syringe (Borosilicate glass barrel 3.3, 100 mL, Co. Lehrmittel- und Verlagsgesellschaft mbH, Hofheim-Diedenbergen, Germany) and inserted into the Biobox via the fitting. A defined volume of air (50 mL for experiments with

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toluene and 100 mL for experiments with 2-ethylhexanol) was drawn through the tube with a flow rate of 100 mL/min. Until analysis, the sorption tubes were stored in gas-tight aluminum bags at room temperature.

2.3 Test Plants and Culture

Well-developed plants of *Dieffenbachia maculata* "Compacta" (Lodd. et al.) G. Don and *Spathiphyllum wallisii* "Daniel" (Regel) (pots with 12 cm diameter, see Fig. 2) were purchased from a wholesale market. Prior to the experiments, the plants were cultivated in a growth chamber (Adaptis A 1000, Co. Conviron) for at least 3 days and up to 3 weeks with 15 h light per day (100 ± 10 μmol m^{-2} s^{-1}; MQ-200 Quantum Separate Sensor, Apogee Instruments, Inc., USA), temperature of 22/20 °C (day/night), and a constant relative humidity of 60–70%. Plants were watered regularly with tap water as needed.

2.4 Experimental Protocol

Three plants of either *D. maculata* or *S. wallisii* or three pots without plants filled with unused potting soil only (potting soil + clay + FE, Co Gramoflor, Vechta, Germany) were tested per Biobox (Fig. 2) under continuous light. All experiments were replicated four times ($n = 4$) using another set of plants or potting soil. For the experiments with potting soil only, the pots were wrapped in aluminum foil before filling with soil, to avoid sorption of VOC on plastic pots. In experiments with plants, the pots and the substrate surface were entirely covered with aluminum foil before placing the plants into the Biobox to ensure that removal of VOC can be attributed to aerial plant parts and soil effects are negligible within the experimental time span. Prior to VOC exposure, the plants were watered to saturation and allowed to drain for 1 h. During that time plants were placed in the Biobox under experimental conditions to get acclimatized. The potting soil was supplied with 100 mL water directly prior VOC exposure.

The climate conditions were set as follows: CO_2 = 500 ppm, RH = 70%, temperature = 22 °C, light = 180 ± 10 μmol m^{-2} s^{-1} (all parameters related within the Biobox). The duration of the experiment is an important factor. In published studies, the exposure time in chamber experiments varied between 2 h (Liu et al. 2007) and several days (Treesubsuntorn and Thiravetyan 2012). It is described that plants may need some time for the induction of VOC removal (at least 24 h) (Orwell et al. 2006; Wood et al. 2002) and that the VOC removal may vary diurnally (Liu et al. 2007). Thus, we decided to run our chamber experiments on the VOC removal by aerial plant parts (and by potting soil) for 48 h. Over this time, plants would have enough time for the induction of VOC removal and the natural VOC decline would have decreased due to saturation of surfaces while the plant would continue to remove VOC. At the beginning of each experiment, toluene or 2-ethylhexanol was injected and allowed to evaporate and equilibrate in the chamber. Thus, the first air samples were taken after 6 min for toluene and after 1 h for 2-ethylhexanol. The following air samples were taken after 5, 24, 29, and 48 h for both VOC. Tests on adsorption in empty chambers were conducted to assess potential effects by sorption on chamber surfaces.

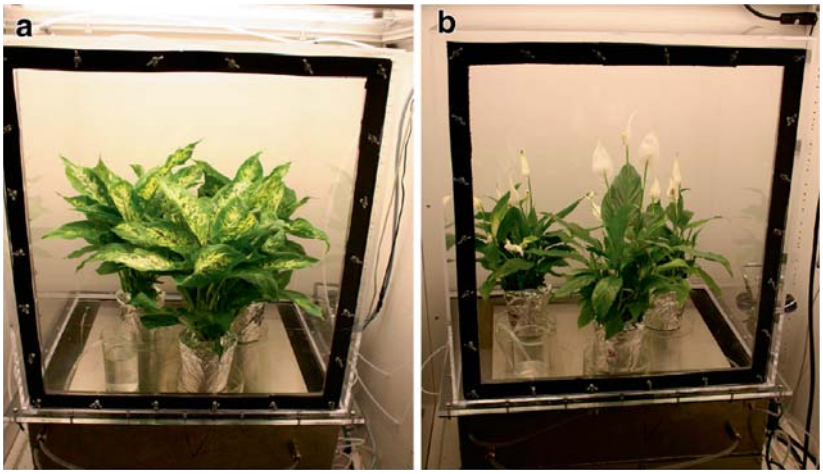

Fig. 2 Biobox equipped with *Dieffenbachia maculata* (**a**) and *Spathiphyllum wallisii* (**b**)

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2.5 Trace Gas Analysis

Trace gas analysis was conducted using an Agilent 6890 gas chromatograph (Agilent Technologies, Inc., Santa Clara, USA) equipped with a Rtx-5MS phase column (60 m, 0.32 mm, 0.5 μm, Restek GmbH, Bad Homburg v. d. Höhe, Germany) and a flame ionization detector and coupled to a mass spectrometer (5979 MSD, Agilent Technologies Inc.). Helium was used as carrier gas. Prior to gas chromatographic analysis, the sorption tubes were desorbed using a Turbomatrix thermal desorber (Perkin Elmer, Waltham, USA). Analyses were performed in accordance with DIN ISO 16000-6 with a detection limit of 1 μg m^{-3}.

2.6 Statistical Analysis

Data were statistically analyzed by one-way ANOVA and post hoc Tukey's HSD test with $p \leq 0.05$ using the statistical software package SPSS 23 (IBM).

3 Results and Discussion

3.1 Operation of the Biobox and Findings on VOC Removal by Aerial Plant Parts and Potting Soil

The control over environmental conditions, namely relative humidity, temperature, and CO_2 content (Fig. 3) as well as light intensity worked well in the Biobox equipped with plants. The relative humidity and temperature varied only slightly around the set points (22 °C; 70%). The CO_2 content showed somewhat higher fluctuations (set point: 500 ppm), but photosynthesis should not be affected too much by this. The light intensity was measured in terms of photosynthetically active radiation (PAR), thus in a light spectrum of 400–700 nm (not shown). The light intensity used (180 ± 10 μmol m^{-2} s^{-1} ≈ ca. 13 klx) exceeds common conditions in, e.g., offices (around 0.5 klx) by far, as well as the relative humidity. These settings were used to create optimal conditions for plants regarding stomatal uptake, thus to achieve maximal removal efficiencies. Furthermore, the Biobox was loaded with a relatively high amount of plants, or rather, leaf surface (see Fig. 2), to be able to detect the limited VOC uptake.

Due to the high sorptive losses of VOC on the surfaces of the Biobox (see Fig. 4), it was necessary to use an excessive amount of VOC (toluene, 5.5 μL representing 20 mg mg^{-3}; 2-ethylhexanol, 4.2 μL representing 14.6 mg m^{-3}). The measured concentration of toluene (which has a high volatility) from the first air samples taken after 6 min, corresponded satisfyingly with the injected amount (Fig. 4a). In contrast, the injected amount of 2-ethylhexanol could not be recovered. Due to its chemical properties, 2-ethylhexanol needs much longer to enter the gas phase than toluene. Hence, the first air samples were taken after 1 h. Nonetheless, the highest amount that could be detected for 2-ethylhexanol was 5.5 mg m^{-3} (measured in the 1 h samples, see Fig. 4b). Apparently, parallel to entering the gas phase, part of the 2-ethyhexanol got adsorbed by the surfaces of the Biobox and/or the aerial plant parts or the potting soil. In general, it can be recommended to applicate volatile compounds in its gas phase, this especially includes compounds with a low volatility.

After 48 h, in the empty Biobox, approximately 60% of the initial toluene concentration remained, but only around 25% of the initially measured 2-ethylhexanol concentration (Fig. 4). Despite these VOC losses in an empty Biobox, significant differences between chambers with and without plants were detected for toluene

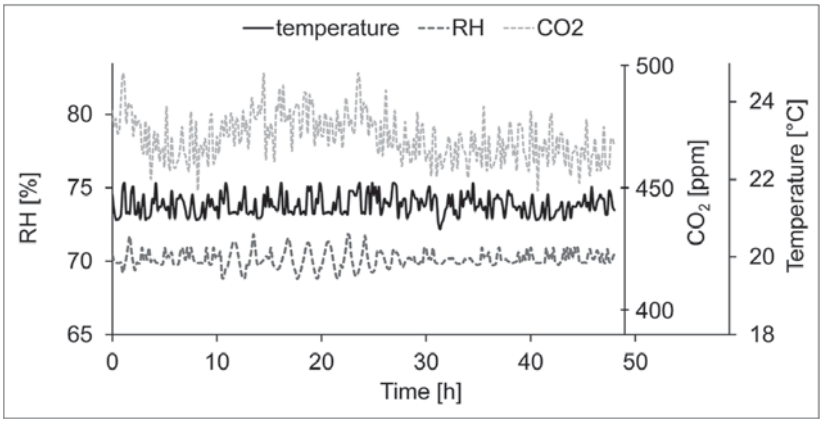

Fig. 3 Section of temperature, relative humidity (RH), and CO_2 measurements during an experimental run in the Biobox containing plants

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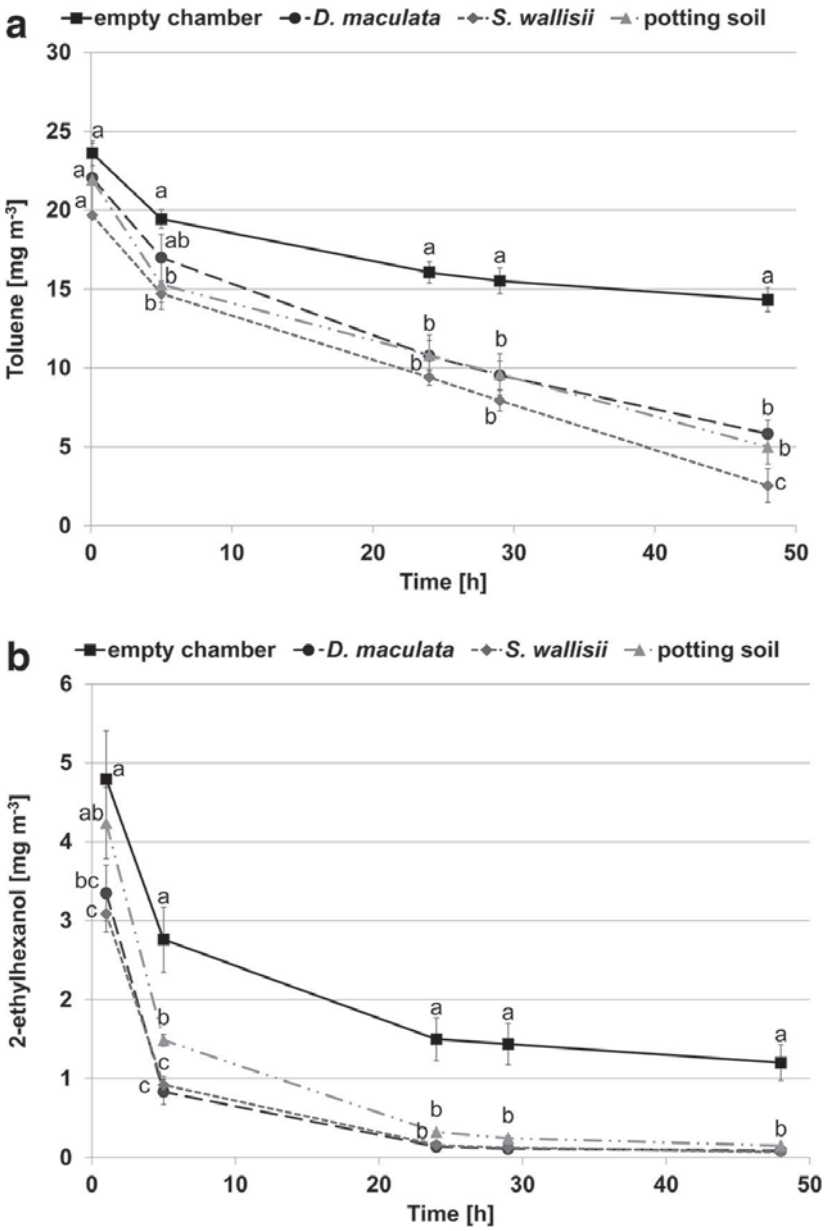

Fig. 4 Toluene (**a**) and 2-ethylhexanol (**b**) concentrations in the Biobox with different plant species under light conditions over time. Data are presented as means ± SD of four experiment replicates ($n = 4$). Different letters indicate significant differences within each sample point ($p \leq 0.05$, one-way ANOVA, followed by Tukey's HSD test)

and 2-ethylhexanol at least after 5 h. This indicates a rapid VOC adsorption not only by the chamber surfaces but also by the aerial plant parts/potting soil. After 48 h, the VOC levels in the chamber experiments with plants were only around 30% of the toluene and around 10% of the 2-ethylhexanol concentration in empty chambers. However, since major removal effects seem to take place shortly after introduction of VOC, air samples should be taken more frequently within the first hour(s), to gain more detailed information on sorption patterns/removal curves. While this issue can be easily adjusted, the high losses of VOC in the empty Biobox are a much greater challenge. The adsorption effects regarding especially 2-ethylhexanol on surfaces of the Biobox properly masked effects by the plants. Therefore, conclusions concerning the removal of 2-ethylhexanol by plants should be considered carefully. The design of the Biobox allows the use of hoods made of different materials. Because we assumed a lower VOC adsorption on glass than on Plexiglas, we tested also a glass hood. However, due to construction faults, the glass hood showed major deficiencies in regard to VOC adsorption and also VOC emission and was not further considered. More information about this issue is provided in the Online Resource.

Significant differences between the two tested plant species were not found with the current set up, except for the last time point in the toluene experiments. Here,

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S. wallisii showed a slightly higher VOC removal than *D. maculata*. Surprisingly, the VOC removal curves of experiments with potting soil only were not different from experiments with aerial plant parts, except for early time points in the 2-ethylhexanol experiments. Here, the VOC removal by potting soil was lower than that achieved by the aerial plant parts (Fig. 4b). Experiments by Irga et al. (2013) have shown that unused potting mix removes also substantial amounts of benzene. The removal was slower than the whole potted plants system; however, here the aerial plant parts were not separated from the potting mix and finally, the same amount of benzene was removed by both treatments at the end of the experiments. Several other studies have also shown that potting substrates remove VOC; however, microorganisms are supposed to be the major contributors (cf. Dela Cruz et al. 2014).

The rapid decrease of VOC within the first 5 h was likely due to sorptive effects on chamber surfaces and on the plant cuticle or the potting soil. After the surface was largely saturated, the decrease of VOC slowed down and the following VOC removal, that was particularly obvious for toluene (Fig. 4a), might be also attributed to stomatal uptake by aerial plant parts (see Fig. 5). In case of 2-ethylhexanol, the VOC concentration was already very low after the surface saturation. Thus, the phase in which stomatal uptake would be the predominant cause for VOC removal is here less obvious.

The VOC removal (meaning the negative slope of graphs in Fig. 4) for chambers with plants or potting soil seems to be sigmoid to exponential. An exponential VOC removal is also described by Hanoune et al. (2013) and Oyabu et al. (2003). A linear concentration-dependent function is also conceivable as outlined by Wetzel and Doucette (2015) and Wood et al. (2002). However, the rate of VOC removal is dependent on the considered time frame and also on the ratio of chamber volume to surfaces, including the plant as well as chamber walls and installed components. The higher the ratio of chamber volume to surfaces, the more flattened is the graph and the more it approaches to a linear function in a certain time frame (Fig. 5). Depending on the considered time frame, the graph appears sigmoid, exponential or linear. It is necessary to repeat experiments with changing treatments (e.g., plant species, VOC identity, and concentration) and frequent air samples to investigate the particular mechanisms of VOC removal by plants in more detail.

3.2 Influence of Environmental Factors and Plant Species on VOC Removal Efficiency

The removal of VOC by potted plants can be divided in the removal by aerial plant parts (through the cuticle and through the stomata) and the removal by growing media, microorganisms, and roots within. The removal efficiency is influenced by many factors, e.g., plant species, temperature, light intensity, growing media, and VOC (identity, concentration, potential mixture effects) (Dela Cruz et al. 2014). Some of these factors are further discussed in the following.

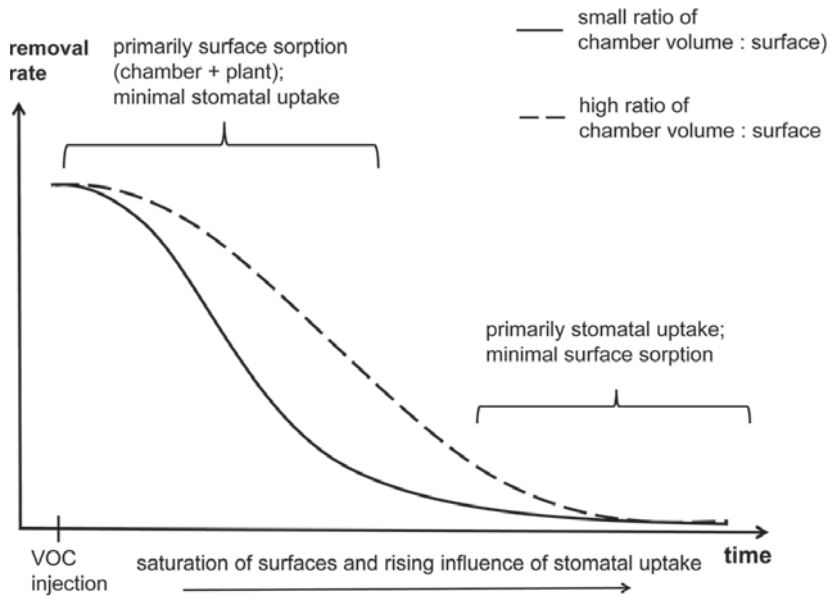

Fig. 5 Simplified scheme of VOC removal by aerial plant parts in chamber experiments in the course of time. VOC removal is indicated as removal rate, representing the amount of VOC removed per unit time per leaf area

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Light condition can affect the VOC removal by plants. Kim et al. (2008) demonstrated that the removal efficiency of aerial plant parts of *Fatsia japonica* Decne. & Planch. and *Ficus benjamina* L. for formaldehyde is higher under light than under dark conditions. Investigations by Xu et al. (2011) revealed a higher uptake of formaldehyde during daytime versus nighttime for aerial plant parts of *Chlorophytum comosum*, *Aloe vera*, and *Epipremnum aureum* as well. In entire-plant experiments (entire plant = aerial parts and growing media including microorganisms and roots), a higher VOC uptake under light conditions was also described by Treesubsuntorn & Thiravetyan (2012) for the removal of benzene by *Dracaena sanderiana.* In contrast, investigations on the removal efficiency of entire plants of different species (*Hedera helix, Chrysanthemum morifolium*, *Dieffenbachia compacta*, and *E. aureum*) by Aydogan & Montoya (2011) showed that the uptake of formaldehyde is higher under dark, rather than at light conditions. In turn, Yoo et al. (2006) have demonstrated no major differences concerning the VOC uptake of aerial plant parts depending on light versus dark conditions. Moreover, light intensity can have an impact. Porter (1994) detected a generally less efficient removal of toluene by entire plants of *Dieffenbachia amoena* under low light intensity (35 μmol m^{-2} s^{-1}) compared to higher light intensity (90 μmol m^{-2} s^{-1}).

Plant surfaces seem to play a more important role at the beginning of VOC exposure due to sorption effects of VOC on the cuticle, while stomata opening—highly dependent on light conditions—plays a greater role when plant surfaces are saturated (Fig. 5). How long the cuticular removal lasts predominantly depends on plant characteristics (e.g., cuticle thickness and lipid composition), which can differ, e.g., between different plant species and between different developmental stages, and VOC identity (functional groups, lipophilicity, etc.). Potential differences for the removal efficiency in light versus dark conditions, and thus an indication of the metabolic pathway of VOC in plants, may be discovered with an extended experimental time span in combination with a continuously emitting VOC source.

Other influencing factors are the plant species and the identity, concentration, and mixture effects of VOC. However, also in this regard, the results reported in literature differ. Investigations by Cornejo et al. (1999) examined the ability of several plant species (*Pelargonium x. domesticum* Bailey, *Saxifraga stolonifera* Mere, *Tradescantia fluminensis*, *Dracena deremensis*, *Kalanchoe blossfeldiana* Poelln., *Primula sinesis*, *Magnesia* sp., and *C. comosum*) to remove benzene, trichloroethylene, and toluene singly or in mixture. The authors found that the VOC removal efficiency varied depending on plant species and VOC identity. Yang et al. (2009) examined the removal efficiency of 28 ornamental plant species for benzene, toluene, octane, trichloroethylene, and α-pinene. In this experiment, aerial plant parts showed also different removal efficiencies depending on both chemical and biological identity. In another investigation, no difference in the VOC removal efficiency between substances was detected when aerial plant parts of *Zamioculcas zamiifolia* were exposed to benzene, toluene, ethylbenzene, or xylene (Sriprapat and Thiravetyan 2013). Trials of Yoo et al. (2006) showed that aerial plant parts of *S. wallisii* Regal have a higher removal efficiency for benzene than foliage of *Syngonium podophyllum* Schott., followed by *H. helix* L. In contrast, all three plant species exhibited the same removal efficiency for toluene.

Depending on VOC concentration, differences in the removal efficiency are likely, at least in the beginning of exposure (at primary sorption-based uptake). Orwell et al. (2006) tested *Spathiphyllum* sp. and *D. deremensis* (entire plants) for their ability to remove toluene and m-xylene at different concentrations (0.758 to 379 mg m^{-3}). It was found that the VOC removal increases with increasing VOC concentration. Other trials showed the same effect, if entire plants (*C. comosum, A. vera* and *E. aureum*) were exposed to different concentrations of formaldehyde (Xu et al. 2011). However, investigations of Porter (1994), who exposed *Dieffenbachia amoena* (entire plant) to increasing concentrations of toluene, showed that up to 200 mg m^{-3} toluene an increasing VOC removal efficiency was achieved only under low light intensity (35 μmol m^{-2} s^{-1}). Above that concentration (tested up to 1200 mg m^{-3}), no further increase in the toluene removal efficiency was observed. At higher light intensity (90 μmol m^{-2} s^{-1}), that in general resulted in a more efficient VOC removal, no clear correlation could be found between VOC concentration and VOC removal efficiency in that study.

Similarly heterogeneous are the results of the removal efficiency in dependence of VOC mixtures. For instance, Yoo et al. (2006) reported that the removal efficiency for toluene is higher than that of benzene when plants were exposed to both VOC concurrently. Studies of Cornejo et al. (1999) showed that benzene is

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taken up selectively over toluene when supplied in a mixture. Results by Porter (1994) in turn revealed that benzene and toluene were removed with the same efficiency when applied in a mixture.

As different plant cultivars (most likely also of different developmental stage or fitness) and/or different VOC were used in all these studies, it is obvious that observed effects are not generalizable. It seems that plants respond specifically to individual VOC as well as to VOC mixtures, depending on the species of the plant (and probably its development and fitness), the VOC concentration, and environmental factors (e.g., light conditions, temperature, and relative humidity). Another factor of uncertainty is the growing media including microorganisms and roots. The exposure or coverage of the growing media may have a strong impact on results, since already unused growing media removed similar quantities of toluene and 2-ethylhexanol compared to the aerial plant parts of *D. maculata* and *S. wallisii* (Fig. 4). However, the removal capacity of the growing media itself is also most likely influenced by several factors like type/composition, its hygrometry, and the kind and abundance of microorganisms present in the media. This makes an entire plant system quite complex and without additional investigations, it remains unclear which portion of VOC removal can be attributed to soil, microorganisms, or the plant itself. Also the impact of plant roots on VOC removal is so far largely unknown (Dela Cruz et al. 2014). It seems recommendable to examine the particular contributors and the respective fate of VOC within the potted plant system by using radio labeled compounds. A limited number of experiments were already conducted (Giese et al. 1994; Jen et al. 1995; Ugrekhelidze et al. 1997); however, only plant parts or plant compartments were investigated but not the fate in the growing media. More research is needed to identify the major contributors of VOC removal and the degradation pathways in dependence of several factors, e.g., plant species, type of growing media, microbial community, and VOC identity.

3.3 Can Results Be Transferred from Chamber Experiments to Rooms?

The ambient conditions in chamber experiments often vary appreciably with conditions found in interiors. Usually, the relative humidity and especially the light intensity, as well as the VOC concentration are much lower indoors. In addition, so far in chamber experiments, just one or two VOC at most are tested at the same time, although indoors there is always a mix of many different VOC. In most chamber experiments, just an initial dose of VOC was injected, while indoor environments commonly have continuously emitting VOC. The air is continuously mixed by fans or the like, whereas a plant in an interior setting does not get in touch with all the air present. Besides, boundary layer effects, which have a significant role in gas exchange, are in chamber experiments usually prevented by an increased air movement. However, chamber experiments are an important tool to receive insights into the capability of plants to remove VOC from air, but such experiments focus on optimal plant growth conditions rather than on real life conditions. As outlined above, the experimental setup can have a crucial impact on the outcome of VOC exposure experiments. To be able to transfer data from chamber experiments to a typical room situation, it is important that the experimental conditions are similar and thus well controllable in the chamber.

In Germany, new construction products for indoor purposes have to be checked against the “principles of health-related evaluation of building products” (DIBt - Deutsches Institut für Bautechnik 2010). Therefore, the emissions of VOC are determined by test chamber measurements under standardized conditions (climate settings, product loading factor, etc.). In general, such test chambers are built from glass or stainless steel and are thus sufficiently inert, and their sizes are widely distributed from several liters up to 1 m^3. Additionally, common emission chambers have an adjustable air exchange which also allows a saturation of surfaces to then perform measurements that are not affected by adsorption (Massold et al. 2005). However, it is difficult to use such chambers as plant exposure chambers, because—since they are made for emission measurements—they are usually inadequately equipped with devices to control environmental factors for plants, such as PAR, relative humidity, and CO_2. In turn, the more devices installed for controlling such factors, the more difficult the analytical examination of VOC becomes, because all additional equipment creates a potential VOC sink or source.

So far, there is no commercial system or a guideline available which is purposed for plant exposure to VOC experiments. Therefore, test chambers and settings for exposure treatments of plants are usually self-made and thus very different, along with the results obtained. For example, Orwell et al. (2006) and Yang et al. (2009)

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treated *S. wallisii* with a similar amount of toluene (37.9 and 36.4 mg m^{-3}, respectively). Orwell et al. (2006) reported a removal efficiency of around 6300 μg m^{-3} m^{-2} h^{-1} (recalculated from 32.8 mg m^{-2} day^{-1}, chamber volume 0.216 m^3) while Yang et al. (2009) reported a far lower efficiency of 2.52 μg m^{-3} m^{-2} h^{-1}. One explanation for part of the discrepancies of the results is that Yang et al. (2009) refers to the aerial plant parts only while Orwell et al. (2006) refers to the entire plant. Furthermore, experiments by Yang et al. (2009) were conducted for 6 h with one VOC injection. In contrast, Orwell et al. (2006) treated plants for 5 days with several VOC injections. Besides, also the chamber design (size, construction, and material of chamber) and the environmental conditions likely affected the results.

One critical point in chamber experiments is the CO_2 content, which decreases quickly in gas-tight test chambers in the presence of plants. In our study, the CO_2 concentration would have dropped from 500 ppm to below 100 ppm within 2.5 h, if additional CO_2 would not have been added. Under low CO_2 conditions, the consumption of primary photosynthates is suppressed while the consumption of stored photosynthates is increased (Pärnik et al. 2007). At the same time, the role of photorespiration increases (Gerbaud and André 1980). Furthermore, falling CO_2 concentrations lead to larger stomatal apertures (Araújo et al. 2011), this of course has an impact on the pollutant removal efficiency.

The control of relative humidity is even more critical. In a gas-tight chamber, the relative humidity rises to more than 90% within minutes or hours, depending on chamber volume and plant size, due to plant transpiration. If water condenses on surfaces of the chamber, which is likely under high humidity, the VOC tested may dissolve to a certain degree in the water which would result in a decreased VOC concentration in the chamber atmosphere without a removal of VOC by plants. Furthermore, a high relative humidity leads to a change in plant physiology: the stomata will be mostly open and thus increasing the stomatal conductance (Turner 1991). In contrast, relative humidity indoors is usually much lower (around 40%) than in most chamber experiments; hence, stomata are often closed to prevent water loss. Because the stomata are one of the major pathways for pollutant uptake, the control of relative humidity would definitely be required for test chambers to achieve realistic results for indoor VOC removal.

Since the uptake of VOC by plants is not continuously linear, the experimental time span influences the results on VOC removal efficiencies. The uptake may slow or accelerate with time, depending also on VOC identity and concentration, saturation of plant cuticles and metabolic activity of plants, potting substrate including microorganisms and roots, and environmental factors. Thus, the results obtained can be regarded as snapshots from dynamic processes with uncertain progression.

Further, very little is known about toxicological effects on plants if they are exposed to VOC for several weeks or even months. Even though experiments under high VOC concentrations were performed, they cannot be used to predict effects on plants at low VOC concentrations (Cape 2003). It is possible that the removal efficiency decreases over time due to damage of the plant. Just as well, it is conceivable that the removal efficiency might increase due to a stimulation of plant metabolism or due to a promotion of microbial communities, including endophytes, microbes associated with the plant surface and those being present in the growing media.

Moreover, our literature survey shows that the removal efficiency of a certain VOC for a specific plant is not transferrable to other VOC or VOC mixtures or to another plant species. Thus, according to the state of the art, removal efficiencies estimated from chamber experiments seem to be difficult to transfer accurately to interior conditions. For this purpose, the design of the experiments and the test chamber and particularly the setting of the environmental parameters need to be reconsidered.

4 Conclusion and Outlook

Our experiments and the literature review revealed major challenges that need to be overcome in test chamber setups to assess the air purification capability of plants. It can be concluded, that test chambers used so far are only of limited suitability to draw exact conclusions for the removal efficiency of VOC by plants at typical indoor conditions. For this, the environmental conditions, which may have a crucial impact on results, are too different compared to interiors. The settings used in our experiments did not reflect real indoor conditions as well. However, this was necessary to overcome high VOC losses in the empty Biobox and to create optimal

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conditions for the plants to trace the limited VOC uptake. It is evident that the Biobox needs to be improved in regard to VOC adsorption to avoid masking of VOC removal by plants. The use of construction materials that are largely inert seems to be mandatory for test chambers. However, the Bioboxes are able to control environmental conditions very precisely and enable experiments under optional setting of important parameters (temperature, relative humidity, CO_2 content, and light conditions) which were consecutively recorded. Therefore, such Bioboxes can be used for example to examine physiological plant characteristics in response to pollutant exposure or to any other stressors under clearly defined conditions. For instance, effects of exposure to single VOC or VOC mixtures on plant physiology could be investigated. Furthermore, it also possible to investigate the influence of changing environmental conditions or of the genetic and physiological status of plants on the plant's VOC removal. Thus, the Bioboxes are well suited for basic research on physiological responses of plants to VOC exposure and to draw general conclusions on plant–environment interactions. Data on the removal efficiency of VOC by plants can be obtained, but cannot be directly transferred to a typical indoor situation.

Similar to the principals for evaluating building products, a guideline for determining the indoor VOC removal efficiency of plants would be helpful to achieve more realistic and coherent findings. Still today, chamber experiments are not well suited to determine the VOC removal efficiency of plants in interiors and results of different investigations are not comparable. However, the following must be regarded when developing such a guideline. The materials from which the chambers are built should be largely inert to avoid sorption of VOC on chamber surfaces. It should be distinguished between experiments with aerial plant parts and experiments with entire plants. In the latter case, the removal of VOC could be assigned to the different parts (foliage, roots, potting substrate) by appropriate additional experiments. Also the experimental time span and important sampling time points should be clearly defined. Since the removal efficiency is markedly influenced by ambient conditions, the test system should be able to control and maintain these conditions. For that, realistic indoor conditions should be set, including light, temperature, relative humidity, CO_2, and (natural) emission sources; the latter would involve a continuously emission of VOC mixtures in realistic concentrations. However, devices that are necessary to control critical environmental factors represent a potential VOC source (or sink), which might be difficult to predict or calculate and would make control measurements on empty chambers inevitable. In conclusion, the experimental set up comprises, besides test duration and distribution of sampling time points, three major parts which all need to be defined and standardized where possible to assess the indoor VOC removal by plants: (i) the test chamber design (e.g., construction materials and size), (ii) the treatments (e.g., VOC identity and concentration, plant species, and plant parts to be exposed), and (iii) the environmental conditions (e.g., light intensity, and climate).

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Suitability of test chambers for analyzing air pollutant removal by plants and assessing potential indoor air purification

Water, Air, & Soil Pollution

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Comparison of the empty glass versus Plexiglas Biobox

The design of the Bioboxes allows the use of hoods made of different materials. Plexiglas is not inert and thus can adsorb VOC itself, what was also revealed by first tests we conducted with the empty Biobox. Since glass is largely inert and not as reactive as Plexiglas, we tested the empty Biobox also with a glass hood expecting lower VOC adsorption. Both hoods were compared for their properties considering VOC adsorption (after injection of toluene) as well as VOC emission.

Material and Methods

The hood that we finally used in our experiments was made of Plexiglas panes that were connected (without glue) directly together (Fig. 1). The other hood, tested in advance, consisted of a metal frame to which glass panes were bonded with a low-emission silicon glue (Ottocoll S 610, Co. Hermann Otto GmbH, Fridolfing, Germany) (Fig. 2). For the empty Biobox pretests, the two hoods were successively connected to the same Biobox base and tested without the addition of plants or soil for VOC adsorption and VOC emission over 5 h using toluene as described in the manuscript (sample times: 6 min, 2.5 h, and 5 h past VOC injection). Depending on the hood used, the entire chamber system was hereafter referred to as "glass Biobox" or "Plexiglas Biobox".

Results and Discussion

Against our expectation, significantly lower toluene levels were detected in the glass Biobox than in the Plexiglas Biobox at 2.5 and 5 h past injection (Fig. 3). Because the glass hood was expected to be characterized by a lower VOC adsorption than the Plexiglas hood, the gas-tightness of the chambers was tested with CO_2 (data not shown). In doing so, the Plexiglas

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Biobox was found to be more gas-tight. Here, the concentration of a single dose of 1500 ppm CO_2 remained almost constant over a period of 48 h whereas the glass Biobox showed a fast decrease of the CO_2 concentration. A further examination, applying soapy water to junctions of glass panes and the metal frame of the glass hood which are subjected to pressure, revealed clear leakages. In comparison, no leakages were found for the Plexiglas hood. Thus, the less gas-tightness is believed to have caused the more pronounced VOC loss in the glass Biobox. Also the glue, used for bonding the glass panes to the metal frame, might have adsorbed some VOC. The potential adsorption by the gluing material and the low gas-tightness of the glass hood, also led to a higher standard deviation within the experiments. In contrast, the initial level of toluene and the concentration pattern over time could be well repeated in the Plexiglas Biobox.

Besides a potential adsorption, the sealing and gluing materials of the glass Biobox obviously emitted extremely high levels of VOC. Different siloxane peaks appeared in the chromatogram of the glass Biobox while the Plexiglas Biobox showed no undesired substances (Fig. 4).

Thus, although glass is theoretically the superior material for VOC removal experiments as its surface is not as reactive as Plexiglas surfaces, the glass Biobox was found to be less suitable. Accordingly, the Plexiglas chamber was used for further experiments due to the higher quality of the obtained results regarding gas-tightness, VOC adsorption, and VOC emission compared to the glass Biobox. The deficiencies of the tested glass Biobox can be contributed to construction faults of the glass hood which could not be fixed satisfyingly by the supplying company. It is evident that the tested glass chamber would require major modifications in mounting and sealing the glass panes before it could be used for VOC exposure experiments. It would probably be better to produce the glass hood from one piece, e.g. a bell-shaped hood on an appropriate base would be feasible, to avoid problems arising from glue or leakages. However, plant loading would be difficult because the whole hood must be lifted. The construction of a gas-tight, non-emitting and non-adsorbing test chamber, which is comfortable in handling and in which plant physiological parameters including light, CO_2 content and relative humidity can be controlled during VC exposure, is a quite challenging matter and until now such a chamber is not existing to our knowledge.

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Fig. 1 Plexiglas hood (without Biobox base; left), edges of Plexiglas hood, which are connected directly together

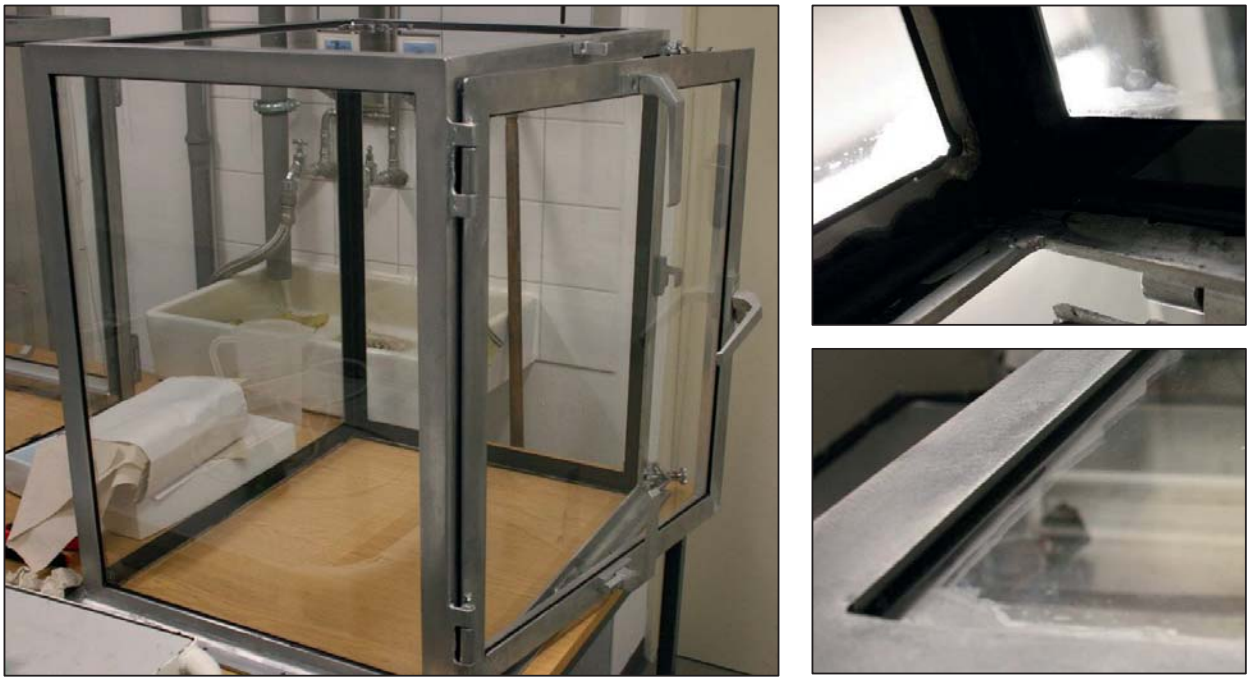

Fig. 2 Glass hood (without Biobox base; left), connection of glass panes to metal frame, interior view (top right), exterior view (down right)

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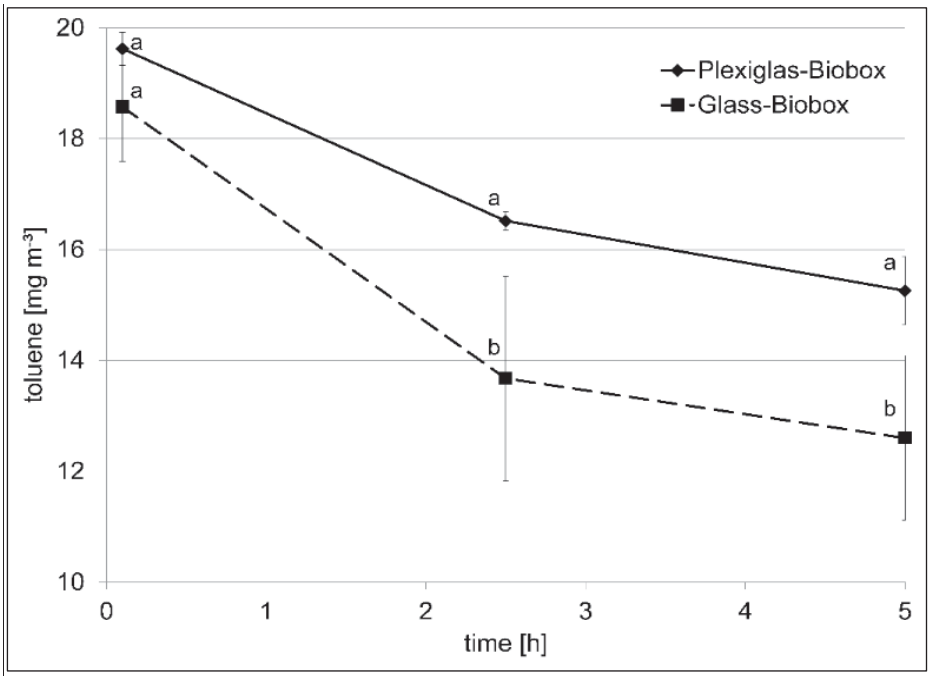

Fig. 3 Toluene concentration in the empty Plexiglas and the empty glass Biobox over time. Data are presented as means ± S.D. (n=4). Different letters indicate significant differences between treatments within each sampling point ($p \leq 0.05$, one-way ANOVA with post-hoc Tukey test)

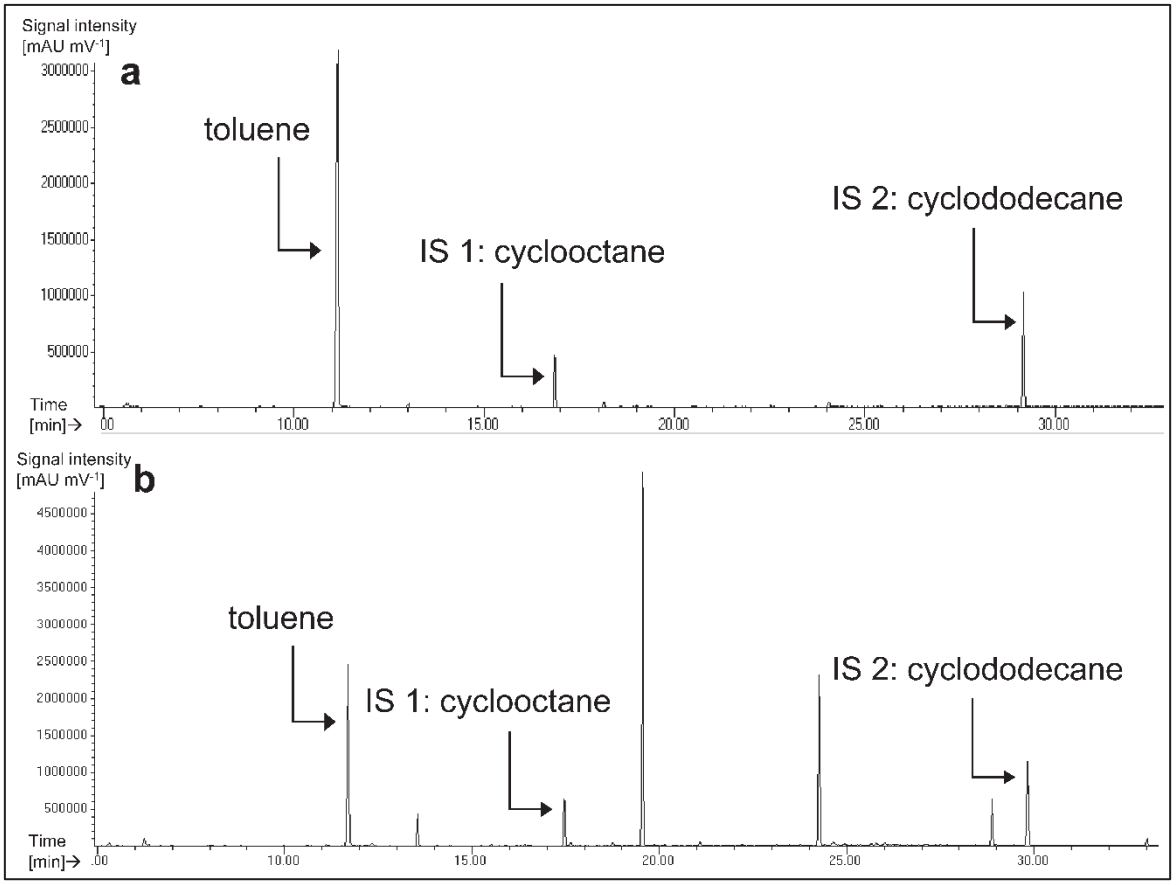

Fig. 4 Air samples from the Plexiglas (a) and the glass Biobox (b) 5 h after toluene injection. Unlabeled peaks represent different siloxanes. IS = internal standard

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Possible error sources of Bioboxes

Two test chambers (called Bioboxes) for experiments of this doctoral thesis were available. Both bioboxes, in combination with plant growth chambers, worked well in regard to the control of environmental conditions. In experiments under light, the CO_2 concentration showed fluctuations only from ca. 450 to 520 ppm (Hörmann et al. 2017a), caused by the delay between CO_2 input, homogeneous distribution within the Biobox, measurement of CO_2 concentration by infrared spectroscopy and the computer which operates the valve of the CO_2 bottle. Since the differences in CO_2 concentration should not affect photosynthesis of the plants too much, the control over the CO_2 concentration under light can be regarded as working well. In experiments under dark, the CO_2 concentration was not controlled, but recorded. The adsorption of CO_2, which was produced by respiration of plants, is actually possible in the Bioboxes by directing the CO_2 rich air through a cartridge filled with soda lime. However, this was not done to avoid a possible sorption of VOC on the soda lime. Of course high CO_2 concentrations, which raised up to more than 2000 ppm in our experiments, might have affected the physiology of plants and thus the VOC removal rate. Because the VOC concentration curves were similar for experiments with plants under light and dark, and the VOC removal by plants seemed to be primarily of passive action (Hörmann et al. 2017a, b) anyway, a disturbing influence by high CO_2 concentrations is not assumed.

While the humidity control worked well in the first Biobox, the second Biobox was not able to maintain the set relative humidity (RH) of 70% in every treatment under light, due to high transpiration of plants. Although both Bioboxes were more or less identical, some technical devices were different. In this special case, the Peltier elements of the heat exchangers, used for the control of RH, differed. The first Biobox was equipped with a Peltier element which was somewhat stronger than that in the second Biobox (unfortunately, another Peltier element of that type was no more available when the second Biobox was built). Therefore, the temperature of the heat exchanger in the first Biobox could be better reduced which led to more efficient RH control. Since under dark the amount of transpired water is much lower and therefore, easier to handle for the “weaker” Peltier element, the second Biobox was mostly used for experiments under dark, while the first Biobox was also suitable for experiments under light.

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Chapter II - Uptake of indoor pollutants by plants and plant physiology upon pollutant exposure

Uptake routes and metabolism of volatile organic compounds by plants

The uptake of VOC by plants can largely be divided in four groups: (i) foliar uptake, (ii) uptake by roots, (iii) uptake by the growing medium, and (iv) uptake by microorganisms residing in the growing medium (Dela Cruz et al., 2014). This paragraph focuses on the foliar uptake and the potential metabolism of VOC by plants. VOC may penetrate the foliage through the stomata or the cuticle (Kvesitadze et al., 2009; Riederer, 1995). The interrelation of the different pathways depends on the combination of volatility and lipophilicity of a particular VOC, together with the degree of stomatal opening and the lipid content of leaves (Collins et al., 2006; Riederer et al., 2002). Usually, low vapor pressure and high affinity to lipoid material will strongly favor the cuticular pathway, while compounds with an opposite combination of these properties will mostly diffuse through the stomata (Riederer et al., 2002). Compounds adsorbed to the cuticle may remain there or diffuse into underlying tissue. Since hydrophobic compounds have a high affinity for the cuticular lipids, the majority of such compounds is expected to remain in the cuticle with little tendency to migrate to other parts of the plant. Either way, in conjunction with the gas exchange, gaseous compounds like VOC can spontaneously enter the foliage through the stomata and directly penetrate the intercellular air space (Paterson et al., 1990). Once in the leaf, VOC may react, be retained there, or they may be transported through the phloem to the stem, roots or flowering parts (Cape, 2003a; Paterson et al., 1990).

To resist toxic actions of VOC, plants are able to activate different biochemical and physiological processes (Keymeulen et al., 1995; Kvesitadze et al., 2009):

- Excretion: VOC undergoes no chemical transformation, but translocation through the apoplast and excretion from the plant (Kvesitadze et al., 2009)
- Immobilization: storage of VOC in e.g. membrane lipids and essential oils (Keymeulen et al., 1995; Paterson et al., 1990)
- Functionalization: due to enzymatic oxidation, hydrolyses etc. VOC acquires a functional group (e.g. hydroxyl, amino), thus increasing polarity and reactivity of the molecule accompanying further transformation (Kvesitadze et al., 2009)
- Conjugation (basic process in phytoremediation): coupling of VOC to endogenous cell compounds (proteins, amino acids, saccharides, lignin etc.), thus decreasing toxicity

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and providing time for mobilization/induction of enzymes for further transformation (Kvesitadze et al., 2009; Sandermann, 1992)

- Deep degradation: oxidation to CO_2 and water or degradation to regular plant cell metabolites (Kvesitadze et al., 2009)
- Compartmentalization: storage of soluble conjugates in cell structures (primarily vacuoles), storage of insoluble conjugates in cell walls (Sandermann, 1992)

The fate of a particular VOC inside the plant depends on its chemical nature, the plant species, the phase of vegetation, and external factors such as temperature (Kvesitadze et al., 2009).

Phytotoxic effects of VOC

Studies regarding effects of VOC on plants are quite limited. Instead, many studies focused on the effects of inorganic gases on plants (Cape, 2003a), since such compounds, like e.g. NOx produced by fossil fuel combustion, have been found to be more toxic to plants than hydrocarbons (Bell et al., 2011; Kammerbauer et al., 1987). Only ethylene received considerable attention because it is a plant hormone and involved in a wide range of processes, such as growth regulation, flower development, and stress responses (Bell and Treshow, 2002; Cape, 2003a). Regarding other VOC, studies often focused on compounds evolved from herbicides, industrial sources or vehicle exhaust emissions. For instance, trichloroacetic acid was widely used as herbicide and has been frequently investigated in relation to forest decline (Bell and Treshow, 2002; Norokorpi and Frank, 1995). Other studies focused on effects of VOC which are persistent in the atmosphere like methyl t-butyl ether or peroxyacetyl nitrate (Beltagi, 2007; Cape, 2003b; Sparks, 2009). Thus, the targets in most investigations were herbaceous or other outdoor plants and compounds mainly found in the atmosphere. Phytotoxic effects were observed in many different terms, e.g. visible injury, damage to reproductive stages, metabolites composition, or changes in morphology (Cape, 2003a). Studies concerning phytotoxic effects of indoor-related VOC on houseplants are quite rare. Here, phytotoxicity was mostly measured in terms of visible injury to leaves and roots (e.g. browning of leaves) or in terms of photosynthetic performance, where the results are not generalizable.

Experiments by Yoo et al. (2006) revealed a deleterious effect in the rates of photosynthesis of different plant species (*Hedera helix* L., *Spathiphyllum wallisii* Regal, *Syngonium podophyllum* Schott., and *Cissus rhombifolia* Vahl.) after exposure to low concentrations of

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benzene and toluene (1 µl l^{-1}). Other experiments with toluene (27 µmol cm^{-3}) revealed no effect as shown for *Glycine max* L. (Jen et al., 1995) or only small transient effects on the photosynthetic performance of plants as shown for *Dieffenbachia amoena* (Porter, 1994). Photosynthesis of *Ficus benjamini* L. was negatively influenced by formaldehyde exposure already at 0.05 ppm, whereas *Epipremnum aureum* L. was not effected (Schmitz et al., 2000). Visible foliar injury of *Chlorophytum comosum* L. treated with formaldehyde at concentrations up to 10 µl l^{-1} could not be overserved (Giese et al., 1994). Also benzene, toluene, ethylbenzene, and xylene were not toxic, neither to leaves and roots nor to the photosynthetic apparatus of *Zamioculcas zamiifolia* after exposure to concentrations around 1 mmol m^{-2} for several days (Sriprapat and Thiravetyan, 2013). Foliar injury and a decrease in photosynthesis of plants exposed to xylene could be provoked in further experiments, but only at concentrations of 2000 ppm or more (Sriprapat et al., 2014). Also other experiments found visible phytotoxic effects of formaldehyde only when plants (*C. comosum*, *E. aureum*, and *Aloe vera*) were exposed to very high concentrations (6.5 - 11.5 µg m^{-3}) (Xu et al., 2011).

As discussed in chapter I, results of VOC removal by plants are difficult to transfer to real-life settings and it is still under discussion to what extent plants might have an impact on indoor air quality. Furthermore, although the uptake routs of pollutants by plants are well established, only little research had been directed toward the impact of plant physiology on pollutant uptake and VOC in particular. Vice versa, also many questions concerning phytotoxic effects of VOC on plants are unresolved. Thus, the following issues were investigated:

- How can the VOC removal by plants be assessed in regard to real-indoor environments?
- Impact of plant morphology and physiology on VOC removal
- Phytotoxic effects of VOC and parameters influencing VOC phytotoxicity

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RESEARCH ARTICLE

Assessment of filtration efficiency and physiological responses of selected plant species to indoor air pollutants (toluene and 2-ethylhexanol) under chamber conditions

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Abstract Three common plant species (*Dieffenbachia maculata*, *Spathiphyllum wallisii*, and *Asparagus densiflorus*) were tested against their capacity to remove the air pollutants toluene (20.0 mg m^{-3}) and 2-ethylhexanol (14.6 mg m^{-3}) under light or under dark in chamber experiments of 48-h duration. Results revealed only limited pollutant filtration capabilities and indicate that aerial plant parts of the tested species are only of limited value for indoor air quality improvement. The removal rate constant ranged for toluene from 3.4 to 5.7 L h^{-1} m^{-2} leaf area with no significant differences between plant species or light conditions (light/dark). The values for 2-ethylhexanol were somewhat lower, fluctuating around 2 L h^{-1} m^{-2} leaf area for all plant species tested, whereas differences between light and dark were observed for two of the three species. In addition to pollutant removal, CO_2 fixation/respiration and transpiration as well as quantum yield were evaluated. These physiological characteristics seem to have no major impact on the VOC removal rate constant. Exposure to toluene or 2-ethylhexanol revealed no or only minor effects on *D. maculata* and *S. wallisii*. In contrast, a decrease in quantum yield and CO_2 fixation was observed for *A. densiflorus* when exposed to 2-ethylhexanol or toluene under light, indicating phytotoxic effects in this species.

Keywords Ornamental indoor plants · Indoor air quality · Air purification · Volatile organic compounds (VOC) · Photosynthesis

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Introduction

Volatile organic compounds (VOC) belong to the most important pollutants indoors (Wolkoff and Nielsen 2001) and are emitted from, e.g., building materials and office equipment (Destaillats et al. 2008; Que et al. 2013). The concentration of total VOC indoors is usually rather low and reported values are, e.g., around 150 μg m^{-3} measured in offices in the USA (Chin et al. 2014) or around 290 μg m^{-3} in Germany (Schulz et al. 2010). However, depending on indoor sources, occupant behavior, etc., certain VOC can occur at very high concentrations like toluene, for which a maximum of 2400 μg m^{-3} could be found when investigating several offices in Germany (Schulz et al. 2010) or 1,4-dichlorobenzene with a maximum of 2126 μg m^{-3} found in the USA (Chin et al. 2014). Human exposure to VOC has in general been related to dizziness, palpitation, and the irritation of eyes, respiratory tract, and skin, whereas the toxicity of a particular VOC depends on its chemical identity (Sarigiannis et al. 2011). For instance, the toxicologically derived guideline value, published by the German Environment Agency, is very low for pentachlorophenol (0.1 μg m^{-3}) and relatively high for monocyclic monoterpenes (1000 μg m^{-3}) (Umweltbundesamt 2017).

Currently, there is no fully satisfying method for air purification (Luengas et al. 2015), but plants may be capable of

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making a substantial contribution to indoor air quality (Orwell et al. 2006). In general, the uptake of VOC can occur through the aerial plant parts (cuticle and stomata), the roots, microorganisms residing in the growing media, and the growing media itself (Dela Cruz et al. 2014). However, in many experiments, it is not clear whether the plant and/or the substrate (with the contained microorganisms and roots) adsorbed the pollutants in particular. Conclusions of previous studies concerning the efficiency of plants as indoor air purifiers differ and the capacity of plants to remove VOC is influenced by many factors, among those are for example plant species, light intensity, and pollutant identity and concentration. Some authors outline plants and plant-soil systems as good air purifiers (Wolverton and Wolverton 1993; Orwell et al. 2006; Xu et al. 2011), others suggest no major impact of plants on indoor air quality (Schmitz et al. 2000; Girman et al. 2009; Llewellyn and Dixon 2011).

The impact of plant physiological characteristics on VOC removal is largely unknown. Vice versa, only little is known about the influence of VOC exposure on plant physiology. So far, physiological responses of plants to pollutant exposure were mostly investigated for outdoor pollutants, mainly inorganic gases (Cape 2003a) and particulate matter (Ulrichs et al. 2008). Literature concerning the influence of indoor pollutants on the physiology of ornamental plants is rare. Available studies mainly focus on photosynthetic performance (Porter 1994; Jen et al. 1995; Schmitz et al. 2000; Yoo et al. 2006; Sriprapat and Thiravetyan 2013; Sriprapat et al. 2014) or on visible symptoms of leaves and roots (Giese et al. 1994; Xu et al. 2011; Sriprapat and Thiravetyan 2013) in dependence of VOC. However, many aspects regarding the correlation between VOC exposure and plant physiology are unknown so far.

The aim of our study was to evaluate the capability of plants to purify indoor air under well-controlled conditions that also allow determination of physiological effects of VOC on plants. Therefore, three different plant species (*Dieffenbachia maculata*, *Spathiphyllum wallisii*, and *Asparagus densiflorus*) that are common indoor ornamentals were exposed to two different VOC: toluene or 2-ethylhexanol. These VOC were chosen because they are important indoor pollutants (Nalli et al. 2006; Sarigiannis et al. 2011) and are chemically dissimilar. Toluene is an aromatic hydrocarbon with relatively high chemical stability and rather low biological activity (Keymeulen et al. 1995). The aliphatic alcohol 2-ethylhexanol is more reactive than toluene, due to its hydroxyl group and acyclic structure. Exposure of plants to either a biological inactive compound or a compound of higher reactivity may trigger different responses in plant physiology that might also affect VOC removal. To identify possible correlations between plant physiology and VOC treatment, different plant physiological characteristics (transpiration, CO_2 fixation/respiration, and photosystem quantum yield) were determined upon VOC exposure and in unexposed controls.

Material and methods

Test plants

Experiments were performed with *Dieffenbachia maculata* "compacta" (Lodd. et al.) G. Don (*Araceae*), *Spathiphyllum wallisii* "Daniel" (Regel) (*Araceae*), and *Asparagus densiflorus* (Kunth) Jessop (*Asparagaceae*). These plant species are typically propagated vegetatively via, e.g., cuttings or dividing of rhizomes. *D. maculata* and *S. wallisii* (pots with 12-cm diameter) were purchased from a wholesale market in Germany in a retail-ready stage (*S. wallisii*: in flower stage). After purchase, the plants were cultivated in a growth chamber (Co. Conviron, Adaptis A 1000) with 15 h of illumination per day (100 ± 10 μmol m^{-2} s^{-1}), temperature set to 22/20 °C (day/night), and a relative humidity (RH) of 60–70% for at least 3 days prior to experiments. *A. densiflorus* (pots with 25-cm diameter) were provided by the horticultural experimental station of the Humboldt-Universität zu Berlin. There, the plants had been cultivated under greenhouse conditions (natural day/night rhythm, summer season) without extra illumination.

The three plant species analyzed in the current study are characterized by marked differences in morphology (see also Online Resource 1). *D. maculata* is a shrubby plant and has an upright shape. The relatively large and waxy leaves, which are mottled with white, are located on several stems emanated from one centrum. *S. wallisii* is an herbaceous plant, which forms offsets at the base and has therefor a clump-forming growth habitat. The dark green and glossy leaves with their long stalks are basal and shaped lanceolate to ovate. It produces flowers which are born in dense spadix with an ovate white spathe. *A. densiflorus* appears bushy or fern-like with a scrambling growth habitat. It has foxtail-like stems with many branches holding the small, liner, needle-like leaves. Because the plants of *A. densiflorus* used in our study were relatively big, only one plant could be tested per experiment replicate. In contrast, plants of the other two species were smaller and slimmer; hence, three plants per replicate were tested.

Experimental setup and test procedure

Two identical gas-tight Plexiglas chambers, called Bioboxes (purchased from the company GMS—Gaswechsel-Messsysteme GmbH, Berlin, Germany), with a total volume of 240 L each and a removable front cover for plant loading, were used. Included instruments allow to log and control the relative humidity and the CO_2 content as follows. The humidity is controlled by a heat exchanger where surplus water

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condensates. From there, the water is fed to a beaker in 30-min intervals. During the experiments, the CO_2 concentration was continuously measured by infrared spectroscopy. If the concentration dropped below the set point, CO_2 was automatically reinjected into the system. A connected PC recorded the injected amount of CO_2, reflecting the CO_2 fixation (see also below). The chambers are provided with two stainless steel fittings to allow VOC injection and air sampling. Furthermore, the chambers are fitted with two fans each to promote complete mixing of the VOC-loaded air. Each Biobox was placed in a growth chamber (Adaptis A1000, Co. Conviron, Winnipeg, Canada) which controlled light intensity and temperature. In all experiments, the climate conditions were set as follows CO_2 = 500 ppm, RH = 70%, temperature = 22 °C, light = 180 ± 10 μmol m^{-2} s^{-1} or dark.

Per Biobox, three plants of *D. maculata* or *S. wallisii* or one plant of *A. densiflorus* were used for chamber experiments (see also Online Resource 1), representing a leaf area of on average 0.72 ± 0.08, 0.57 ± 0.04, and 0.94 ± 0.46 m^2, respectively. Prior to VOC injection, plants were watered to saturation (until all pores of the potting substrate were filled with water) and placed in the Bioboxes under experimental conditions. Plants were allowed to drain for 1 h and get acclimatized to the set conditions. The pots and the substrate surface were entirely covered with aluminum foil, so that just the aerial plant parts would be exposed to the VOC. After 1 h of acclimatizing in the Bioboxes, the quantum yield of photosystem II (PS II) was determined as described below. Subsequently, the Bioboxes were sealed, and one of the VOC was introduced.

Plants were exposed to a concentration of 20 mg m^{-3} toluene (by injecting 5.5 μl of ≥ 99.5% toluene for synthesis, Co. Carl Roth GmbH & Co. KG, Karlsruhe, Germany) or 14.6 mg m^{-3} 2-ethylhexanol (by injecting 4.2 μl of 99% 2-ethyl-1-hexanol, Co. Alfa Aesar, Karlsruhe, Germany) for 48 h. To analyze the concentration of VOC in the chamber air, air samples were taken using sorption tubes (C1-CXXX-5003, Tenax TA. C6-C30. Inert-coated., Co. Markes, Frankfurt, Germany) spiked with internal standards (100 ng of each, cyclooctane and cyclododecane). The first air sample in the experiments with toluene was taken 6 min after injection, after an initial equilibrium was achieved. In experiments with 2-ethylhexanol, the first air samples was taken 1 h after injection. Due to a low vapor pressure, this VOC needed much longer to enter the gas phase; here, an initial equilibrium was thus hardly obtained. The best time point for the first air sampling was determined in pretests using empty chambers (data not shown). Following air samples were taken after 5, 24, 29, and 48 h for both VOC. All air samples were taken with a flow rate of 100 ml min^{-1} and a duration of 30 and 60 s for toluene and 2-ethylhexanol, respectively. After VOC exposure, the chlorophyll fluorescence was measured again.

Experiments without exposing plants to VOC were used as controls for analyzing physiological plant responses to VOC. For the determination of VOC removal by plants, controls with empty chambers were conducted and results on VOC removal rates of plants were corrected using the empty chamber data. Therefore, average loss of VOC in empty chambers after 48 h was added to the measured value of VOC after 48 h in each experimental run with plants. The obtained value was then used as final concentration in the equation given below. All experiments were replicated fourfold under continuous light and under continuous dark, always using another set of plants (that was not used before) in every replicate.

Determination of plant leaf area

The total leaf area was carefully determined for all test plants after performing the chamber experiments. For *D. maculata* and *S. wallisii*, this was done with a leaf area meter (3100 Area Meter, Co. LI-COR. Inc., Lincoln, NE, USA). Due to the high number of very small three-sided leaves of *A. densiflorus* (see also Online Resource 1c), the leaf area determination with the leaf area meter was here not possible. Instead, the leaf surface of 25 leaf-sides was measured using a digital microscope (VHX-1000, Co. Keyence, Neu-Isenburg, Germany), the number of leaves per branch was counted as well as the number of branches per 20-cm stem (each n = 5). So the average leaf surface of 1-cm stem could be estimated. The length of all stems per test plant was determined and the entire leaf surface was extrapolated.

Analysis of morphological plant parameters and stomatal conductance of unexposed plants

Leaf surface structure as well as density and conductance of stomata were examined for representative plants (not VOC exposed) of *D. maculata* and *S. wallisii*. Due to the leaf shape of *A. densiflorus*, these analyses were not performed for this plant species.

The surface structure and stomatal distribution of leaves were determined using negative imprints. Therefore, transparent nail polish was applied on the abaxial and adaxial surfaces of two leaves of five plant (representing 10 negative imprints) and allowed to dry for 5 min. Afterwards, the polish was removed with an adhesive tape which was subsequently transferred on a microscope slide. The digital microscope (VHX-1000, Co. Keyence) was used to determine the wax deposits and stomata density. Stomata were counted on five different square millimeter per negative imprint, resulting in a sample size of 50 mm^2 per plant species.

The stomatal conductance of *D. maculata* and *S. wallisii* was non-invasively measured with help of a portable diffusion porometer (AP4 Porometer, Co. Delta-T Devices Ltd., Cambridge, UK). Twelve plants per species (not exposed to any VOC) were placed in a growth chamber (Adaptis A1000, Co. Conviron) under experimental conditions and allowed to

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acclimatize for 1 h. From each plant, two older and two younger leaves were examined (in total $n = 48$).

Analysis of physiological plant parameters upon VOC exposure

The quantum yield, CO_2 fixation/respiration, and transpiration were analyzed for all three plants species upon VOC exposure and in control experiments without exposing plants to VOC.

The quantum yield of PS II was non-invasively determined after acclimatization in the Bioboxes and at the end of each experiment. For this, chlorophyll fluorescence was measured in the absence of photosynthetic light with a portable chlorophyll fluorometer (MINI-PAM, Co. Walz, Effeltrich, Germany) and the help of Leaf-Clip Holder 2030-B (Co. Walz). The quantum yield of PS II was automatically calculated by the MINI-PAM. Plants of experiments under light were put in a dark environment 10 min prior to measurements. For *D. maculata* and *S. wallisii*, chlorophyll fluorescence was measured on four leaves per plant with three replications per species. Because experiments on *A. densiflorus* were performed with only one big plant per Biobox, chlorophyll fluorescence was here measured on 12 leaves (i.e., four leaves of three different branches) per plant. For statistical analysis, the average chlorophyll fluorescence per plant (for *D. maculata* and *S. wallisii*) or per branch (for *A. densiflorus*) was used ($n = 3$).

CO_2 fixation rate and the respiration of all plants per Biobox under light and dark, respectively, were determined in each chamber experiment ($n = 4$). During experiments, the CO_2 concentration in chamber air was measured continuously by infrared spectroscopy. Under light, the CO_2 concentration decreased due to photosynthesis. If the concentration dropped below the set point, CO_2 was automatically reinjected into the system and the amount of reinjected CO_2, thus CO_2 fixation rate, was recorded. Under dark, respiration of plants was monitored by recording the increase of CO_2 concentration. Data on CO_2 fixation/respiration were expressed on a leaf area basis to obtain comparable results for gas exchange rates.

The transpiration rate of all plants per chamber that took place during the time span of each experiment was determined by quantifying the transpired water that was collected in the beaker of the Biobox system. The volume of the transpired water was related to the experimental duration and the transpiration rate was expressed on a leaf area basis to obtain comparable results for gas exchange rates.

Trace gas analysis and determination of VOC removal

The sampling of chamber air is described above in the experimental setup. Prior to analysis of VOC in chamber air samples, sorption tubes were desorbed using a Perkin Elmer Turbomatrix thermal desorber. For gas chromatographic analysis, an Agilent 6890 gas chromatograph (Agilent Technologies, Inc., Santa Clara, USA) with flame ionization detector, coupled to a mass spectrometer (Agilent Technologies, Inc., 5979 MSD), was used. Helium was used as carrier gas and analysis were performed according to DIN ISO 16000-6.

VOC concentrations were expressed in micrograms per cubic meter and were corrected with those of empty chamber experiments by adding the average loss of VOC in empty chambers after 48 h to the final concentration in each experiment with plants. Calculation of the VOC removal rate constant was based on the 48-h data to cover also potential effects of active uptake instead of passive sorption only. Assuming an exponential VOC decrease (see Online Resource 2 for change in VOC concentrations over time), the removal rate constant of VOC was calculated for each combination of VOC, plant species, and light condition using the following formula (Girman 1992). In doing so, data were normalized according to chamber volume and experimental duration and scaled up to 1-m^2 leaf area:

$$\text{VOC } \textit{removal rate constant } \left[\text{L m}^{-2}\,\text{h}^{-1}\right] = \left(-\left(1/t\right)\ln\left(C/C_0\right)\right) \times V/A$$

where t is the experimental time span [h], C is the final concentration of VOC [$\mu g\ m^{-3}$], C_0 is the initial concentration of VOC [$\mu g\ m^{-3}$], V is the chamber volume [L], and A is the total leaf area [m^2]. To consider uncertainties of the measured VOC concentrations and the leaf area, data for every experimental run were calculated individually. Afterwards, the means and standard deviations were determined.

Statistical analyses

Tests for significant differences with $p \leq 0.05$ were performed with the statistical software package SPSS 23 (IBM). To check for significant differences between the tested light conditions (within each VOC and each plant species) or between the tested plant species (per VOC and light condition) sub-sets of data were analyzed by one-way ANOVA and post hoc Tukey's HSD test.

To identify main factor(s) and putative interactions between factors in the entire 3 × 2 × 2 experimental setup (plant species, VOC identity, and light condition) regarding the VOC removal constant, a three-way ANOVA with post hoc Tukey's HSD test was applied. To find possible correlations between plant physiological parameters (CO_2 fixation/respiration and transpiration) and VOC removal constant, a Pearson's correlation analysis was performed. For the three-way ANOVA and the correlation analysis, the data were log-transformed in advance to establish normal distribution. To balance missing

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symmetry, it was necessary to add a constant to particular data prior to applying the log. Data on CO_2 fixation/respiration and transpiration from VOC-exposed plants had been offset against data from control plants prior to correlation analysis. For that, mean values of the control experiments were subtracted from the individual values obtained in every experiment with VOC-exposed plants.

Results and discussion

Characterization of the test plants

We selected three common indoor ornamentals that are characterized by marked differences in morphology. Besides differences in the macroscopic surface structure (e.g., leaf size and shape), that are most pronounced for *A. densiflorus*, differences also occur on microscopic level (see representative micrographs in Online Resource 1). Negative imprints of leaf surfaces were analyzed for *D. maculata* and *S. wallisii* where epicuticular waxes and stomata were well depicted (Online Resource 1). The wax structures of both species were pronounced with prominent ridges, but the wax depositions of *D. maculata* were somewhat rougher, thicker, and more distinctive than that of *S. wallisii*. Further, *D. maculata* had only stomata on the abaxial leaf surface, whereas *S. wallisii* had a few on the adaxial leaf surface as well (Fig. 1).

Although the abaxial stomata density was found to be similar for *D. maculata* (30.3 ± 4.6 mm^{-2}) and *S. wallisii* (29.6 ± 6.6 mm^{-2}) (Fig. 1), the stomatal conductance measured on the abaxial surface was significantly higher for *S. wallisii* (0.12 ± 0.03 cm s^{-1}) in comparison to *D. maculata* (0.03 ± 0.01 cm s^{-1}) (Fig. 2). A higher stomatal conductance may be an indication of a higher metabolic activity, which was also indicated by a higher transpiration rate of *S. wallisii* (14.8 ± 1.2 ml h^{-1} m^{-2}) under light without VOC exposure compared to the leaf ornamentals *D. maculata* (8.4 ± 1.0 ml h^{-1} m^{-2}) as well as *A. densiflorus* (5.7 ± 0.4 ml h^{-1} m^{-2}) (Fig. 3). In contrast, all three species had a similar CO_2 fixation rate (2.5 ± 0.4, 1.7 ± 0.7, and 1.8 ± 0.3 μmol s^{-1} m^{-2} for *D. maculata*, *S. wallisii*, and *A. densiflorus*, respectively) and a similar respiration (1.2 ± 0.1, 1.4 ± 0.2, and 1.0 ± 0.1 μmol s^{-1} m^{-2} for *D. maculata*, *S. wallisii*, and *A. densiflorus*, respectively) when not exposed to any VOC (Fig. 4). The results of chlorophyll fluorescence measurements are presented in Fig. 5. The results are expressed as quantum yield, reflecting the proportion of photons absorbed by PS II, thus indicating the efficiency of PS II (Maxwell and Johnson 2000). This measurement was used as additional marker for the photosynthetic activity (and for the stress response of the plant to VOC). Compared to *D. maculata* (0.803 ± 0.008) and *S. wallisii* (0.808 ± 0.008), *A. densiflorus* (0.752 ± 0.012) showed under light a somehow lower basal quantum yield.

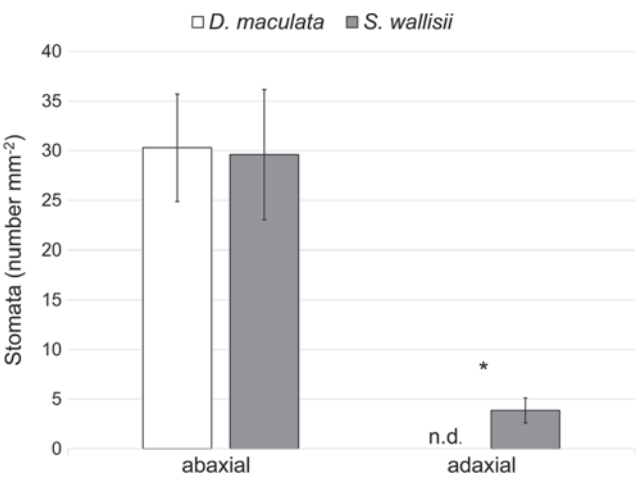

Fig. 1 Stomatal density of *Dieffenbachia maculata* and *Spathiphyllum wallisii*. Data are presented as mean values ± SD (n = 50). Asterisks indicate significant differences between plants. *n.d.* not detected

VOC removal by aerial plant parts in chamber experiments

It is suggested that the uptake of lipophilic VOC by aerial plant parts can be divided in two main actions, the uptake by the cuticle and the uptake by stomata (Paterson et al. 1990; Dela Cruz et al. 2014). Initially, the uptake is dominated by adsorption effects to the cuticle. This is a process which is of passive nature and thus strongly influenced by VOC concentration and lipophilicity (Paterson et al. 1990). Furthermore, it seems to be a relatively fast action that slows down with decreasing availability of vacant surface sites for sorption on the cuticle and will likely follow an exponential decay. Once the cuticle is (almost) saturated, the stomatal uptake becomes the

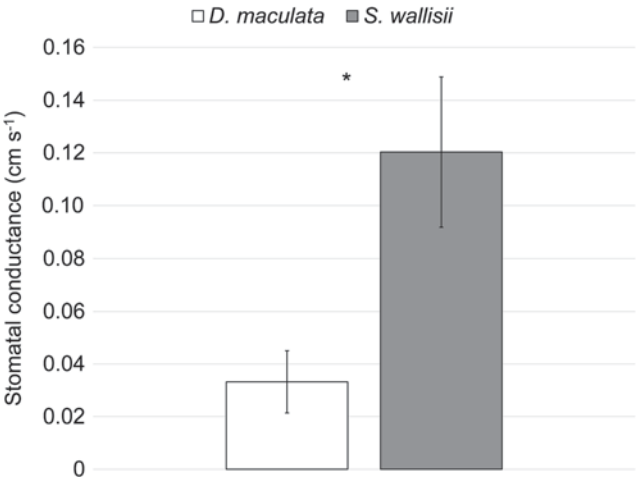

Fig. 2 Stomatal conductance of *Dieffenbachia maculata* and *Spathiphyllum wallisii*. Data are presented as mean values ± SD (n = 48). Asterisks indicate significant differences between plants

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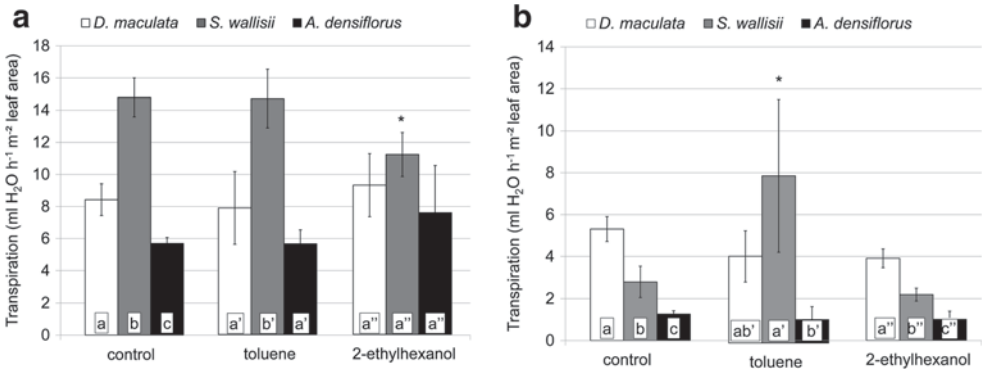

Fig. 3 Transpiration under light (**a**) and dark (**b**) of *Dieffenbachia maculata*, *Spathiphyllum wallisii*, and *Asparagus densiflorus* during VOC exposure for 48 h. Values are presented as means ± SD (n = 4). Different letters indicate significant differences between plants and within treatments, and asterisks indicate significant differences between treatments within plant species ($p \leq 0.05$, one-way ANOVA, followed by Tukey's HSD test)

dominant action of VOC removal. Here, the limiting factor of VOC uptake would be the subsequent translocation and/or metabolism of VOC. The stomatal uptake is believed to be reduced in speed compared to sorption and highly dependent on VOC identity (e.g., kind of functional groups) and on genetic and physiological status of plants (e.g., existence and activity of relevant metabolic pathways for VOC degradation/transformation, stomatal conductance). In general, VOC that are biologically inactive may be stored in plant tissue, rather than metabolized (Paterson et al. 1990). This would lead to an equilibration between concentration in leaves and concentration in air (Cape 2003a), and further uptake would nearly come to a stop. In contrast, VOC that are biologically active might undergo further transformation to decrease their toxicity (Kvesitadze et al. 2009). Thus, their uptake would continue and may even accelerate due to an induction of biosynthesis and activity of relevant enzymes.

The VOC removal curves obtained in our experiments are steeper at the beginning of exposure and become more flattened in the curse of time (see Online Resource 2). Furthermore, our correlation analyses revealed no relation of gas exchange and VOC uptake (see below). This indicates that the first action of uptake—namely the passive sorption on plant surfaces—is predominant in our experiments. Therefore, an exponential function was chosen to determine the VOC filtration of plants, which was expressed as removal rate constant. This expression has the advantage that it can be compared with volumetric flow rates, expressing how much liters of air get cleaned from VOC by 1-m^2 leaf area per hour. Hence, a comparison of data with other pollutant removal processes is possible, like absorption-based air filtration or natural ventilation rates. However, since the VOC removal efficiency by plants can generally be dependent on many factors (including plant characteristics, VOC identity and concentration, and environmental conditions), it should be mentioned that the presented values are only valid for the given experimental setup. Further, data on 2-ethylhexanol removal are only indicative for the removal capacity by aerial plant parts. Here, due to the low vapor pressure, it was not possible to apply concentrations as high as for toluene.

The VOC removal rate constants achieved in our study by the aerial parts of the investigated plant species are outlined in Table 1. Our calculation was conducted for every single experimental run and therefore, included every single value of initial

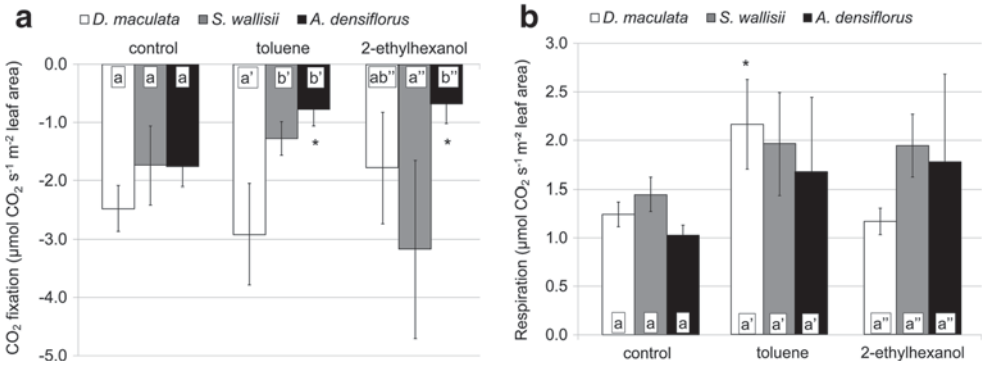

Fig. 4 CO_2 fixation during light (**a**) and respiration during dark (**b**) of *Dieffenbachia maculata*, *Spathiphyllum wallisii*, and *Asparagus densiflorus* during VOC exposure for 48 h. Values are presented as means ± SD (n = 4). Different letters indicate significant differences between plants and within treatment, and asterisks indicate significant differences between treatments within plant species ($p \leq 0.05$, one-way ANOVA, followed by Tukey's HSD test)

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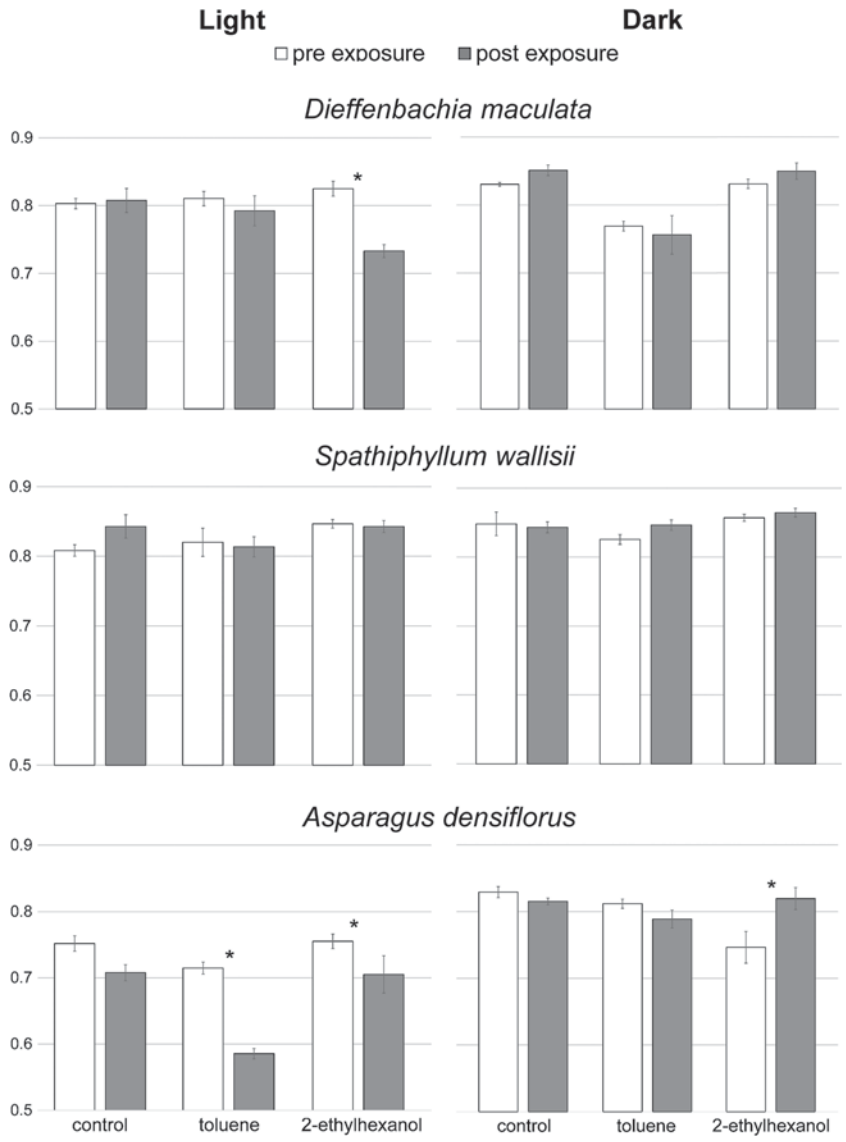

Fig. 5 Quantum yield of *Dieffenbachia maculata*, *Spathiphyllum wallisii*, and *Asparagus densiflorus* under continuous light and continuous dark. Values are presented as means ± SD ($n = 3$) of measurements before and after VOC exposure over 48 h. Asterisks indicate significant differences between pre and post exposure ($p \leq 0.05$, one-way ANOVA, followed by Tukey's HSD test)

Table 1 Removal rate constant for 2-ethylhexanol and toluene in experiments with *Dieffenbachia maculata*, *Spathiphyllum wallisii*, and *Asparagus densiflorus* under different light conditions (light/dark). Values are means ± SD ($n = 4$) after 48 h of VOC exposure

	Plant species	Removal rate constant [L h^{-1} m^{-2} leaf area]	
		Light	Dark
2-Ethylhexanol	*D. maculata*	1.8 ± 0.2	1.4 ± 0.1*
	S. wallisii	2.4 ± 0.2	1.9 ± 0.2*
	A. densiflorus	2.0 ± 1.1	1.7 ± 1.0
	Significance among species	n.s.	n.s.
Toluene	*D. maculata*	5.6 ± 1.8	5.5 ± 1.2
	S. wallisii	5.7 ± 1.5	4.0 ± 0.8
	A. densiflorus	4.0 ± 2.1	3.4 ± 2.0
	Significance among species	n.s.	n.s.

Asterisks indicate significant differences between light conditions within plant species and VOC identity ($p \leq 0.05$, one-way ANOVA, followed Tukey's HSD test)

n.s. not significant

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and final VOC concentration as well as leaf area. Hence, uncertainties of these parameters are considered in the presented data. Overall, the data ranged from 1.4 ± 0.1 to 5.7 ± 1.5 L h^{-1} m^{-2} leaf area. To identify the main factor(s) of the VOC removal, a three-way ANOVA was performed. Although the Levene' test showed that variance homogeneity was not fulfilled, the ANOVA was used for this analysis since it is robust against such violations if normal distribution is met and sample size is equal (which was the case in the present study). The main factor in our experiments on the VOC removal rate constant was the VOC identity, which had a significant effect ($F(1, 36) = 60.4$, $p = .000$, partial $\eta^2 = .626$). The plants species showed only in tendency an effect ($F(2, 36) = 3.0$, $p = .061$) and the light condition showed no significant impact ($F(1, 36) = 2.4$, $p = .128$). An interaction between these factors was not found.

In our study, the removal rate for toluene was in general around three to four times higher than for 2-ethylhexanol (Table 1). This may be caused by exposing plants to a higher concentration of toluene. In case of 2-ethylhexanol, nearly two thirds of the injected amount appeared to get lost due to absorption on chamber surfaces within the first hour, as revealed by the control with empty chambers (see Online Resource 2). Thus, the actual concentration of 2-ethylhexanol in the chamber air was approximately three to four times lower in comparison with toluene (for toluene, the concentration of the first air sample corresponded satisfyingly with the injected amount as shown in Online Resource 2). Due to these uncertainties, the results on removal of 2-ethyhexanol have to be considered carefully.

The VOC removal rate achieved by the aerial plant parts seems, however, to be too low to be of impact for indoor air quality. For instance, the toluene removal rate constant of *D. maculata* (in regard to 1-m^2 leaf area) corresponds to a ventilation rate of approximately 0.0002 h^{-1} in a 30-m^3 room. This is not even close to a low natural ventilation rate of 0.2 h^{-1} of highly airtight houses (Münzenberg et al. 2003). Thus, more than 1000 m^2 of leaf area would be necessary to gain a similar ventilation rate in a 30-m^3 room. Basically, it is suggested that aerial plant parts do not improve indoor air significantly regarding VOC pollution. This statement is underlined by different authors (Levin 1992; Schmitz et al. 2000; Girman et al. 2009; Llewellyn and Dixon 2011; Hanoune et al. 2013). The filtration capability, metabolism, etc. may be different for plant-soil systems, especially those containing a potent microflora and that are equipped with devices which allow an active ventilation of the substrate as described by Llewellyn and Dixon (2011). However, such systems were not subject of the current study.

Putative impact of plant characteristics on the VOC removal rate

In general, the VOC removal efficiency might differ between plants depending on their genetic and physiological status. The three-way ANOVA showed in tendency an effect of the plant species on the VOC removal rate constant ($F(2, 36) = 3.0$, $p = .061$). Performing chamber experiments under highly controllable and repeatable conditions enables evaluation of plant physiological parameters in parallel to VOC exposure. To check whether in the current study a relation exists between the VOC removal rate and key parameters for the plant's gas exchange, a correlation analysis was done. The Pearson's correlation revealed a significant linear correlation between CO_2 fixation (light)/respiration (dark) and transpiration ($p = .004$, $r = .410$). This result was expected, since both parameters are related to the gas exchange of plants and are therefore connected naturally. However, the VOC removal rate constant was found to correlate neither with CO_2 fixation/respiration, nor with transpiration. The correlation of these physiological parameters with the removal constant for each VOC singly revealed equal results. This indicates that most of the VOC was removed passively (first action of uptake) and got likely adsorbed on the cuticle. The fact that hydrophobic chemicals (like toluene and 2-ethylhexanol) have a high affinity for cuticular lipids (Paterson et al. 1990) supports this finding. Furthermore, the uptake of toluene (that is due to its aromatic nature supposed to be largely biological inactive) may be a simple physicochemical process, with an equilibration between concentration in air and in leaves (Cape 2003a). Also Wetzel and Doucette (2015) found that changing concentration of six different VOC, including toluene, in air paralleled the corresponding VOC concentration in leaves of four different plant species (*Ficus benjamina*, *Epipremnum aureum*, *Chlorophytus comosum*, and *Schlumbergera truncate*). The authors found that the leaf air concentration factors (VOC concentration in leaves divided by VOC concentration in air) generally increased with increasing lipid content of leaves, implying again a passive uptake by sorption to the cuticle (Wetzel and Doucette 2015).

For toluene, the removal rate constant was found to be similar for all plant species under light and under dark (Table 1). Since plants largely close their stomata during night, it seems that toluene uptake through the stomata was indeed negligible. In contrast, the 2-ethylhexanol removal rate constant was for *D. maculata* and *S. wallisii* significantly higher under light (when stomata are open) versus dark (when stomata are almost closed). This indicates an uptake of 2-ethylhexanol not only by cuticle sorption but also through the stomata, probably followed by degradation/transformation of this VOC which would have facilitated the stomatal uptake. Different to the other investigated species, *A. densiflorus* did not show a significantly higher removal rate of 2-ethylhexanol under light versus dark, which might also be attributed to a higher variation in data (likely caused by the estimation of leaf area, which seems to be not sufficiently accurate due to the filigree leaves and/or by a higher biological variation since here only one plant per chamber could be used

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due to the size of the plants). Furthermore, due to experimental timeline and low concentration of 2-ethylhexanol after surface sorption, it may be difficult to observe stomatal uptake of this compound. Nonetheless, *S. wallisii* showed in tendency the highest removal of 2-ethylhexanol (Table 1), which might have been facilitated by the high stomatal conductance (Fig. 2).

Other studies have also shown that the level of VOC removal can depend on plant species (Wolverton and Wolverton 1993; Liu et al. 2007; Yang et al. 2009). However, since our study revealed no significantly different VOC removal rate constant between plant species, the differences in physiology or macroscopic and microscopic morphology seem to be not distinctive enough to have an impact on the removal of toluene and 2-ethylhexanol in these experiments. Although microscopic analyses revealed a generally thicker wax layer of *D. maculata* compared to *S. wallisii* and *A. densiflorus* leaves, the VOC removal rate constant did not differ between the species. Thus, despite the lipophilic character of both VOC, sorption appears to be primarily depending on the surface area and VOC diffusion within the cuticle or translocation to other leaf tissue seems to be low in the present study. However, it can generally be considered that with increasing lipophilicity of VOC identity, the composition and quantity of the cuticular layer has a greater impact. Depending on VOC identity and plant species/physiology (thus level and composition of enzymes), passive uptake by sorption on plant surfaces may change to active uptake by stomata in the second action phase, which is characterized by translocation, metabolism, and/or storage of VOC. According to this, other genetic and/or physiological characteristics (e.g., stomatal conductance and metabolic activity) may have an impact on VOC removal as well. Furthermore, VOC exposure may also affect plant physiology, as outlined in the following, which in return might have an impact on the VOC removal again.

Effect of VOC exposure on physiological plant characteristics

To assess the impact of VOC exposure on plant physiology, transpiration (providing general insight into the plant's metabolic activity), CO_2 fixation/respiration (reflecting photosynthetic activity and together with transpiration the plant's gas exchange with the environment), and quantum yield of PS II (as additional marker for the photosynthetic activity and as indicator for the location of possible damage within the photosynthetic apparatus) were determined upon exposure to toluene and to 2-ethylhexanol. Control plants were not exposed to either VOC.

In *D. maculata* and *A. densiflorus*, the transpiration rates were not affected by VOC exposure and showed no difference between treatments (control, exposure to toluene, and exposure to 2-ethylhexanol), neither under light (Fig. 3a) nor under dark (Fig. 3b). In detail, transpiration rates of *D. maculata* were 8.4 ± 1.0, 7.9 ± 2.3, and 9.3 ± 2.0 ml H_2O h^{-1} m^{-2} leaf area under light and 5.3 ± 0.6, 4.1 ± 1.2, and 3.9 ± 0.05 ml H_2O h^{-1} m^{-2} leaf area under dark for control, toluene, and 2-ethylhexanol treatments, respectively. *A. densiflorus* had somewhat lower transpiration rates compared to *D. maculata* with 5.7 ± 0.4, 5.6 ± 0.9, and 7.6 ± 3.0 ml H_2O h^{-1} m^{-2} leaf area under light and 1.3 ± 0.2, 1.1 ± 0.6, and 1.0 ± 0.4 ml H_2O h^{-1} m^{-2} leaf area under dark for control, toluene, and 2-ethylhexanol treatments, respectively. In contrast, *S. wallisii* showed a significantly lower transpiration rate when exposed to 2-ethylhexanol under light (11.2 ± 1.4 ml H_2O h^{-1} m^{-2} leaf area) in comparison to control (14.8 ± 1.2 ml H_2O h^{-1} m^{-2} leaf area) and toluene treatment (14.7 ± 1.8 ml H_2O h^{-1} m^{-2} leaf area) (Fig. 3a). Under light, this plant species showed in our study the highest basal transpiration rate (i.e., when not exposed to VOC). The lowered transpiration indicates a decrease in metabolic activity upon 2-ethylhexanol exposure, which might become obvious at higher basal transpiration rates only. The basal transpiration rates of the other two plant species might not have been high enough to be lowered significantly. Further, a detrimental effect on transpiration might also be attributed to the flowering stage of *S. wallisii*, because in this developmental stage, plants were found to be more susceptible to VOC exposure (Cape 2003b). The higher transpiration rate of *S. wallisii* when exposed to toluene under dark (8.0 ± 3.7 ml H_2O h^{-1} m^{-2} leaf area) compared to control (2.8 ± 0.7 ml H_2O h^{-1} m^{-2} leaf area) and 2-ethylhexanol treatment (2.2 ± 0.3 ml H_2O h^{-1} m^{-2} leaf area) could have been caused by a measurement error since the standard deviation is quite high (Fig. 3b).

Regarding CO_2, in control experiments without VOC exposure, the decrease (light) and the increase (dark) of its concentration in the chamber air was largely regular and linear during the experimental time span (Online Resource 3), indicating a constant stomatal conductivity and photosynthetic activity and a constant respiration, respectively. In accordance, also the quantum yield of control plants remained unchanged after 48 h under continuous light or dark. The CO_2 fixation and respiration rates for *D. maculata* and *S. wallisii* were not significantly different for VOC-exposed plants compared to plants from control treatment (Fig. 4). In contrast, *A. densiflorus* showed a significant decrease in CO_2 fixation after exposure to either VOC; respiration was not altered. This indicates damage to the photosynthetic apparatus of *A. densiflorus* under light. Adverse effects of the photosystem in this plant species upon VOC exposure under light became also obvious in the quantum yield, which was significantly reduced when *A. densiflorus* was exposed to either VOC under light (Fig. 5). The other two species were not affected in quantum yield except a decrease found for *D. maculata* exposed to 2-ethylhexanol under light. Thus, the photosynthetic apparatus of *A. densiflorus* seems to be more susceptible to

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VOC exposure than for the other two species. Furthermore, these data indicate that the damage took place in PS II, thus in the light reactions of photosynthesis, rather than in the carbon fixation reactions. A reduced absorption of photons in PS II might have reduced synthesis of NADPH + H^+ and ATP, which is further lacking in the Calvin cycle, what in turn might reduce CO_2 fixation. However, a lowered quantum yield does not necessarily lead to a reduced CO_2 fixation (as shown for *D. maculata*) since several competing processes (e.g., nitrogen metabolism, electron donation to oxygen, or photorespiration) might be reduced instead (Maxwell and Johnson 2000). Interestingly, a damage to the photosynthetic apparatus appeared in our study to take place only under light, thus when it is operating. Under dark, when the photosystem is inactive, no negative impact of VOC on PS II quantum yield was observed. In experiments with *A. densiflorus*, the quantum yield was even increased after 48 h dark and 2-ethylhexanol exposure. However, the reason for this is unknown.

A damaging effect on PS II was also observed in similar experiments when *Zamioculcas zamiifolia* was exposed for 7 days (12 h light/dark cycle) to high concentration of xylene (6000 ppm). However, under exposure to lower concentrations, no decrease in quantum yield was detected in that study (Sriprapat et al. 2014). Other experiments revealed no effects on quantum yield of *Z. zamiifolia* when exposed to 20 ppm of either benzene, toluene, ethylbenzene, or xylene (Sriprapat and Thiravetyan 2013). Here, the authors mentioned that this concentration might be not high enough to affect the photosynthesis of the plant. The impact of VOC concentration on phytotoxicity is also outlined by Jen et al. (1995).

The chemical properties of VOC and the accompanying routes of uptake by plants may also be of impact with regard to VOC toxicity. Generally, hydrophobic compounds like toluene and 2-ethylhexanol have a high affinity for cuticular lipids (Paterson et al. 1990). Thus, the cuticle is likely a major sink for both VOC. Usually, lipophilic compounds remain in the cuticle with little tendency to migrate to other plant parts. But a diffusion into underlying cells and tissues is also possible, depending on the chemical properties of the particular compound (Paterson et al. 1990). The one-way ANOVA showed no differences in the toluene removal rate between light (stomata open) and dark (stomata almost closed); further, correlation analysis revealed no relation between the toluene removal rate constant and the gas exchange parameters (CO_2 fixation/respiration and transpiration). Also Yoo et al. (2006) found that the removal of toluene does not appear to be rate limited by stomatal conductivity. Hence, it is assumed that the majority of removed toluene is fixed within the cuticle and not further translocated, and that stomatal uptake is negligible. Consequently, the contact between toluene and the intracellular space is likely very limited, which would reduce potential phytotoxic effects per se. Nonetheless, differences might occur between plant species as shown in the present study in case of toluene exposure of *A. densiflorus* under light. Regarding 2-ethylhexanol, even though statistical analysis revealed no correlation between the removal rate constant and the plant gas exchange parameters, it is suggested that the stomatal pathway has a certain significance for this compound due to the higher removal rates of 2-ethylhexanol under light versus dark found by one-way ANOVA. Because of the hydroxyl group, 2-ethylhexanol has a higher affinity for plant tissue and is more reactive against plant-based components than toluene (Kvesitadze et al. 2009). Therefore, it could more easily enter the intracellular space where it affects plant physiology. This would explain why *D. maculata* and *S. wallisii* showed a stronger response in the analyzed physiological parameters when exposed to 2-ethylhexanol in comparison to toluene exposure.

Briefly summarized, our experiments revealed only a minor impact of 2-ethylhexanol on the analyzed physiological parameters of *D. maculata* and *S. wallisii* and toluene seems to have no impact in these plant species. In contrast, *A. densiflorus* was affected by both VOC, but only under light. This indicates that plants of different genetic and/or physiological status can respond specifically to VOC and that phytotoxicity depends on VOC identity and light condition. Other studies support these findings. For example, a reduction in photosynthesis for *Dieffenbachia amoena* following toluene exposure was only found under high light intensities, but not under low light intensities (Porter 1994). Further, the level of deleterious effects of VOC on plants was shown to vary with plant species (Schmitz et al. 2000; Yoo et al. 2006) and VOC identity (Yoo et al. 2006). However, phytotoxic effects might be only of concern at high concentration of VOC. Typical VOC concentration found indoors have likely no short-term effects on plants (Cape 2003a; Guieysse et al. 2008), though it was shown that low VOC concentrations may affect the timing and extend of flowering and fruiting of outdoor plants (Cape 2003a). Studies concerning long-term effects of low-concentrated VOC on indoor plants are missing so far.

Conclusions

Generally, it is difficult to transfer results on VOC removal by plants from chamber experiments to real-life settings. Nevertheless, they can give an estimation about the potential efficiency of plants to improve indoor air quality. Our results indicate that aerial plant parts have no major impact on indoor air quality in regard to VOC pollution. However, further research is needed to examine the potential impact of whole potted plant systems, whereas experimental conditions should be set similar to real environments to obtain results that can be easily transferred from chambers to interiors. Overall, differences in morphological plant parameters could not be related to any significant differences in VOC removal by aerial parts

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of the three tested plant species. The same was true for the physiological parameters, although the high stomatal conductance of *S. wallisii* might have (in tendency) facilitated the uptake of 2-ethylhexanol. Overall, the uptake of toluene and 2-ethylhexanol seemed to be primarily of passive action, at least until saturation of plant surfaces. The further uptake of VOC depends strongly on VOC identity and plant species. An influence of VOC on plant physiology was observed in all plant species, but in regard to different characteristics. There was a remarkable decrease of CO_2 fixation and quantum yield in *A. densiflorus* after exposure to either VOC under light. A decrease in quantum yield was also observed for *D. maculata*, but only after exposure to 2-ethylhexanol under light. No influence on photosynthesis of *S. wallisii* was observed, but there was a decrease in transpiration. Here again, this effect was noted only under light and 2-ethylhexanol exposure. These data indicate that phytotoxicity varies with plant species and VOC identity, as was found also by Schmitz et al. (2000) and Yoo et al. (2006). Under dark, no major effects of VOC exposure on photosynthesis or transpiration were detected. Thus, light condition seems to have an impact on VOC toxicity on plants too, as is also suggested by Porter (1994). The VOC uptake pathway and the fate of VOC within the plant likely have an effect on plant physiology. However, since only three plant species were considered, the capability to identify species-related responses of VOC is limited. Further research is needed to fully understand the interaction of plant physiology and VOC regarding air purification and toxicity to plants, taking the exposure time, more plant species, VOC identities/mixtures, and their concentrations into account.

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Assessment of filtration efficiency and physiological responses of selected plant species to indoor air pollutants (toluene and 2-ethylhexanol) under chamber conditions

Environmental Science and Pollution Research

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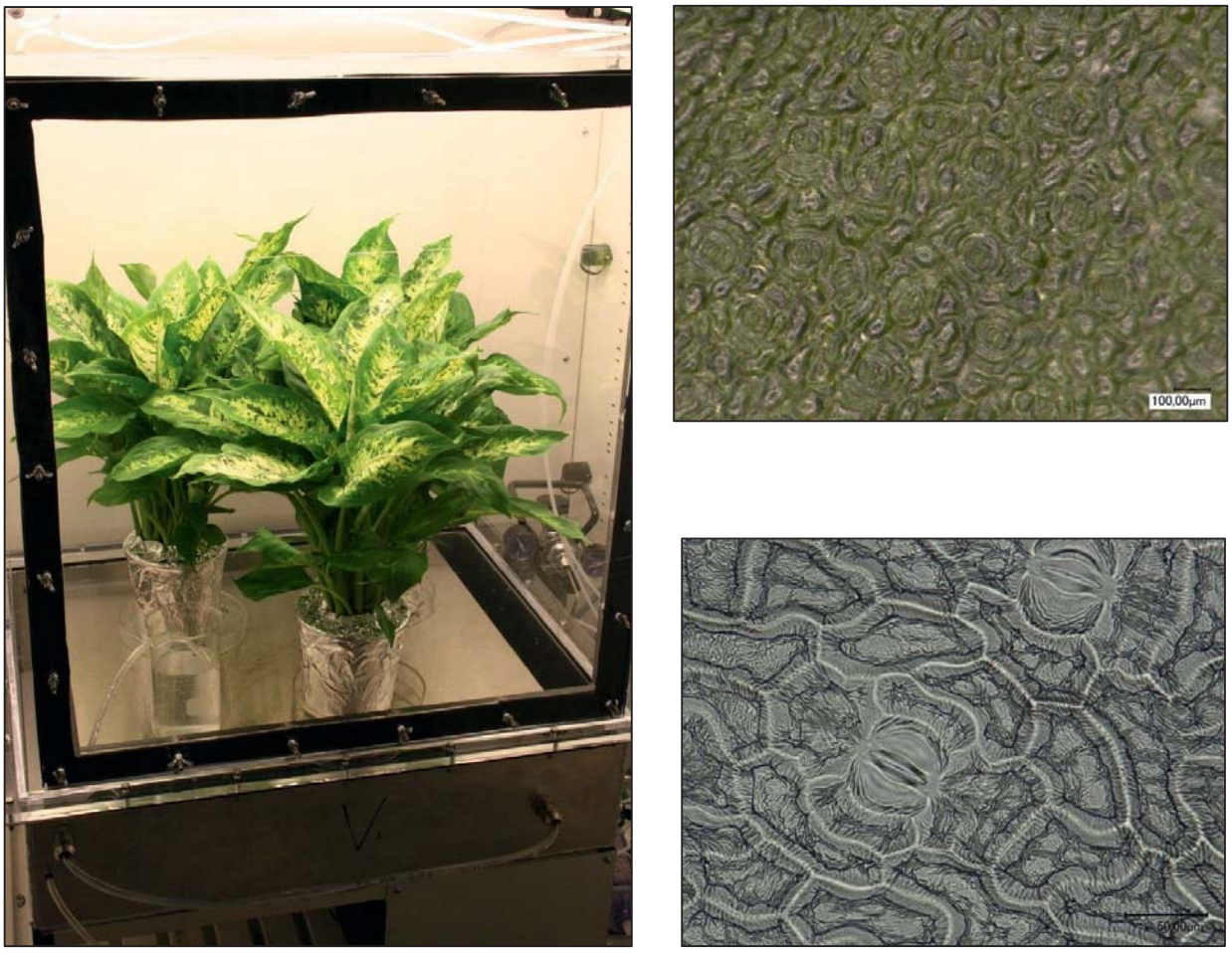
Online Resource 1 a - c: Photographic material of plants used in this study, *Dieffenbachia maculata* (a), *Spathiphyllum wallisii* (b), and *Asparagus densiflorus* (c)



Online Resource 1a: Photographic material of *Dieffenbachia maculata*: three plants during experimental run in test chamber (left), microscopic picture of abaxial leaf side (top right), and microscopic picture of lack imprint of abaxial leaf surface (down right)

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Online Resource 1b: Photographic material of *Spathiphyllum wallisii*: three plants during experimental run in test chamber (left), microscopic picture of abaxial leaf side (top right), and microscopic picture of lack imprint of abaxial leaf surface (down right)

Online Resource 1c: Photographic material of *Asparagus densiflorus*: one plant during experimental run in test chamber (left), leaves (top right), and microscopic picture of a leaf side (down right)

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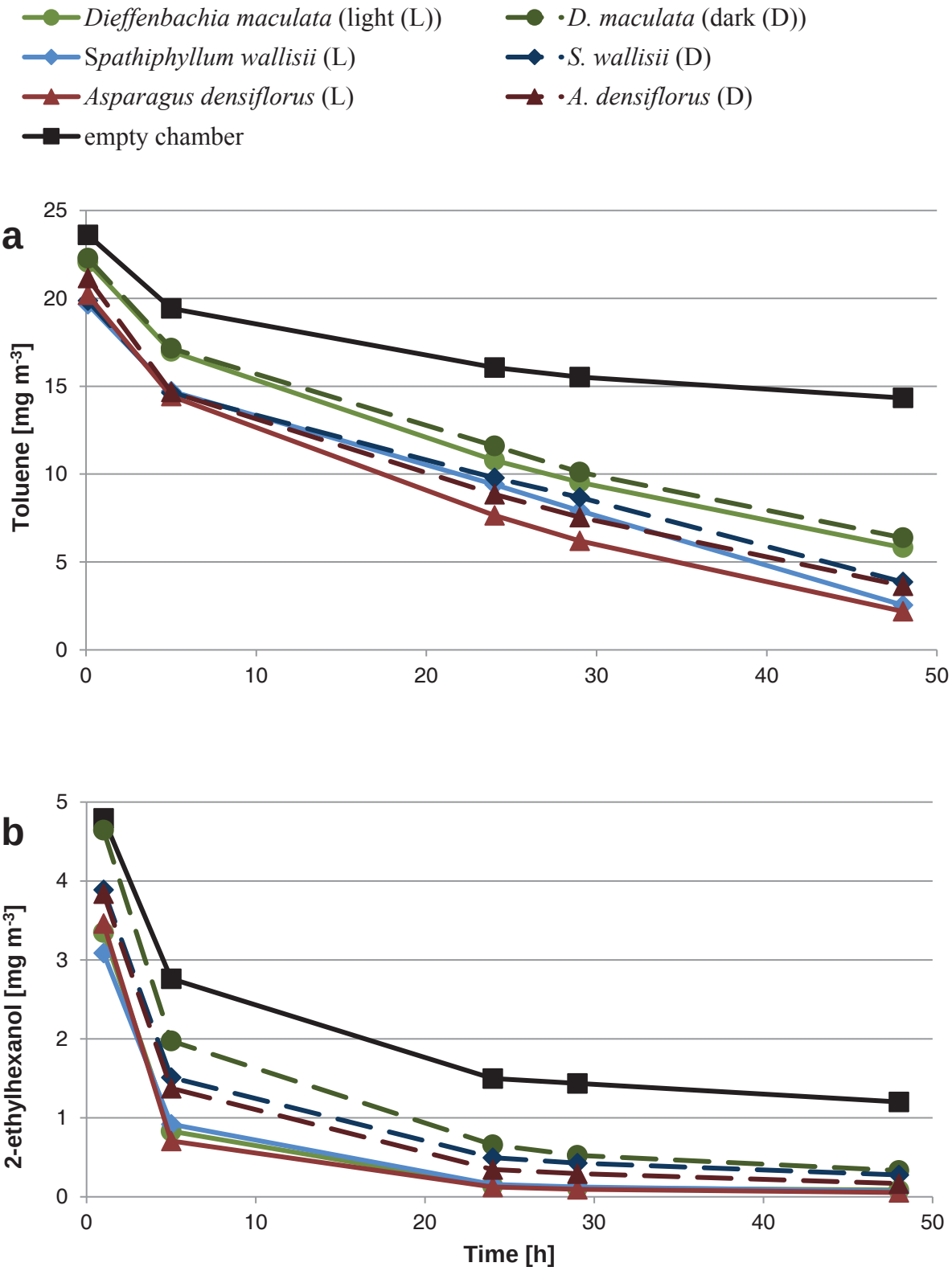
Online Resource 2: Removal curves of toluene (a) and 2-ethylhexanol (b) in chamber experiments depending on different treatments



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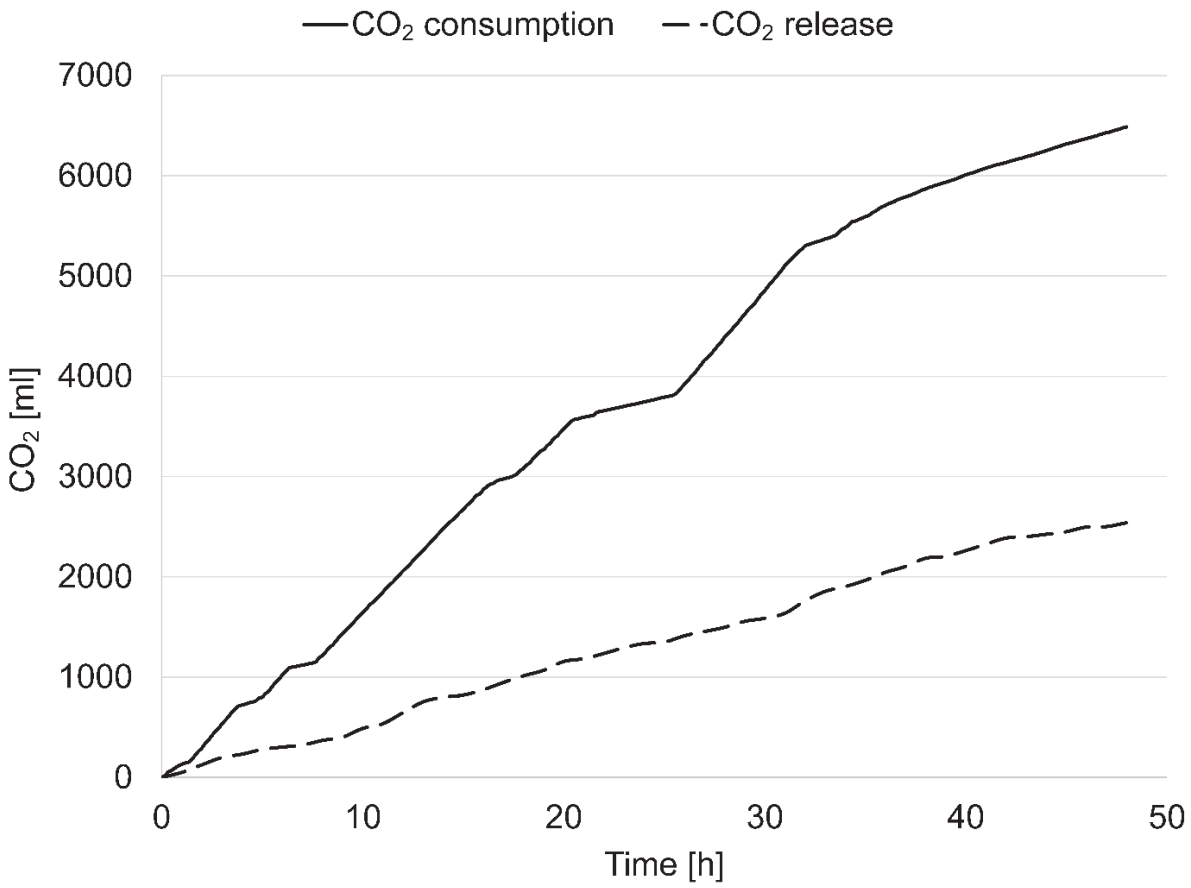
Online Resource 3: Consumed and released CO_2 [ml] during experimental run with plants in control treatment (i.e. without VOC exposure) representing photosynthetic activity and respiration, respectively



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Results on removal of volatile organic compounds compared to literature

The settings in the chamber experiments should create conditions that support plant metabolism and therefore also the uptake of VOC by plants. Nevertheless, a high amount of leaf area was necessary to trace the limited amount of VOC removed by plant foliage.

The results of this doctoral thesis revealed no differences in removal rate constants of either toluene or 2-ethylhexanol between the different plant species (*Dieffenbachia maculata*, *Spathiphyllum wallisii* and *Asparagus densiflorus*) (Hörmann et al. 2017b). While removal rate constants represent the volume of air which is purified per unit time per leaf area, the units used to report the removal of VOC by plants in literature are rather expressed as the amount or the percentage of VOC removed per unit time per leaf area (Dela Cruz et al. 2014). To allow a comparison of the results obtained in the present study with results reported in literature, the results of this doctoral thesis are presented additionally as amount of removed VOC per unit time per leaf area in table 1.

Table 1 Accumulated removal of toluene and 2-ethylhexanol (± S.E.) by *Dieffenbachia maculata*, *Spathiphyllum wallisii* and *Asparagus densiflorus* after 48 h of exposure in Biobox under continuous light and, continuous dark, respectively.

Plant species	Light condition	Removal [µg h^{-1} m^{-2} leaf area]	
		toluene	2-ethylhexanol
D. maculata	light	110 ± 24	22 ± 2
S. wallisii	light	117 ± 24	29 ± 2
A. densiflorus	light	79 ± 45	25 ± 14
D. maculata	dark	110 ± 11	18 ± 2
S. wallisii	dark	90 ± 8	23 ± 3 [A]
A. densiflorus	dark	70 ± 45	22 ± 12

Since 2-ethylhexanol was not tested yet against removal by plants, these data cannot be compared with data from the literature. The calculated values for toluene removal were found to be in between values reported in literature, which, however, show a high heterogeneity (Hörmann et al. 2017a). For instance, Orwell et al. (2006) reported a toluene removal for *S. wallisii* of round 6300 µg m^{-3} m^{-2} h^{-1} (recalculated from 32.8 mg m^{-2} d^{-1}, chamber volume: 0.216 m^3) while Yang et al. (2009) reported a far lower toluene removal of 2.52 µg m^{-3} m^{-2} h^{-1} for the same plant species. In this study *S. wallisii* removed around 490 µg m^{-3} m^{-2} h^{-1} and

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375 µg m^{-3} m^{-2} h^{-1} of toluene under light and dark, respectively. As presented in detail in Hörmann et al. (2017a), the high diversity of results is likely attributed to a diversity in experimental setups. To get a better impression about the influence of the experimental setup on VOC removal by plants, it would be interesting to repeat experiments under changing environmental conditions and with different treatments.

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Chapter III - Potential value of plants in regard to indoor air quality and human health

Removal of pollutants by plants in real-life settings

So far, many studies were conducted concerning the filter capability of plants for improving IAQ. Most of these studies were conducted in test chambers, whose results are hard to transfer to real-life settings. Studies done at real-life settings, however, revealed that plants seem to have no major impact on IAQ. In the early 90s, Healthy Buildings International (HBI) conducted one of the first field studies. Plants were installed on one of two similar floors after baseline measurements were made. The un-planted floor served as reference. Even after several months, no difference in concentration of VOC between both floors could be detected. The authors suggest that overwhelming influences, like the effect of ventilation, masked the effect of the plants (HBI, 1992). The influence of plants on the concentration of formaldehyde in portable office buildings (ca. 20 m^3) was investigated by Dingle et al. (2000). The offices were first equipped with five plants and every two days five more plants were added up to a maximum of 20. The results show only a reduction of 11% of the formaldehyde concentration in the presence of 20 plants compared to controls without plants. In the presence of 0 - 15 plants no reduction in formaldehyde concentration could be detected. A large field study in three different office buildings, two with air-conditioning systems (building 1 and building 2) and one which was naturally ventilated (building 3), was conducted by Wood et al. (2006). The offices were equipped with either 0, 3 or 6 plants. The results revealed no differences in either CO_2 concentration or in relative humidity independent of building or planting regime. Total VOC levels could only be reduced significantly in building 2 and building 3 if the level was higher than 100 ppb in the reference offices and data with total VOC < 100 ppb were excluded. The number of plants (3 or 6) had no effect on VOC reduction. Comparing the means of all data (data with total VOC < 100 ppb included) no significant differences in any building between the different planting regimes could be found (Wood et al., 2006). Song et al. (2007) conducted an investigation in newly built laboratory chambers of approximate 30 m^3 each. The more plants placed in a chamber (5 or 10% of room space), the more VOC could be reduced. Unfortunately, no information about number or size of plants or significant levels was given. Another investigation, this time in open plan offices, was done by Smith and Pitt (2011). Within six months after installation of plants no differences in relative humidity and CO_2 levels between planted and non-planted offices were found. Interestingly, the VOC levels were consistently higher in the planted area during the whole experimental period. Pegas et al. (2012) determined the IAQ in terms of concentrations of VOC, CO_2, CO, carbonyl and

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particulate matter in a school in Portugal. After placement of six potted plants in one classroom all parameters could be reduced significantly. However, no replicates were conducted and no reference room was monitored. Because the study was conducted from February to May, first three weeks without plants following six weeks with plants, variations in ventilation could have had an impact on the measured values what is quite possible, due to a higher ventilation rate in the warmer period. In summery, the data that are currently available are lacking consistency in the potential effect of plants on IAQ.

People-plant relationships

A more beneficial effect of indoor plants may be the psychological effect on human rather than their impact on IAQ. Different hypotheses, like the Biophilia hypothesis (Wilson, 1984) or the psycho-evolutionary theory (Ulrich et al., 1991), suggest that nature has positive effects on health and well-being of human. Also houseplants are thought to exhibit potential positive psychological and physical effects on human and several studies support this assumption, although here the data vary too. For example, Nieuwenhuis et al. (2014) conducted three studies in large commercial offices in the Netherlands and the UK to evaluate the effect of plants on employees. Office workers in planted offices showed an increase in subjective concentration, workplace satisfaction, and perceived IAQ. However, an improved productivity could only be detected in one of the three studies. A survey of office workers from several companies in Norway revealed an increased productivity and decreased sick leave if indoor plants are placed proximal to a worker´s desk (Bringslimark et al., 2007). Larsen et al. (1998) reported higher levels of mood, perceived office attractiveness, and perceived comfort when plants were present in the office. However, results of a productivity task showed in this study an inverse linear relationship to the number of plants per office. This may be attributed to a distraction due to the plants (Larsen et al., 1998). A survey about user perception and satisfaction in green and non-green office buildings was conducted in China. Office workers from green buildings were found to be more satisfied but felt not necessarily more comfortable (Gou et al., 2013). Another study concerning the influence of indoor plants on health and discomfort of office workers was conducted by Fjeld et al. (1998). In offices containing plants, neuropsychological symptoms (including fatigue and feeling heavy headed), mucous membrane symptoms (e.g. hoarse, dry throat, cough), and skin symptoms (dry or flushed facial skin) could be reduced by 21%.

Beside beneficial effects of plants on office workers, investigations in nursing homes and hospitals revealed that plants can act as therapeutic tools too. Thus, older people showed an

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improvement in life satisfaction and social network as well as a decrease in perception of loneliness after an indoor gardening program (Tse, 2010). A survey among 65 nursing staff members from ten homes revealed a prominent contribution of plants to the psychological and social well-being of residents with dementia (Rappe and Lindén, 2004). Studies of Park and Mattson (2009, 2008) have shown that patients in hospital rooms with plants had shorter hospitalization, fewer intakes of analgesics, lower ratings of pain, anxiety, and fatigue, and more positive feelings after surgery compared with patients in the control group. Similar effects could be recorded if patients were assigned to rooms with windows looking out on a natural scene than patients whose windows faced a brick building (Ulrich, 1984). Different laboratory studies have already shown that the view on pictures showing landscapes can lead to positive psychological and physiological effects. For example, less stress (Dijkstra et al., 2008) or quicker recovery from stress and greater immunization to subsequent stress (Parsons et al., 1998) as well as a decreased blood pressure, heart rate, and fingertip pulse (Li et al., 2012) could be demonstrated.

Although there are many studies published dealing with the potential impact of plants on either IAQ or human health, there are only few studies available that tested both issues simultaneously. Lohr and Pearson-Mims (1996) evaluated the accumulation of particulate matter on horizontal surfaces in presence and absence of plants. The tests were performed in a windowless computer laboratory as well as in an office space. In both rooms the accumulation of particulate matter was significantly lower (up to 20%) in the presence of plants. While the temperature remained unchanged, the relative humidity slightly increased from 41.2 to 42.0% (Lohr and Pearson-Mims, 1996). Furthermore, the influence on productivity and stress of workers in the presence or absence of plants in the computer laboratory that lacked any windows was examined. The results revealed that the productivity was increased by 12%, the stress level decreased, and attentiveness was higher in the presence of plants (Lohr et al., 1996). Lim et al. (2009) investigated the influence of plants in newly built apartments on their capacity to remove certain VOC and their influence on symptoms of sick building syndrome (SBS). Only the formaldehyde content could be reduced in the households with plants. The content of other examined VOC (toluene, ethylbenzene, and xylene) was not reduced; in some cases they even increased in households with plants, especially in the winter season. The authors mentioned that this effect is the consequence of reduced ventilation by indoor-dwellers, done to protect plants from cold injury. Nevertheless, the frequency of SBS symptoms was significantly lower in households with plants at least in the summer season (Lim et al., 2009). Similar results were obtained by Kim et al. (2011). Here, the IAQ was

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examined in terms of benzene, toluene, ethylbenzene, xylene (all together abbreviated as BTEX), and formaldehyde concentration. Interiors used were offices in aged buildings as well as in newly-built buildings. Again, only the formaldehyde concentration could be reduced by the plants, and only in the aged buildings. The concentrations of the other investigated VOC were mostly higher in the presence of plants. In accordance with a lowered formaldehyde concentration, symptoms of SBS were reduced in offices with plants only in the aged buildings (Kim et al., 2011). In another study, the IAQ in schools in Seoul was monitored in four rooms with plants and in two rooms without plants (Kim et al., 2013). More than twice as many plants were installed compared to the study conducted by the aforementioned Pegas et al. (2012), which showed significant reduction of VOC, particulate matter, and other air pollutants. However, here the amount of measured VOC (formaldehyde and BTEX) was not markedly affected by the plants. Although the symptom score of SBS showed a constant decrease in classrooms with plants but not in control classrooms, a multiple regression analysis revealed that this had only little relation to the indoor plants (Kim et al., 2013).

In general, contrary to reports from studies with chamber experiments, a moderate planting does not seem to improve IAQ markedly. Nevertheless, positive effects of plants on human well-being have frequently been reported. Therefore, the last chapter of this doctoral thesis covered the following issues:

- Does *Spathiphyllum wallisii* improve IAQ in real-life settings?
- Have indoor plants a positive effect on human well-being and is this attributed to a placebo effect?
- Has nature relatedness of human beings an impact on the effect of plants?

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Human well-being through plants – a placebo effect?

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Highlights

- A field study was conducted to evaluate if psychological and physiological effects of indoor plants on office workers are related to a placebo effect
- Effects of plants on well-being, perceived air quality, subjective, and objective chronic stress could not be detected
- Effects of plants on humans in a working environment seem to be small and affected by multiple causes

† both authors contributed equally

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Abstract

A field study was conducted to evaluate if psychological and physiological effects of indoor plants on humans in working environments are based on a placebo effect. Furthermore, it was examined if self-reported nature relatedness of human beings affects the influence of the plant. The study was conducted with three experimental groups, each with three experimental repetitions to form a 3x3 experimental design. In order to prove the existence of a placebo effect, a sample of 74 university employees were randomly chosen and divided into two experimental groups and one control group. Each experimental group received a single indoor plant, with the outstanding difference that one experimental group was provided with an information sheet about a good pollutant removal efficiency of the plant. This information was supposed to raise expectations about the plant's impact to increase the air quality, which might enhance subjective well-being and perceived air quality. Furthermore, providing information might led to a more pronounced reduction of subjective and objective chronic stress (the latter was measured via a hair cortisol analyses). The results revealed no significant differences between the three sample groups and confirm the heterogeneous findings of former studies on the impact of plants on human well-being. The outcome of our study implies that possible effects of indoor plants on humans in working environments are subject to a more complex system of varying factors.

Keywords: People-plant interactions, stress level, human well-being, indoor plant, indoor air quality

1. Introduction

Several hypotheses, including the *biophila hypothesis*, the *psycho-evolutionary theory*, and the *attention restoration theory*, postulate that plants have positive effects on human health and well-being (Kaplan, 1995; Ulrich et al., 1991; Wilson, 1984). In addition, indoor plants are thought to have positive psychological and physiological effects on humans. They can reduce symptoms that are related to the sick building syndrome (SBS) like irritation of the respiratory system, cough, fatigue, and dry or flushed facial skin (Fjeld & Bonnevie, 2002; Fjeld, Veiersted, Sandvik, Riise, & Levy, 1998). Plants have also been shown to increase

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concentration and well-being (Nieuwenhuis, Knight, Postmes, & Haslam, 2014), reduce stress (Dijkstra, Pieterse, & Pruyn, 2008), and improve working productivity (Lohr, Pearson-Mims, & Goodwin, 1996). Furthermore, they might improve indoor air quality (IAQ) in terms of relative humidity (RH) and pollution (Tarran, Torpy, & Burchett, 2007; Wolverton, Johnson, & Bounds, 1989). However, the findings concerning the psychological and physiological effects of plants on humans are very diverse, due to different test designs, methods and manipulations used (Bringslimark, Hartig, & Patil, 2009). The same is true for the impact of plants to improve IAQ. Many chamber experiments have shown that plants remove air pollutants, e.g. volatile organic compounds (VOC) (Dela Cruz, Christensen, Thomsen, & Müller, 2014; Hörmann, Brenske, & Ulrichs, 2017). VOC belong to the most important pollutants indoors, because they are emitted from various indoor sources, including building material, furniture, technical equipment, and human activities (Berrios, Zhang, Guo, Smith, & Zhang, 2005; Que, Wang, Li, & Furuno, 2013; Rösch, Kohajda, Röder, von Bergen, & Schlink, 2014) and are associated with several health complaints like symptoms of the SBS (Burge, 2004), and eye and airway irritation (Wolkoff & Nielsen, 2001). Furthermore, several VOC (e.g. acetaldehyde, formaldehyde and benzene) are characterized as carcinogenic contaminants (Sarigiannis, Karakitsios, Gotti, Liakos, & Katsoyiannis, 2011). However, results of chamber experiments are difficult to transfer to real-life settings because environmental conditions, which may have a crucial impact on results, are quite different in test chamber versus interiors (Hörmann et al., 2017). Therefore, field studies are more informative in regard to the actual impact of plants on IAQ. Most field studies examined the impact of plants on IAQ in terms of total VOC (TVOC) and / or several VOC in detail. Indeed, it is important to examine specific VOC in field studies, since values of TVOC alone are not suited for a health related evaluation of indoor air due to the variability in composition (Jones, 1999; Seifert, 1999). However, the results of field studies have shown that the level of TVOC as well as the concentration of benzene, toluene, ethylbenzene, and xylene (BTEX) and others is often unaffected or even higher in the presence of plants (HBI, 1992; Kim et al., 2011, 2013; Lim et al., 2009; Smith & Pitt, 2011). Interestingly, studies that investigated the influence of plants on IAQ in terms of formaldehyde and BTEX and human well-being simultaneously have shown that symptoms of SBS could be significantly reduced although IAQ parameters were hardly affected by plants (Kim et al., 2011; Lim et al., 2009).

A placebo is a sham substance or sham intervention inducing positive effects without active ingredients. Two distinctive neurophysiological mechanisms are of concern in the placebo

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response: associative learning processes and the expectation of subjects towards the benefit of a treatment (Enck, Benedetti, & Schedlowski, 2008). Even though it is stated in the Cochrane report that placebo interventions have no major health effects in general, except a modest effect on pain and nausea (Hróbjartsson & Gøtzsche, 2010). A reduction of SBS in the presence of plants was found although objective factors of IAQ were not improved (Kim et al., 2011; Lim et al., 2009). It is generally unlikely that potted plants improve IAQ to a large extent (Girman, Phillips, & Levin, 2009; Soreanu, Dixon, & Darlington, 2013) and the reported positive effects of plants on humans, e.g. an increased subjective well-being (Evensen, Raanaas, Hagerhall, Johansson, & Patil, 2013; Nieuwenhuis et al., 2014), reduced self-reported symptoms of stress (Lohr et al., 1996), and perceived fresher air in a room (Lohr & Pearson-Mims, 2000), are properly not attributed to objective factors, but maybe to a placebo induced process which might be based on former / evolutionary conditioning as well as expectation.

The aim of this study was to evaluate if reported psychological and physiological effects of indoor plants on humans are attributed to a placebo effect. It was expected that one plant in the working environment has positive effects on several subjective and objective parameters of participants, but not on parameters of IAQ.

Based on the above mentioned field studies, which found no major improvement of IAQ by plants, the first hypothesis of the current study was:

H1: One plant in the working environment has no effect on IAQ,

H1.1: in terms of TVOC concentration, temperature and RH

H1.2: in terms of several VOC in detail

As pointed out, reported benefits of plants on humans in the working environment seem to be not attributed to objective factors. An explanation for this could be a placebo-induced relaxation. Since the placebo may be induced by expectation, stronger effects were assumed if participants are provided with an information sheet about a good pollutant removal efficiency of the plant (placebo manipulation). Therefore, the following hypotheses were stated:

H2: Plants in the working environment improve the subjective well-being.

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H2.1: An additional information about a good pollutant removal efficiency of plants leads to a stronger increase in subjective well-being in comparison to plant installation without additional information.

H3: Perceived air quality is rated more positively after plant installation.

H3.1: An additional information about a good pollutant removal efficiency of plants leads to an increase in perceived air quality in comparison to plant installation without additional information.

H4: The level of chronic stress decreases after plant installation.

H4.1: An additional information about a good pollutant removal efficiency of plants leads to a stronger reduction of subjectively perceived chronic stress.

H4.2: An additional information about a good pollutant removal efficiency of plants leads to a stronger reduction of objective chronic stress (hair cortisol).

In addition, the assumed impact of plants may also depend on the individual's nature relatedness, which comprises the notion of being connected with nature (Schultz, 2002). Thus, it was further hypothesized:

H5: The impact of plants on subjective well-being, perceived air quality, and subjective, as well as objective chronic stress by plants depends on individual's nature relatedness. The higher the nature relatedness,

H5.1: - the higher the effect on subjective well-being.

H5.2: - the higher the effect on perceived air quality.

H5.3: - the higher the reduction of subjective chronic stress.

H5.4: - the higher the reduction of objective chronic stress.

2. Methods

2.1 Participants

The participants of this study were composed of 74 healthy employees of the Department of Mathematics and the Department of Computer Science at Humboldt-Universität zu Berlin.

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Among the employees were 16 women with an average age of 39 ± 15.5 years and 58 men with an average age of 34 ± 15.8 years. The majority of participants were scientific assistants and professors (74% and 11%, respectively). This sample ensured nearly constant conditions in regard to the level of education and socio-economic status and to location factors like conditions of light, temperature, office design, etc.

Participants were divided into three groups: Group A got one plant with a written care instruction ("plant group", n=29, average age of 32.38 ± 7.51 years, 5 women and 24 men); group B has been provided with one plant including the care instruction plus a detailed information sheet about a good pollutant removal efficiency of the given plant ("plant+info group", n=24, average age of 36.63 ± 12.43 years, 2 women and 22 men); and group C got no plant ("control group", n=21, average age of 33.62 ± 13.54 years, 9 women and 12 men).

Both groups with plants were considered as placebo groups, whereas the information sheet in the plant+info group should create an additional expectation in regard to indoor air improvement by the plant (placebo manipulation). All groups were compared for the following demographic variables which might affect results: gender, age, children, educational level, smoking, cups of coffee per day, cups of tea per day, glasses of caffeinated drinks per day, health, disease symptoms, allergies, work resources, daily break time, and weekly working time. The analyses indicated no differences for these variables, except gender. Significantly more women were found in the control group compared to both groups with plants $F(2,71) = 4.56$, $p = .014$, $\eta 2 = .114$. Therefore, gender was considered as control variable in hierarchic regression analysis.

The participants joined the study voluntary, at no charge, were informed beforehand about the anonymity and confidentiality of individual responses, and gave written informed consent to take part in the study. A personal code was generated by each participant to anonymize the hair samples for cortisol analysis. The study followed the rules stated in the Declaration of Helsinki (1964).

2.2. Materials and Design

The plant selected for this study was *Spathiphyllum wallisii* 'Alfetta' with a height of 95 cm (Fig. 1), purchased from a wholesale in a retail-ready state in 25 cm diameter plastic pots.

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This plant species was chosen because it is a common indoor plant, widely used, and easy to handle. Furthermore, this species has been associated with air purifying effects (Orwell, Wood, Burchett, Tarran, & Torpy, 2006; Yang, Son, & Kays, 2009; Yoo, Kwon, Son, & Kays, 2006). Each office was supplied with either one plant in field of view of the employee(s) or no plant.

Fig. 1 *Spathiphyllum wallisii* in the office setting

2.2.1 Monitoring of subjective parameters

The subjective parameters, namely well-being, perceived air quality, and subjective stress level were monitored by online surveys (in German language). Well-being was assessed using the WHO-5, version II (Bech, 2004), which is one of the most widely used questionnaires assessing subjective psychological well-being (Topp, Østergaard, Søndergaard, & Bech, 2015). General health was assessed with the appropriate item derived from the German version of the SF-36 health survey (Bullinger & Kirchberger, 1998). This item was shown to

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be a good predictor of physical morbidity and mortality (Idler & Benyamini, 1997). Demographic and work-specific questions, examining possible stressors and perceived air quality, were assessed with an international widely used questionnaire for office workers in regard to sick building syndrome (MM040 Örebro) (Andersson & Stridh, 1992). The "Trierer Inventar zum chronischen Stress" (TICS screening scala) was used for evaluating the subjective perceived chronic stress in a global and unspecific manner (Schulz, Schlotz, & Becker, 2004).

Nature relatedness was assessed using the short version of nature relatedness scale (NR-6) (Nisbet, Zelenski, & Murphy, 2009; Nisbet & Zelenski, 2013). This scale predicts briefly happiness, environmental concern, and nature contact and enables the assessment of the connectedness of elements rather than environmental attitudes (Nisbet & Zelenski, 2013). Because this instrument was not used in the German-speaking area so far, it was translated from English to German by an English teacher and back again by a native English speaker. Small discrepancies from the original version were discussed and the German version revised accordingly. To avoid any impact on results of hypothesis 5 (nature relatedness as moderator), the survey of NR-6 was conducted at the end of the study. Additionally at the last sample time, participants in the groups with plants were asked if indoor climate changed after plant placement with a simple yes/no question and how they rate their knowledge about the benefit of plants for human health using a five-point rating scale (1 = "poor", 5 = "excellent") .

A follow-up survey was conducted in February 2016 to verify if the induction of an additional expectation in the plant+info group was successful (placebo manipulation). Therefore, participants in this group were asked if they read the information sheet and how credible it was perceived. Both questions were measured by using a five-point rating scale (1 = "not at all", 5 = "very detailed" and "very credible", respectively). In order to verify that participants did not influence each other, the total sample (N = 57) was asked with which intensity they exchanged with their colleagues about the study (1 = "not at all", 5 = "very intensive"). The total sample was further asked to what extent they perceive indoor plants as part of living nature, assessing the influence of nature relatedness on the perception of indoor plants. Participants were asked if they assess a plant in the office positively. Both questions were assessed using a four-point rating scale (1 = "disagree", 4 = "agree").

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2.2.2 Monitoring of Objective parameters

The level of cortisol in human hair is a good marker to assess physiologically chronic stress, because of its retrospective character (Honold, Lakes, Beyer, & van der Meer, 2015; Wosu, Valdimarsdóttir, Shields, Williams, & Williams, 2013). In comparison to salivary cortisol it has the advantage that it is not dependent on moods or daily form (Kudielka & Kirschbaum, 2003). Therefore, we chose this indicator to assess objective chronic stress. Assuming an hair growth of 1 cm per month, the cortisol secretion can be determined over a period of several months (Russell, Koren, Rieder, & Van Uum, 2012). An independent laboratory conducted the analysis using 2 cm segments of the most recent hair, representing the period of 2 months prior to sample time.

Examined parameters for assessing IAQ were the concentration of TVOC and several VOC in detail, temperature, and RH. The measurement of TVOC is a usual method for evaluating IAQ. It can be used as measure for ventilation of a building as well as for the identification of highly polluting activities (European Collaborative Action (ECA), 1997; Tham, 2016) or rather for indication of air purifying effects by the plant in our study. Also, specific VOC were quantified for profiling the main VOC in indoor air and for evaluating VOC that are of special concern for human health like BTEX (Jones, 1999; Tham, 2016). Temperature and RH were measured as additional markers for IAQ.

2.3. Procedure

The field study was conducted from 26th of October till the 17th of December 2015. The offices of participants are located either on the second, third, or fourth floor of Johann von Neumann-Haus in Berlin, Adlershof. The whole complex is comb-shaped and consists of four separate but identical rear buildings (house 1 – 4) which are connected via a large construction block. Offices have a size of approximately 20-30 m² each and were of height 2.7 m. The employees worked either in a single-man office, in a two-man office, or in a three-man office. In addition to common office furnishing, all offices were equipped with carpets and heat-insulated windows. While arranging the groups, care was taken to avoid an exchange between both groups with plants. This was done by selecting participants for the plant group only from employees of the Department of Mathematics (house 3 and 4) and participants for the plant+info group only from employees of the Department of Computer Science (house 1

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and 2). Within each department, participants were assigned randomly for the respective group. The control group was spread throughout the houses 1 to 4.
All participants were informed that the approaching field study is associated to laboratory studies in test chambers concerning the filtration of indoor pollutants by indoor plants, done at Thaer-Institute of Agricultural and Horticultural Sciences, Division Urban Plant Ecophysiology at Humboldt-Universität zu Berlin. It was communicated that two conditions will be applied: offices with a plant versus offices without a plant.
Online surveys were conducted at three sample times: at the beginning of the study, i.e. one day before plant placement, comprising the baseline (T0), in the middle of the time span, i.e. 4 weeks after plant placement (T1), and at the end of the experimental duration, i.e. 8 weeks after plant placement (T2). The hair samples were taken at T0 and at T2. Therefore, strands of hair with a diameter of ca. 3 mm were separated approximately 2 cm below the cranial bone. The hair sample was cut as close as possible to the scalp and stored in aluminum foil until analysis. Due to very short hair of some persons, hair samples could not be taken among all participants. In the baseline measurement hair samples of 66 participants were available and at T2 hair samples of 59 participants were available.

To provide similar conditions for analyses of IAQ, 19 similar two-man offices with about 20 m² were selected. The IAQ in terms of VOC concentration was monitored in 12 offices with a plant, and 7 without plant. Air samples for VOC analyses were taken passively using sorption tubes (C1-CXXX-5003, Tenax TA. C6-C30. Inert-coated., Co. Markes, Frankfurt, Germany), which were spiked with internal standards (100 ng of each cyclooctane and cyclododecane). The tubes were equipped with diffusion caps and fixed upside down at a height of approximately 2.5 m in the middle of the room. After one week of exposure (according to 3 l of sample volume), the tubes were removed and stored in air tight aluminum bags at room temperature until analysis. Prior to trace gas analysis, the sorption tubes were desorbed using a Perkin Elmer Turbomatrix thermal desorber. Following on, air samples were analyzed using an Agilent 6890 gas chromatograph (Agilent Technologies, Inc., Santa Clara, USA) with flame ionization detector (FID), coupled to a mass spectrometer (MS) (Agilent Technologies, Inc., 5979 MSD). Helium was used as carrier gas and analysis were performed in accordance with DIN ISO 16000-6. The VOC monitoring was conducted at the same sample times like the surveys (T0, T1 and T2) whereas one week of exposing sorption tubes ended at the date of survey. Due to the limited number of data loggers (EL-USB-2+, Lascar

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Electronics Ltd., Wiltshire, United Kingdom), climate conditions namely temperature and RH, could only be measured in 14 of the selected offices: 8 offices equipped with a plant and 6 offices without plant.

2.4 Data analyses

The statistical analysis of subjective and objective parameters comprising well-being of participants was conducted in three steps:

1) The three groups were examined regarding significant differences in mean values compared to the baseline using one-way ANOVA. Nominal data were analyzed with the Chi-square test in regard to different frequency distributions between groups.
2) Hierarchical regression analysis was applied for single dependent variables. The plant group served as reference group, because of anticipated expectations in plant+info group. Furthermore, nature relatedness was considered as z-standardized variable to examine its impact on effects.
3) Because the first step revealed that gender is significantly different between groups, this parameter was encoded and taken into the model as control variable. Thereby it was verified that results of former steps remain valid.

The analysis of the follow-up survey was conducted by differences in mean values using one-way ANOVA. Following, Pearson´s correlations between dependent variables and “sorption tube for monitoring IAQ in office” were examined in the total sample to verify a possible relation between variables and randomly assigned sorption tubes.

Data on IAQ were statistically analyzed by one-way ANOVA, examining potential differences in temperature, RH, and concentration of VOC between offices with and without plant. For temperature and RH, mean values of the whole time-span were analyzed; for TVOC and single VOC, raw data per each of the sample times were used. For determining differences of VOC concentration over time, a post-hoc Tukey´s HSD was applied for TVOC and single VOC within planted and non-planted offices.

All data were tested with a level of significance of $p < 0.05$ using the statistical software package SPSS 23.0.0.0 (IBM).

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3. Results

3.1 General descriptive statistics

Analysis of dependent variables, i.e. subjective well-being, perceived air quality, subjective and objective chronic stress (hair cortisol) revealed no differences between groups in the baseline. The same was true for the self-reported nature relatedness and the knowledge about the benefits of plants for human health, which were surveyed at T2 (Table 1).

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Table 1 Descriptive statistics of participants assigned to experimental groups

	Plant group			Plant+info group			Control group				
	n	*M*	*SD*	*n*	*M*	*SD*	*n*	*M*	*SD*	F/χ^2	*p*
Dependent variables (T0)											
Subjective well-being	29	2.86	1.02	24	2.87	.95	21	2.87	.84	$F(2,71) = 0.00$	1.000
Perceived air quality	29	1.34	.34	24	1.54	.50	21	1.37	.44	$F(2,71) = 1.70$	.190
Subjective chronic stress	29	1.18	.70	24	.85	.44	21	1.08	.49	$F(2,71) = 2.28$	.110
Hair cortisol	24	13.86	16.92	22	23.14	27.52	17	35.54	59.29	$F(2,60) = 1.78$	.177
Independent variable (T2)											
Self-reported nature relatedness	28	3.17	.90	24	3.40	.89	21	3.29	.86	$F(2,70) = 0.12$	.886
Self-rated knowledge about benefits of plants	28	3.79	.88	24	3.79	1.10	21	3.90	.70	$F(2,70) = 0.44$	.646

Note: T0 = Baseline; T2 = 8 weeks after plant placement (last sample time); *n* = sample size; *M* = mean; *SD* = standard deviation; F = test value one-way ANOVA; χ^2 = χ^2-indipendence test; * $p < .05$

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3.2 Indoor air quality in planted and non-planted offices

The temperature and RH were comfortable and remained steady during the test period in planted and non-planted offices. Confirming our hypothesis (No. 1.1), no significant differences were found for temperature: 22.4 ± 0.7°C and 22.3 ± 0.4°C, as well as for RH: 40.9 ± 1.7% and 40.6 ± 1.6% in planted (n=8) and non-planted offices (n=6), respectively.

The results of TVOC and single VOC analyses are presented in Table 2. The concentration of TVOC and singly examined VOC showed no significant differences according to planting regime, except for toluene, mp-xylene, and ethylbenzene which were significant lower in planted offices. Thus, our hypothesis concerning IAQ in regard to VOC (H1: 1-2) is largely confirmed. Significant changes of specific VOC over time were found, but followed no clear pattern. For instance, while the concentration of a-pinene decreased from T0 to T1 significantly and further remained unchanged, the concentration of toluene first decreased (T0 to T1) and later increased again (T1 to T2). The variation in VOC concentration were largely similar in planted and non-planted offices, except TVOC, 2-ethylhexanol, and o-xylene. The level of TVOC remained unchanged in planted offices throughout the study, while an increase was observed in non-planted offices. The concentration of o-xylene in planted offices decreased from T0 to T1, ending in between both values at T2, in contrast non-planted offices showed unchanged concentrations of o-xylene at all sample times. Contrary to this, 2-ethylhexanol decreased significantly in non-planted offices but remained unchanged in planted offices.

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Table 2 Concentrations of total volatile organic compounds (VOC), toluene, 2-ethylhexanol, ethylbenzene, mp-xylene, o-xylene, styrene, a-pinene, and hexanal in offices with plant (n=12) and without plant (n=7). Data are presented as means ± S.D., different letters indicate significant differences between sample times ($p < 0.05$, one-way ANOVA, followed by Tukey's HSD test), indications for significant differences between planting regime within sample times are given below specific data sets ($p < 0.05$, one-way ANOVA)

VOC	Planting regime	VOC concentration [µg m^{-3}]		
		T0	T1	T2
total VOC	+ plant	101.42 ± 39.03^{a}	109.83 ± 33.78^{a}	117.17 ± 43.33^{a}
	- plant	65.54 ± 23.13^{a}	91.00 ± 17.02^{ab}	104.86 ± 35.59^{b}
	Significance	n.s.	n.s.	n.s.
toluene	+ plant	2.47 ± 0.39^{a}	1.45 ± 0.32^{b}	1.96 ± 0.41^{c}
	- plant	2.72 ± 0.49^{a}	1.87 ± 0.51^{b}	2.32 ± 0.34^{ab}
	Significance	n.s.	*	*
2-ethylhexanol	+ plant	4.13 ± 3.09^{a}	3.57 ± 1.87^{a}	2.71 ± 1.44^{a}
	- plant	3.10 ± 0.75^{a}	2.58 ± 0.72^{ab}	2.27 ± 0.41^{b}
	Significance	n.s.	n.s.	n.s.
ethylbenzene	+ plant	0.84 ± 0.19^{a}	0.50 ± 0.09^{b}	0.61 ± 0.11^{c}
	- plant	0.95 ± 0.12^{a}	0.68 ± 0.20^{b}	0.82 ± 0.12^{ab}
	Significance	n.s.	*	*
mp-xylene	+ plant	1.66 ± 0.45^{a}	0.92 ± 0.21^{b}	1.22 ± 0.23^{c}
	- plant	1.72 ± 0.45^{a}	1.15 ± 0.43^{b}	1.46 ± 0.29^{ab}
	Significance	n.s.	n.s.	*
o-xylene	+ plant	0.70 ± 0.24^{a}	0.46 ± 0.14^{b}	0.60 ± 0.14^{ab}
	- plant	0.75 ± 0.29^{a}	0.58 ± 0.28^{a}	0.70 ± 0.21^{a}
	Significance	n.s.	n.s.	n.s.
styrene	+ plant	0.93 ± 0.34^{a}	0.78 ± 0.29^{a}	0.83 ± 0.21^{a}
	- plant	0.91 ± 0.19^{a}	0.72 ± 0.21^{a}	0.88 ± 0.21^{a}
	Significance	n.s.	n.s.	n.s.
a-pinene	+ plant	3.59 ± 0.81^{a}	2.24 ± 0.38^{b}	2.47 ± 0.33^{b}
	- plant	3.22 ± 0.30^{a}	1.99 ± 0.38^{b}	2.35 ± 0.37^{b}
	Significance	n.s.	n.s.	n.s.
hexanal	+ plant	3.22 ± 1.59^{a}	3.03 ± 0.98^{a}	2.66 ± 1.26^{a}
	- plant	2.88 ± 0.72^{a}	2.49 ± 1.06^{a}	2.44 ± 0.99^{a}
	Significance	n.s.	n.s.	n.s.

Note: T0 = Baseline; T1 = 4 weeks after plant placement; T2 = 8 weeks after plant placement; + plant = offices with plant; - plant = offices without plant; n.s. = not significant; * = significant

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3.3. Effects of plants on subjective and objective parameters of human well-being

The results of the online survey concerning subjective well-being, perceived air quality, and subjective chronic stress, as well as hair cortisol analyses are given in Table 3. The hierarchical regression analysis regarding a possible placebo effect with respect to subjective parameters revealed no significant changes from baseline (T0) to T1, as well as from T1 to T2. Therefore, only results from T0 to T2 are further specified. The plant group served as reference for effects of plants on dependent variables and the plant+info group and control group were compared to reference using regression weights.

Contradictory to our hypothesis (H2), the analysis revealed no improvement of subjective well-being in the plant group (p = .41). By comparison, the subjective well-being is not improved more in the plant+info group (β = -.02, t(70) = -.17, p = .87), and not less improved in the control group (β = -.05, t(70) = -.07, p = .73).

The perceived air quality did not change in the plant group as well (p = .43). By comparison, the perceived air quality did not improve stronger in the plant+info group (β = -.08, t(70) = -.62., p = .54) and the control group showed no minor improvement (β = .08, t(70) = .61, p = .54). The additional question at T2, which queried if indoor climate changed after plant placement, was answered by a narrow majority with “no” equally in both groups with plants (χ^2 = 0.002, p = .966). This result again fails to confirm our hypothesis (H3).

A reduction of subjective chronic stress was found in the plant group (p = .03). However, contradictory to our hypothesis (H4.1) the plant+info group showed no stronger reduction in subjective chronic stress (β = .24, t(70) = 1.86, p = .07) and the control group showed no minor reduction (β = .22, t(70) = 1.72, p = .09).

Contrary to our hypothesis (H4.2) the level of hair cortisol, representing objective chronic stress, remained unchanged from T0 to T2 in the plant group (p = .34). By comparison, the reduction of objective chronic stress was not stronger in the plant+info group (β = -.10, t(57) = -.66, p = .51) and not smaller in the control group (β = -.02, t(57) = -.12, p = .91).

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Table 3 Mean values and standard deviation of dependent variables at baseline, 4 weeks and 8 weeks after plant placement

Experimental group	Sample time	Well-being			Perceived air quality			Subjective chronic stress			Objective chronic stress		
		n	*M*	*SD*	*n*	*M*	*SD*	*n*	*M*	*SD*	*n*	*M*	*SD*
Plant group	T0	29	2.86	1.02	29	1.34	.34	29	1.18	.70	24	13.86	16.92
	T1	28	2.81	1.02	28	1.29	.32	28	1.06	.76			
	T2	26	2.98	.70	28	1.29	.35	26	.93	.79	26	9.76	6.04
Plant+info group	T0	24	2.87	.95	24	1.54	.50	24	.85	.44	22	23.14	27.52
	T1	24	3.01	1.01	23	1.42	.30	23	.85	.58			
	T2	24	2.94	1.01	24	1.41	.41	24	.90	.65	22	15.79	15.01
Control group	T0	21	2.87	.84	21	1.37	.44	21	1.08	.49	17	35.54	59.29
	T1	21	2.79	.90	21	1.40	.51	21	1.13	.58			
	T2	21	2.90	.90	21	1.38	.31	21	1.11	.64	17	31.17	67.39

Note: T0 = Baseline; T1 = 4 weeks after plant placement; T2 = 8 weeks after plant placement; *n* = sample size; *M* = mean; *SD* = standard deviation

3.4 Influence of nature relatedness on impact of plants on human well-being

The self-reported nature relatedness was not found to influence any effects of plants on humans (p = .57; p = .97; p = .38, and p = .65 for well-being, perceived air quality, subjective chronic stress, and objective chronic stress, respectively), which does not support our hypothesis (H5: 1-4).

The final models of regression analyses were not found to explain variances for the criteria subjective well-being (R^2 = .04, F(6,66) = .50, p = .81), perceived air quality (R^2 = .03, F(6,66) = .283, p = .94), subjective chronic stress (R^2 = .12, F(6,66) = .298, p = .20), and objective chronic stress (hair cortisol) (R^2 = .12, F(6,53) = 1.191, p = .33). However, the final model showed that the subjective chronic stress is less reduced in the control group compared to the plant group (β = .29, t(72) = 2.15, p = .04).

3.5 Follow-up survey

The analysis of the follow-up survey revealed that most participants in the plant+info group (n=18) read the information sheet (describing a good pollutant removal efficiency of the given plant) quite detailed (M = 3.8, SD = .96) and that the information was rated as quite credible (M = 3.8, SD = 1.07). Further, an item approved that participants did not influence each other to a large extent, since they exchanged opinions “rather seldom” (M = 2.37, SD = .88). This was similarly responded in all experimental groups and the control group (F(2,54) = 2.25, p = .12).

Independent of each other, the total sample assessed an indoor plant in the office as “rather part of living nature” (M = 2.53, SD = .89) and as “rather positive” (M = 2.96, SD = .87), and all groups answered these questions equally (F(2,54) = 1.68, p = .20 and F(2,54) = .34, p = .72, respectively). Finally, a point-biserial correlation ensured that there was no coherence between randomly assigned passive samplers (for monitoring VOC in indoor air) and response behavior of particular participants and the remaining sample for subjective well-being, perceived air quality, subjective, and objective chronic stress (all p > .05).

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4. Discussion

This was the first study that examined a possible placebo effect in regard to psychological and physiological benefits of plants on human in the working environment and further, if this benefits depend on self-reported nature relatedness of human beings. Office workers, representing mainly scientific assistants, were supplied with either one plant, one plant plus an information sheet about a good pollutant removal efficiency of the plant or no plant. IAQ was monitored by measuring temperature, RH, and VOC. Well-being was assessed by online surveys regarding general health, perceived air quality, and subjective perceived stress, and by cortisol level of hair as physiological indicator. Results were controlled for gender, remaining demographic variables were not different in experimental groups.

Our results on IAQ revealed no differences between planted and non-planted offices concerning level of TVOC, temperature and RH. This indicates no major impact of the plant on IAQ which confirms part one of our first hypothesis. Although the concentrations of ethylbenzene, toluene, and mp-xylene were partly significantly lower in planted versus non-planted offices, the concentrations the 5 remaining VOC showed no differences between planting regime. The level of 2-ethylhexanol and hexanal were even in tendency lower in non-planted offices. Therefore, also the second part of our first hypothesis is largely confirmed. Fluctuations in VOC concentrations over time appear to be random and show no clear pattern. Even though TVOC concentration in non-planted offices increased significantly during time, which may be attributed to a very low concentration measured in the baseline (around 65 $\mu g\, m^{-3}$), the level of TVOC in non-planted offices did not exceed the concentration measured in planted offices. Indeed, the level of TVOC is in tendency lower in non-planted offices at any measure. However, differences in VOC concentrations between planting regime or during time can be hardly attributed to the plant. Other factors like various emission sources or variations in ventilation are more credible explanations. Generally, the indoor environment is dynamic and influenced by e.g. equipment and processes that may generate pollutants or heating/cooling processes, ventilation and air distribution that mix the air and might generate flow paths that carry contaminants (Tham, 2016). Also human activities, including e.g. the use of room fragrances or cosmetics and other products for personal care, have a great influence on the level and composition of VOC in indoor air (Rösch et al., 2014). Indeed, we observed passive evaporating room fragrances in some offices, which might lead to higher concentrations of VOC (Uhde & Schulz, 2016). Also the absence of employees, e.g.

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due to school holidays or illness-related absence, could have led to changing compositions of VOC. Furthermore, many participants kept their office door open during the day, which leads to very high ventilation rates. This comprises an overwhelming effect in comparison to the plant, since already slight differences in ventilation influence VOC concentration to a much greater extent than indoor plants (HBI, 1992). Other field studies have shown that even an excessive indoor greening could not improve IAQ markedly, especially in regard to VOC. In some studies, measured VOC were even higher in planted versus non-planted rooms (Dingle, Tapsell, & Hu, 2000; HBI, 1992; Kim et al., 2011, 2013; Lim et al., 2009; Smith & Pitt, 2011). This might be caused by a VOC emission by plants (Kesselmeier & Staudt, 1999; Yang et al., 2009), or by an emission from plant containers (Smith and Pitt 2011). Other authors assume a reduced ventilation in the presence of plants (Kim et al., 2013; Lim et al., 2009). Another reason for a poor pollutant removal by plants is likely the limited natural mass transfer of pollutants to the relatively small leaf area. Thus, only one plant is likely not powerful enough to purify the air in an office significantly. This is underlined by a number of critical views that suggest that even a moderate planting regime cannot have an impact on IAQ to a large extent (Girman et al., 2009; Levin, 1992; Llewellyn & Dixon, 2011; Schmitz, Hilgers, & Weidner, 2000; Soreanu et al., 2013).

In regard to psychological and physiological effects of plants on humans we did not find the expected benefits, and a possible placebo effect could not be determined. Contrary to our expectations we found no improvement of well-being and perceived air quality. However, participants showed already in the baseline no deficiencies and air analysis revealed an excellent IAQ. The concentrations of TVOC were well below 0.3 mg m^{-3} in every treatment at all sample times and was thus of no hygienic objections (Umweltbundesamt, 2007). The singly evaluated VOC were also far below the toxicologically derived guideline values of the German Environment Agency (Umweltbundesamt, 2017) and the chromatograms showed no striking values of VOC in general. The temperature and RH remained in a comfortable range throughout the whole test period. Apparently, participants had no reason to complain all along and therefore, an improvement was hardly obtainable.

We found a reduction in subjective chronic stress, though, only in the plant group but not in the plant+info group. This finding is contradictory to our hypothesis (H4.1) as well. However, the significant reduction found in the plant group (comprised of employees of the Department of Mathematics) cannot be necessarily attributed to the plant. Otherwise, this effect should

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have been found also in the plant+info group (comprised of employees of the Department of Computer Science). Although both groups work at the same university, they may differ by their workload in December.

The hair cortisol analysis, comprising our measure of objective chronic stress, revealed no changes over time in any of the experimental groups and therefore, also did not confirm our hypothesis (H4.2). Since it was already shown that hair cortisol level is influenced by vegetation (Honold et al., 2015), this indicator seems reasonable to use. However, one plant in the office is apparently not equivalent to e.g. vegetated pedestrian trails used in the study by Honold et al. (2015) or other natural environments with restorative character (Roe & Aspinall, 2011; van den Berg & Custers, 2011). Besides, office workers likely focus their attention on their work, rather than on a plant which is possibly just regarded as part of the office equipment. Thus, studies of Shibata and Suzuki (2001) have shown that stress-reducing effects by plants are more effective during a work break than while working on a task.

In general, effects of plants on humans in the working environment seem to be a small effect (f = .10), which is not detectable with the sample size used in our study (n = 74), rather than a medium effect as assumed in advance. As stated in the Cochran report placebo intervention can influence patient-reported outcomes only in certain settings. Large effects on pain were found, whereas the effects on pain varied from negligible to clinically important. Effects on nausea were small but consistent. Besides, it is stated that placebos seem to have no important clinical effects in general (Hróbjartsson & Gøtzsche, 2010). Also other findings suggest that placebos are more effective when negative symptoms are clearly noticeable (Meissner & Ziep, 2011). This might be caused by a requirement for an effective reduction of complaints (Price & Fields, 1997). According to this, placebo-induced effects of plants on human in the working environment might only be expected if stress-related suffering or other health complaints are large enough to be equated with pain.

Furthermore, the information sheet about a good pollutant removal efficiency of the given plant failed to trigger an (additional) expectation in the plant+info group. Although the information was read, as revealed by the follow-up survey, the potential change of perceived air quality was rated equally by both groups with plants. Possibly, the information sheet and only one plant per room were not sufficient to generate or strengthen respective expectations among participants. Basically it has to be questioned, if the information sheet used in the present study would be equivalent to a suggested effect in a therapeutic treatment of

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conventional placebo studies. Usually, studies focusing on a medical effect of placebos are characterized by participants that suffer from disease or have deficiencies in well-being and health. A verbally-induced expectation on e.g. a pain reduction of chronic low back pain (Carvalho et al., 2016) has probably another effect on suffering participants than our written information about a good pollutant removal efficiency of a plant for healthy participants.

Generally, the selection of participants appeared to be not optimal, since persons with a high level of education have usually a superior job satisfaction in comparison with employees of low education levels (Oreopoulos & Salvanes, 2011). Studies have shown that well-designed jobs and jobs that provide learning opportunities improve health (Rau, 2004, 2006). These features belong to working activities of our sample comprised of university employees. Furthermore, the participants in the current study showed already at the baseline no deficiencies in regard to well-being and stress level. Subsequently, an improvement of the evaluated parameters was difficult to achieve.

Our extended psychological issue concerned a possible relationship between nature relatedness and dependent variables (subjective well-being, perceived air quality, subjective and objective chronic stress between). This was the first time that the aspect of nature relatedness was regarded in a workplace-related context. Our results revealed that the self-rated nature relatedness did not influence psychological and physiological effects of humans as expected (H5: 1-4). However, it has to be questioned if the parameter nature relatedness is suited to examine this issue at all. Although it is regarded as a relatively stable personal characteristic (Nisbet et al., 2009) and associated with happiness, which is a component of well-being (Capaldi A., Dopko L., & Zelenski, 2014), it remains unclear if this parameter is important for the work routine in general and for this group of persons in detail. Further research is needed to analyze the impact of nature relatedness on workplace-related aspects of human in more detail.

In summary, the main limiting factors of this study were the small sample size and the selection of participants. The sample was not optimal due to the good subjective and objective well-being at the start of the experiment, the overall low stress level, the high level of education, and the focus on cognitively-challenging activities of participants. In further studies it should be ensured that participants suffer considerably from stress, because a

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recovery from stress is only possible if a need is existing (Bringslimark, Hartig, & Patil, 2007). Moreover, the number and diversity of applied plants should be increased to examine if variety of plants influences the impact on mentioned effects as described by Honold et al., (2015). Thereby, it should be assured that selected plants are rated positively by participants.

5. Conclusion

In conclusion, our study failed to determine significant effects of plants on humans in the working environment. However, our results confirm the heterogenic findings reported in literature concerning such effects (Bringslimark et al., 2009; Larsen, Adams, Deal, Kweon, & Tyler, 1998; Nieuwenhuis et al., 2014) and furthermore, this study closes a research gap. The assumption of an existing linear, monocausal relationship between the presence of plants in a working environment and the subjective well-being and stress level of humans cannot be supported by our findings. These parameters appear to be affected rather multicausal. Also, aspects like size, species, and color of plants, that concern subjective perception and valuation, might affect the outcomes of a study (Bringslimark et al., 2007). The people-plant relationship in working environments consists of a complex interaction among the way the plant is applied, the characteristics of the plant, and the characteristics of the individual employee (Thomsen, Sønderstrup-Andersen, & Müller, 2011). Further research is needed to fully understand the relationship between plants in the working environment and human well-being. Special care should be taken in the selection of participants, which should show pronounced deficiencies of dependent variables like well-being and stress level in the baseline.

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Limitations of plants to reduce volatile organic compounds and approaches to enhance the uptake by plants

In recent decades more than 100 plant species have been shown to remove indoor pollutants, mainly VOC (Dela Cruz et al. 2014; Hörmann et al. 2017a). Usually, those results were produced by chamber experiments. Since the capacity for a removal of VOC could be generally proven, it is a widespread assumption that plants can be used to clean indoor air. However, independent of the results in particular, it is difficult to rate these values in regard to real-life settings and data may be misinterpreted in regard to actual VOC removal under indoor conditions. Therefore, in this doctoral thesis the removal of VOC was expressed as a removal rate constant (see Hörmann et al. 2017b), representing the volume of air which is purified per unit time per leaf area ($l\ h^{-1}\ m^{-2}$). This parameter can be compared with the volumetric air flow and is thereby useful for two reasons (Girman 1992):

- It allows the transfer of the plant´s removal capability to any indoor environment (other than the test chamber).
- It allows the comparison with the most common VOC removal process, i.e. ventilation.

Ventilation means the introduction of outdoor air into a building or room by passive (uncontrolled) air exchange through leaks in the building envelope or actively by opening of windows/doors. The rate of ventilation is expressed as air changes per hour and represents the volumetric air flow in relation to the room volume (Vedavarz et al. 2007):

$$ventilation\ rate\ \left({}^{1}/_{h}\right) = \frac{volumetric\ air\ flow\ \left({}^{m^3}/_{h}\right)}{room\ volume\ (m^3)}$$

As described in detail in Hörmann et al. (2017a), results from chamber experiments are difficult to transfer to real-life settings. However, data obtained in this doctoral thesis can be considered as rough estimates. The results indicate a very limited removal capability of aerial plant parts of *Dieffenbachia maculata*, *Spathiphyllum wallisii*, and *Asparagus densiflorus* in comparison with active ventilation. Even natural (passive) ventilation (also referred to as infiltration, considering exclusively the air exchange through leaks in the building envelope) seems to cause a much more pronounced air exchange than plant foliage. To achieve similar ventilation rates or volumetric air flows with the VOC removal of plants under real-life

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settings, several hundred square meters of leaf area would be necessary in e.g. a 30-m³ room, irrelevant of the plant species tested in this PhD (Tab. 3).

Table 1 Ventilation rate and volumetric air flow of infiltration and correspondences of *Dieffenbachia maculata, Spathiphyllum wallisii*, and *Asparagus densiflorus* with respect to a room with 30 m³. Data for plants are estimated from experiments with toluene under light conditions (n=4)

	Infiltration[a]	*D. maculata*[b]	*S. wallisii*[b]	*A. densiflorus*[b]
Ventilation rate (h^{-1})	0.26	0.00019	0.00019	0.00013
Volumetric air flow ($m^3\ h^{-1}$)	7.8	0.0056	0.0057	0.0040

[a]Average data from a survey of 80 buildings in Germany (Münzenberg et al. 2003)
[b]Data are referred to one square meter leaf area and to the aerial plant parts only

Although the removal rates for the different plant species were similar in the present study (Hörmann et al. 2017b), different removal rates for plant species are described (Yoo et al. 2006; Liu et al. 2007; Yang et al. 2009). However, the values for removal of VOC by aerial plant parts seems to be too low to be of impact for IAQ in general. Weidener and Teixeira da Silva (2006) even stated that hydrophobic compounds are solely adsorbed in small amounts by the cuticle and neither assimilated nor accumulated and that the decline of VOC concentrations in former studies had been misinterpreted as true detoxification.

The field study of this doctoral thesis had shown, that a potted plant (*S. wallisii* of height 95 cm) had no major effect on IAQ (Hörmann et al. submitted). Unlike our chamber experiments, we tested the whole potted-plant complex including potting soil, roots and microorganisms. Nevertheless, we expected no improvement of IAQ which was largely confirmed. The concentration of total VOC, and the relative humidity and temperature, which were measured as additional markers for IAQ, were not significantly different in offices with a plant compared to offices without a plant. Even though some particular VOC were partially lower in planted vs. non-planted offices, this is likely attributed to random fluctuations of VOC emission and ventilation, which is also indicated by VOC fluctuations over time found for total VOC and certain VOC in particular (Hörmann et al. submitted). Moreover, several studies revealed that even an excessive indoor greening has only little (if any) effects on IAQ (HBI 1992; Dingle et al. 2000; Lim et al. 2009; Kim et al. 2011, 2013; Smith and Pitt 2011).

Transgenic plants may be more efficient in removing VOC. Studies have shown that the uptake of formaldehyde by transgenic plants (e.g. modified with a bacterial formaldehyde-fixing pathway) is increased by up to 20% (Sawada et al. 2007; Chen et al. 2010). The

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modification of *Nicotiana tabacum* cv. Xanthii (modified with a mammalian cytochrome) resulted in greatly increased removal rates of several VOC, including benzene, toluene and trichloroethylene (James et al. 2008). However, consumers still have ethical and moral concerns about genetically modified organisms (Frewer et al. 2013; Wunderlich and Gatto 2015) and the perception of transgenic houseplants may be negative as well. An increasing effect of VOC removal may also be induced by epiphytic bacteria in the phyllosphere. Studies have shown that *Pseudomonas* spp. and others considerably increase the removal rates of plants for toluene (De Kempeneer et al. 2004), phenol (Sandhu et al. 2007), and also polycyclic aromatic hydrocarbons (Yutthammo et al. 2010).

Assuming that the growing media and included microorganisms play a major role in the removal of VOC by potted plants (Wolverton and Wolverton 1993a; Wood et al. 2002; Orwell et al. 2006; Weidener and Teixeira da Silva 2006; Xu et al. 2011), removal rates are expected to be appreciably higher for the potted plant complex than for plant foliage only. Also, experiments of this doctoral thesis have shown that pots with common potting soil (unused) without a plant remove similar amounts of toluene and 2-ethlyhexanol like aerial plant parts (Hörmann et al. 2017a). Furthermore, it is reported that different porous materials (growstone, expanded clay and activated carbon), used as hydroponic growing media, are efficient in uptake of VOC. The best results were revealed by activated carbon, which is, however, an unfavorable growing media for plants (Aydogan and Montoya 2011). It could be rewarding to test different mixtures of activated carbon and usual growing media against plant growth and VOC removal.

However, independent of plant species and growing media, the most limiting factor for VOC removal by potted plants seems to be the natural mass transfer of pollutants to the relatively small leaf area / substrate surface, which results in little VOC removal under most conditions. Therefore, it is unlikely that simple potted plants can contribute to good IAQ in regard to pollution to a large extent, unless the amount of plants is excessively high like in the study conducted by BMW (Mohrlang 2003). In general, a moderate indoor greening with ordinary potted plants cannot replace ventilation.

A more promising approach in purifying indoor air is the application of an active botanical biofiltration system, which combines plants, growing media and air circulation equipment. Technical components actively draw air through the system which leads to a much higher air flow and thus effectivity. Such a system was developed by Canadian researchers and is described in Llewellyn et al. (2000) and Llewellyn and Dixon (2011). In brief, plants are

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grown in a vertical plane with roots located in a thin layer of porous medium. Air is drawn horizontally through the rooting medium by fans located in a plenum behind the biofilter and exhaust air is recirculated to the indoor environment (Llewellyn and Dixon 2011). Similar systems were already developed and patented, e.g. in Germany (Weidner and Schmitz 1996) and the USA (Saceman 1990; Wolverton and Wolverton 1993b). Here the technical component was integrated in a planting container. While the Canadian biofilter is still distributed, patents for the other systems have lapsed. For instance, in Germany the demand for these relatively expensive products was very limited. Furthermore, technical or patent related problems occurred, hence the distribution has been discontinued (Molitor 2015). However, although actively ventilated biofilter systems seem to be very promising, it must be kept in mind that such systems may release and spread bioaerosols, microbial borne VOC etc. This should be avoided by appropriate measures. Otherwise, such systems may lead to a changed, but not necessarily better IAQ. Apart from that, an actively ventilated biofilter is a very good approach, because it seems to be efficient enough to impact IAQ by treating significant amounts of indoor air. Furthermore, planted systems provide many other benefits. For instance, the advantage of a botanical air filter in comparison with pure technology-oriented systems is that pollutants get metabolized by e.g. soil-borne microorganisms and are not just accumulated in a filter medium. Moreover, it can be easily integrated in the building design, which has further the potential to affect stress, and thus health, of humans. Several design elements concerning stimulation (e.g. odor, color, and visual exposure), coherence (e.g. landmark and floorplan complexity), control (e.g. boundaries and privacy), and restorative (e.g. minimal distraction and fascination) may influence stress (Evans and McCoy 1998) and could be improved by plants. For instance, a vertical plant system could serve as room divider making interiors more comfortable by e.g. providing a privacy screen and through stimulation and restoration of cognitive resources. Thus, apart from a potential improvement of IAQ, one of the most important benefits of plants may be the psychological effects on humans.

Impact of plants on indoor air quality in regard to relative humidity and carbon dioxide

IAQ comprises aspects that affect well-being and health of humans. The contamination of indoor air by gases and house dust is used to measure IAQ objectively. Among gases, the concentrations of VOC and carbon dioxide are regarded as significant indicators (AGÖF

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2004; Schlechter et al. 2004; Umweltbundesamt 2007). However, the perceived IAQ is also of high relevance and usually measured by questionnaires (Ruotsalainen et al. 1991; Kosonen and Tan 2004). Important factors are e.g. room temperature, odors, dust and dirt, and dry air (Andersson and Stridh 1992).

The chamber experiments in this doctoral thesis enable the measurement of not only the removal of VOC by plants, but also the transpiration of water vapor, and CO_2 uptake and release. Since, the concentration of VOC, CO_2, and relative humidity (RH) are of relevance for IAQ, the potential impact of indoor plants on these parameters is further discussed.

Spathiphyllum wallisii showed a significantly higher transpiration compared to the two other tested species (*Dieffenbachia maculata* and *Asparagus densiflorus*), while the CO_2 uptake and release were equal for all plant species (Hörmann et al. 2017b). For interpretation of data in regard to IAQ, data of *S. wallisii* (control treatment) are set in relation to human activity (Tab. 2).

Table 2 Data for transpiration and CO_2 metabolism of *Spathiphyllum wallisii* and humans

	S. wallisii, 1 m² leaf area (Hörmann et al. 2017b)	Human (Kunsch and Kunsch 2007)
Transpiration (ml h^{-1})	14.8 (light); 2.8 (dark)	31.25[a,b]
CO_2 uptake (-); release (+) (l h^{-1})	-0.151 (light) +0.125 (dark)	+14.6 to +29.2[b]

[a]Insensible water loss (release of water via skin and mucous membrane by diffusion and evaporation without contribution of perspiratory glands); [b]Relaxed status.

Under well-illuminated conditions (180 ± 10 µmol m^{-2} s^{-1}), *S. wallisii* transpired 14.8 ml water per hour per square meter leaf area, ca. half as much as a human. Under dark conditions, the amount of transpired water was markedly lower. Usually, the light intensity in interiors (e.g. recommended for offices are 500 lx (DIN5035-2:1990-6) which corresponds to ca. 37.5 µmol m^{-2} s^{-1}) is substantially lower than in the Biobox experiments. Hence, it is assumed that the amount of transpired water under indoor conditions is in between the values found under light and dark. However, depending on plant parameters (e.g. species, water status, and leaf area) and on environmental conditions (light intensity, relative humidity, temperature etc.), the amount of transpired water can vary and therefore, plants might increase the relative humidity indoors. For instance, to raise the relative humidity in a 30 m³ room

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(22°C) from 35% to 40%, 29 ml of water would be necessary. Thus, it seems realistic that plants could have a significant impact on RH indoors.

The uptake of CO_2 by the three tested plant species was very limited, although a high light intensity was applied. In comparison to the amount a human exhales, the CO_2 metabolism of plants seems to be negligible as it was also outlined by other authors (Wood et al. 2006; Llewellyn and Dixon 2011). For example, the uptake of 150 ml CO_2 per square meter leaf area would correspond to a change of the CO_2 concentration of only 5 ppm per hour in a 30 m³ room. Taking into account that light intensity and accompanying CO_2 uptake is lower in interiors than in our chamber experiments and that plants do not take up CO_2 24 hours per day, but also release CO_2, plants do not seem to reduce CO_2 in interiors to a large extent. Vice versa, plants also do not seem to increase the CO_2 concentration to a detrimental level.

Health concerns related to plants

Although indoor plants are generally not regarded as originators of health complaints (the mishandling and potential threat of poisonous indoor plants is not considered here), they may be of concern in healthcare facilities. The substrate of potted plants is a natural habitat for a high diversity of microorganisms. Among them are also molds which are actually saprophytic, but can be considered as pathogenic fungi to plants as well as to human. For instance, *Fusarium* spp., *Penicillium* spp., and especially thermophilic fungi like *Aspergillus* spp. belong to the most virulent species found in potting soil of plants in hospitals (Summerbell et al. 1989; Hedayati et al. 2004). These fungi can cause invasive mycoses or allergies in humans and can produce mycotoxins which may lead to dysfunction of multiple organs (Hardin et al. 2003; Curtis et al. 2004). However, fungal infections and disorders due to mycotoxins are almost exclusively restricted to immunocompromised persons (Hardin et al. 2003; NRZMyk 2014) and plants should therefore not be prohibited categorically in hospitals, especially because of the beneficial effects on patients like shorter hospitalization and lower ratings of pain (Park and Mattson 2008, 2009). Under proper maintenance plants seem to pose no greater risk than other sources of infections like infective dust or aerosols from washbasins, nebulizers, or furniture (Schaal 1991). Another risk for human health is comprised of allergic reactions against plants. Well described is e.g. a sensitization and allergic airway symptoms of persons exposed to *Ficus benjamina* (Axelsson et al. 1991; Mahillon et al. 2006). However, hypersensitive persons (Brehler and Theissen 1996) or

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persons with a cross-sensitivity to latex (Schenkelberger et al. 1998) are primarily affected. In summary, only a few studies about the hazardous health effects of indoor plants are published, indicating no major risks of plants to healthy persons.

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Conclusion and Outlook

In conclusion, common potted indoor plants seem not to improve indoor air quality markedly, at least in regard to concentrations of total VOC and CO_2 (Hörmann et al., 2017b; Hörmann et al. submitted). Nevertheless, indoor greening can be recommended, because plants in the indoor environment have many other benefits (making interiors more comfortable and attractive, use as a room divider to give some privacy, potentially improving human well-being, and so on). Further research would be helpful to better understand the mechanisms of VOC removal by plants – covering passive sorption to cuticle and active uptake through stomata – depending on environmental conditions (especially climate, light and VOC identity, -concentration, and -mixtures) and morphological and physiological characteristics of plants. Moreover, the removal of other pollutants that occur indoors, like particulate matter and inorganic compounds (e.g. ozone, NOx), should be examined, as well as the influence of plants on other parameters of IAQ, e.g. RH and CO_2 / O_2 concentration. With such information, plants could be better selected and properly applied in a more efficient way in regard to IAQ improvement. In addition, the microbial consortia should be addressed as a further research goal, since they seem to be the main agents for VOC uptake and degradation in a plant system (Weidener and Teixeira da Silva 2006; Guieysse et al. 2008). Also the examination of potting mixes, which could be blended with adsorption-based filter materials like activated carbon, is highly rewarding. Moreover, the use of alternative materials which are known to be good absorbers like diatomaceous earth (Tsai et al. 2006) could be promising. Since common potting soil itself shows already good filter capabilities (Hörmann et al. 2017a) the combination with another absorbing material could lead to enhanced filtration capabilities. This is also conceivable for mixtures with inorganic substrates like expanded clay. However, the ratio of potting substrate to activated carbon or others must provide a good habitat for plants as well as for microorganisms. Further, it must be ensured that the absorber does not become saturated by compounds originating from the usual potting substrate. Another approach would be the incorporation of the absorber in planting container or fleeces used for (vertical) greening. A combination of plant(s), substrate, additional absorber (in the substrate itself or incorporated in the vessel), and appropriate technique(s) for moving significant amounts of air through the biofilter could be a very promising approach to improve IAQ additionally to all the other benefits of usual potted plants.

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Summary

People spend more and more time indoors, thus, indoor air quality (IAQ) and also the indoor environmental design become increasingly important for human health and well-being. A variety of studies have shown, that plants have the potential to improve IAQ as well as human health and well-being. However, published results in regard to both issues are very diverse. The actual impact of plants on IAQ and the examination of a putative interrelation between plant physiology and pollutant removal were investigated in the presented thesis. The filtration capability and plant physiological parameters were examined in test chamber experiments under laboratory conditions. Three common plant species, namely *Dieffenbachia maculata*, *Spathiphyllum wallisii*, and *Asparagus densiflorus*, were exposed under light or dark for 48 h to toluene and 2-ethylhexnaol, which are important indoor air pollutants belonging to the group of *volatile organic compounds* (VOC). Furthermore, a field study was conducted to evaluate the actual impact of plants on IAQ in real-life settings and to examine the influence of plants on human well-being. Therefore, half of overall 50 offices (ca. 20 m^2) were supplied with one plant (*S. wallisii* of height 95 cm) each and IAQ was monitored by data loggers and passive air sampling. The evaluation of well-being and stress level of 74 office workers was examined with questionnaires and hair cortisol analysis.

The chamber experiments revealed a toluene uptake of 70 to 117 $\mu g\ h^{-1}\ m^{-2}$ leaf area with no significant differences between plant species or light condition (light/dark). The values for 2-ethylhexanol were fluctuating around 22 $\mu g\ h^{-1}\ m^{-2}$ leaf area for all plant species tested, whereas differences between light and dark were observed for *D. maculata* and *S. wallisii*. Due to adsorption effects by the test chamber and differences in environmental conditions in test chamber versus real-life settings, the data obtained must be regarded as rough estimates. However, the extrapolation of data shows that the removal capability of one square meter plant foliage correspondence to a ventilation rate of only 0.0002 h^{-1}, with respect to a 30 m^3 room, what can be regarded as negligible. Therefore, a moderate planting regime will probably not reduce VOC concentrations in interiors markedly. The field study verified the limited performance of potted plants in regard to IAQ improvement. The relative humidity (~ 40%), the temperature (~ 22°C), and the concentration of total VOC (~ 100 $\mu g\ m^{-3}$) were not different between planted and non-planted offices throughout the whole test period of 8 weeks.

A relation between VOC removal and plant physiology was not found in the chamber experiments. Since no correlation between gas exchange of plants (transpiration, CO_2

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exchange) and uptake of toluene / 2-ethylhexanol was found, the uptake of these VOC seems to be primary based on passive sorption to plant surfaces rather than on active uptake by stomata. Phytotoxic effects of VOC were found to depend on plant species, VOC identity, and light condition. While the photosynthetic performance, in terms of quantum yield and CO_2 fixation, of *A. densiflorus* was clearly reduced by both VOC's, the other plant species were only affected by 2-ethylhexanol. Here, *D. maculata* showed a significant lower quantum yield and *S. wallisii* a decreased transpiration. Independent of plant species, a detrimental influence of VOC was only observed under light, not under dark.

The field study revealed no impact of the plant (*S. wallisii*) on health of office workers in terms of subjective well-being, perceived air quality, and subjective and objective chronic stress. Apparently, effects of single plants on human in the working environment are rather small and subject to a more complex system of varying factors.

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Zusammenfassung

Heutzutage verbringen die Menschen den Großteil ihrer Zeit in Innenräumen. Daher gewinnen die Qualität der Innenraumluft und auch das Design von Innenräumen eine immer größere Bedeutung für die Gesundheit und das Wohlbefinden von Menschen. Viele Studien haben gezeigt, dass Zimmerpflanzen das Potential haben die Qualität der Innenraumluft zu verbessern und die Gesundheit und das Wohlbefinden von Menschen zu steigern. Allerdings ist die publizierte Datenlage in beiden Themenfeldern sehr heterogen. Daher wurde in der vorliegenden Doktorarbeit untersucht, inwiefern Zimmerpflanzen tatsächlich einen Effekt auf die Innenraumluftqualität haben und weiterhin, ob es einen Zusammenhang zwischen der Pflanzenphysiologie und der Schadstoffaufnahme von Pflanzen gibt. Die Filterleistung von Pflanzen in Abhängigkeit diverser pflanzenphysiologischer Parameter wurde in Versuchskammern unter Dauerlicht oder Dauerdunkel für 48 Stunden untersucht. Dabei wurden die drei Pflanzenarten *Dieffenbachia maculata, Spathiphyllum wallisii* und *Asparagus densiflorus* den Substanzen Toluol und 2-Ethylhexanol ausgesetzt. Beide Stoffe gehören zur Gruppe der Innenraumluftschadstoffe (*volatile organic compounds* (VOC)). Darüber hinaus wurde eine Feldstudie durchgeführt, um den tatsächlichen Einfluss von Zimmerpflanzen auf die Innenraumluftqualität zu beurteilen und um die Wirkung von Pflanzen auf das Wohlbefinden von Menschen zu untersuchen. Hierfür wurde die Hälfte von insgesamt 50 Büroräume (ca. 20 m²) mit jeweils einer Pflanze (*S. wallisii* mit einer Höhe von 95 cm) ausgestattet. Die Raumluftqualität wurde mittels Dataloggern und passiver Luftproben beobachtet. Die Bewertung des Wohlbefindens sowie des Stressniveaus von 74 Büroangestellten erfolgte mit Hilfe von Fragebögen und Haarcortisolanalysen.

Die Kammerversuche ergaben eine Aufnahme für Toluol zwischen 70 und 117 $\mu g\ h^{-1}\ m^{-2}$ Blattfläche und keine signifikanten Unterschiede zwischen den Pflanzenarten und den Lichtbedingungen (Dauerlicht/Dauerdunkel). Die Werte für 2-Ethyhlhexanol schwankten um 22 $\mu g\ h^{-1}\ m^{-2}$ Blattfläche für alle Pflanzenarten, wobei hier signifikante Unterschiede zwischen Dauerlicht und Dauerdunkeldunkel für *D. maculata* und *S. wallisii* beobachtet wurden. Aufgrund von Adsorptionseffekten der Versuchskammern und weil die Umweltbedingungen in den Kammern im Vergleich zu realen Bedingungen in Innenräumen sehr unterschiedlich sind, sollten die Ergebnisse nur als grobe Richtwerte betrachtet werden. Dennoch zeigt die Hochrechnung der Daten, dass die Filterleistung von einem Quadratmeter Blattfläche, in Bezug zu einem 30-m^3-Raum, einer Ventilationsrate von nur 0,0002 h^{-1} entspricht, was als vernachlässigbar betrachtet werden kann. Von daher hat eine moderate Innenraumbegrünung

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wahrscheinlich keinen großen Einfluss auf die VOC -Konzentration im Innenraum. Die Feldstudie bestätigte, dass die Verbesserung der Innenraumluft durch Topfpflanzen sehr eingeschränkt ist. Die relative Luftfeuchte (~ 40 %), die Temperatur (~ 22°C) und die Summe der VOC (~ 100 µg m^{-3}) unterschieden sich nicht während der gesamten Versuchsdauer von acht Wochen in den begrünten und unbegrünten Büros.

Ein Zusammenhang zwischen VOC -Aufnahme und Pflanzenphysiologie konnte in den Kammerversuchen nicht nachgewiesen werde. Da zwischen den Parametern Gasaustausch (Transpiration und CO_2-Austausch) und Aufnahme von Toluol bzw. 2-Ethylhexanol keine Korrelation gefunden werden konnte, scheint die Aufnahme dieser VOC primär auf einer passiven Adsorption an Oberflächen der Pflanze zu basieren als auf einer aktiven Aufnahme durch die Stomata. Phytotoxische Effekte wurden in Abhängigkeit von Pflanzenart, VOC-Typ und Lichtbedingung gefunden. Während die Photosyntheseleistung hinsichtlich der Quantenausbeute und Kohlenstofffixierung von *A. densiflorus* durch beide VOC deutlich reduziert wurde, führte bei den anderen beiden Pflanzenarten nur die Exposition gegenüber 2-Ethylhexanol zu einer Reaktion. Bei *D. maculata* war hier die Quantenausbeute signifikant geringer und bei *S. wallisii* zeigte sich eine geringere Transpiration. Unabhängig von der Pflanzenart wurden schädliche Effekte der VOC aber nur unter Dauerlicht gefunden, nicht unter Dauerdunkel.

Eine positive Wirkung von Pflanzen auf die Gesundheit von Menschen konnte in der Feldstudie nicht festgestellt werden. Einzelpflanzen (*S. wallisii*) hatten keinen Einfluss auf das Wohlbefinden, die wahrgenommene Luftqualität sowie den subjektiven und objektiven chronischen Stress der Büroangestellten. Anscheinend sind Effekte von einzelnen Pflanzen auf den Menschen im Arbeitsumfeld eher klein und Gegenstand eines komplexen Wirkgefüges verschiedener Einflussfaktoren.

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