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Review

Nature-based solutions as tools for air phytoremediation: A review of the current knowledge and gaps



POLLUTION

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ABSTRACT

Monitoring of air quality and the application of strategies for its improvement are perceived as key areas for reducing environmental pollution. The research on Nature Based Solutions for the mitigation of pollutant concentrations in the air has increasingly developed in the last twenty years. The purpose of this review is to evaluate whether the current knowledge about Nature-Based Solutions provides a quantitative answer of the real benefits of air phytoremediation. To address this question, the literature on air phytoremediation over the last twenty years was analyzed. Altogether, 52 variables were selected, grouped into six categories, to briefly characterize the contents, methodology and outcome of the peerreviewed articles. Altogether, 413 plant species found in the analyzed studies were recorded. The results show the trends about the most studied pollutants and on the methodologies mostly applied, in relation to the study outcomes. The analysis demonstrated that particulate matter (PM_x) was the most frequently examined pollutant, most studies on NBS are based on experiments with exposure chambers, and scaling up the results with models has been limited. Although effective reductions in pollutant concentrations have been shown in the majority of studies, there is a strong fragmentation of the approaches, most studies have looked at a single pollutant and detailed information for model parameterization is only available for a few species. Thus, the review highlights that studies of Nature Based Solutions in air phytoremediation require unification of methodologies, and should consider a broader range of pollutants and plant organisms useful for mitigating the impacts of air pollutants in indoor and outdoor human environments.

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1. Introduction

1.1. The air pollution challenge

Nowadays, more than 55% of the world's population lives in urban areas, and this proportion is expected to increase to 68% by 2050 (United Nations, 2018). In the anthropo-ecosystems, people are exposed to about 200 different air pollutants some of which can affect the life quality and well-being of citizens, enhancing respiratory and cardiovascular diseases and mortality (Sicard et al., 2018; Orellano et al., 2020). As a matter of fact, air pollution has been recognized as the single largest environmental health risk worldwide, causing about 4.2 million premature deaths per year, with 400,000 cases per year in Europe, (WHO, 2016; EEA, 2019). Moreover, poor air quality may have several important environmental impacts and may directly affect natural ecosystems and biodiversity (WHO, 2015; EEA, 2019). Particulate matter (PM), carbon monoxide (CO), nitrogen oxides (NO_x), volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), potential toxic elements (PTEs), sulfur dioxide (SO₂), and secondary pollutants, such as ozone (O_3) , are typical anthropic sourced (Gawronski et al., 2016). However, there is no shortage of air pollutants of natural origin, such as biogenic volatile organic compounds (BVOCs) and radon. Since people in industrialized countries spend more than 90% of their lives in closed environments (Parseh et al., 2018), even indoor air pollution may pose a serious threat to human health (Soreanu et al., 2013; Brilli et al., 2018). Continuous exposure to indoor air pollutants, may cause diseases eventually contributing to the so-called "sick building syndrome" and "building-related illnesses" (Brilli et al., 2018). Therefore, the air



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pollution challenges facing the actual society are very complex and include the development of mitigation tool.

1.2. Nature-based solutions and ecosystem services

An increasingly promoted method for air pollution mitigation is the use of nature-based solutions (NBS). NBS to cope with atmospheric pollution refer to (i) the plant ability to absorb and catabolize almost any airborne pollutant and (ii) efficient pollutant deposition onto vegetation, rather than onto artificial surfaces. Indoor plants in living rooms and work places, trees in streets and parks, green walls, green roofs, and other green infrastructures and systems are increasingly being used to green the urban environment and reduce air pollution, by providing regulation ecosystem services (ES) (Escobedo and Nowak, 2009; TEEB, 2010; Brilli et al., 2018; Roeland et al., 2019; Hewitt et al., 2020). Once the importance of ecosystems for sustaining human well-being, has been realized, interest has grown in understanding how and how much NBS, present in anthropo-ecosystems, can meet human needs by providing ES (Davies et al., 2017; Sicard et al., 2018; Gopalakrishnan et al., 2019). Although many studies have looked at air phytoremediation, the empirical evidence for the effectiveness of NBS in enhancing air quality is still weak, because of the absence of a systematic method to assess NBS impacts on urban air (Hewitt et al., 2020), both for outdoor and indoor experimental systems used to characterize phytoremediation (Brilli et al., 2018; Moya et al., 2019; Sicard et al., 2018; Hewitt et al., 2020; Teiri et al., 2020). Moreover, within indoor studies, vegetation systems have shown a potential for mitigating pollutant concentrations, but there is still a lack of solid body of data to understand the pollutant removal mechanisms and to quantitatively characterize the impact of plants on pollutant concentrations in these systems (Moya et al., 2019; Kumar et al., 2019). In addition, the application of the NBS in air phytoremediation is controversial as several plant species, in particular trees, have the capacity to emit a considerable amount of different biogenic volatile organic compounds (BVOCs) (Niinemets et al., 2011; Calfapietra et al., 2013) which play a major role in urban air quality. The photochemically-driven reactions between NO_x and BVOCs lead to formation of various air pollutants such as ozone and particulate matter, ultimately contributing to smog formation (Calfapietra et al., 2013). Given this background, it is critically important to quantitatively evaluate the real mitigating effects as the result of NBS, and assess the trade-offs between ecosystem services and disservices (Hewitt et al., 2020).

1.3. Different approaches for studying the effectiveness and applications of NBS in air phytoremediation

The search for an exhaustive methodology that can confirm the effective value of the different plant species proposed as NBS, has led to development of various lines of research. For different pollutants, environmental conditions and approaches, the experimental methods used can be divided among (i) active or direct methods, (ii) passive or indirect methods and (iii) mathematical models (Amorim et al., 2015; Sicard et al., 2018). For the analysis of particulate pollutants, a series of extractions with different solvents is typically used to isolate the pollutants from the plant matrix and allow their qualitative and quantitative analysis (Dzierżanowski et al., 2011; Baldacchini et al., 2019). The ratio of plant absorption and emission of gaseous pollutants depends on the compensation point for the given pollutant, i.e. the pollutant concentration at which the uptake and release are equal and net exchange is zero (Niinemets et al., 2011). The techniques to determine the compensation points include measurements of variations in gas concentration in the open field, combined with modelling approaches, measurements of compound uptake and release under constant environmental conditions, or modelling approaches (Schjoerring et al., 1998; Fowler et al., 2009; Niinemets et al., 2011; Soreanu et al., 2013; Sicard et al., 2018). From the 2000s onwards, numerical dry deposition models have been developed for the quantification and prediction of air quality ESs delivered by NBS (Nowak and Crane, 2000; Nowak et al., 2006; Niinemets et al., 2014). One of the most used dry deposition models is the Urban FORest Effects (UFORE) and following developments (i-Tree) (Nowak and Crane, 2000; Nowak et al., 2006; Sicard et al., 2018). These models estimate the deposition rates (both stomatal and non-stomatal deposition) for a wider range of pollutants (O₃, SO₂, NO₂, CO and PM) by using the concepts of dry deposition on plant surfaces and into plant leaves, and modelling stomatal conductance (Nowak and Crane, 2000; Nowak et al., 2006; Sicard et al., 2018).

1.4. Aims of the review

Given the great variety of methodologies, and the consequent variety of research outcomes in the literature the main question that gives rise to this review is: "Does current knowledge about Nature-Based Solutions offer a clear answer to the real benefits of NBS in air phytoremediation?" Guided by this question, we analyzed the scientific literature aiming at (i) estimating the trends of the research lines over time, (ii) evaluating the availability of data and models on plant-atmospheric pollutant interactions and (iii) comparing different approaches and suggesting promising methodologies to assess the capacity of NBS in air detoxification.

2. Methods

2.1. Literature search process and refinement criteria

A literature search was conducted with the aim of finding all relevant studies that have examined air phytoremediation and NBS, and address the three aims of the study (Fig. 1). A robust and comprehensive literature search was conducted using three research databases: The Web of Science, Scopus, and PubMed. Each database enables access to a wide range of peer-reviewed literature. We included all research articles written in English from the year 2000 until July 2020 to depict a trend of research topics over the last twenty years.

The keywords "air pollution", "plants", "phytoremediation", "air quality", and "urban forest" were included in the search in all databases used. To obtain as wide a range of available evidence in this field as possible, no restrictions were imposed on the research method, experimental design, and air pollutant type, neither on the type of environmental conditions (indoor or outdoor). The query used was ("air pollution" AND "plants" AND "phytoremediation") OR ("air pollution" AND "air quality" AND "urban forest"), which

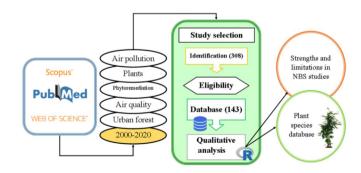


Fig. 1. Illustrated workflow of the present study.

returned the broadest number of peer-reviewed articles relevant to the topic under review.

The search found 153 articles in Scopus, 121 in Web of Science, and 34 in PubMed databases (308 in total). Additional refinement criteria were used to exclude articles that were not relevant for the topic. The refinement process was carried out by analyzing the title, keywords, and abstract of each article. For the articles, topic of which was unclear after the first screening, an in-depth analysis of the full article text was conducted. Duplicate records were identified and removed. As we were interested in the primary research, review articles, included in the three databases were removed. Ultimately, 143 unique research papers were identified.

2.2. Data extraction and database construction

All articles were carefully reviewed and all relevant information was identified to construct a database for analysis of potential trends in the literature and answer our research questions (Fig. 1). Altogether 52 relevant variables were identified, and the data matrix was filled based on the presence (coded as 1) or lack (coded as 0) of information about each variable for each scientific manuscript. The procedure was repeated twice by two co-authors of the MS, and when there were discrepancies among the two independent assessments, the specific cases were jointly analyzed and a consensus assessment was made. To evaluate the relationship between species taxonomy and use in air phytoremediation, a second database was constructed that included the full taxonomic identity of all plant species (species names, genus and family) found in the articles together with the 52 selected variables. Species taxonomy follows International Plant Names Index (IPNI, 2020).

Relevant variables were classified according to seven categories. The first category "Article information" encloses all the information useful to identify the individual peer-reviewed article. Second, the

Table 1

Acronyms and full names of pollutants reported as variables in the matrix.

Abbreviation	Full name	
BC	Black carbon	
Benz	Benzene	
BT	Bromotoluene	
BVOCs	Biogenic Volatile Organic Compounds	
CH ₄	Methane	
CO	Carbon monoxide	
CO ₂	Carbon dioxide	
DehP	Di (2-ethylhexyl) phthalate	
EA	Ethyl acetate	
EB	Ethylbenzene	
FA	Formaldehyde	
H ₂ S	Hydrogen Sulfide	
HNO ₃	Nitric acid	
IsoP	Isoprene	
MonoT	Monoterpene	
NH ₃	Ammonia	
NO	Nitrogen monoxide	
NO ₂	Nitrogen dioxide	
03	Ozone	
PAHs	Polyciclic Aromatic Hydrocarbons	
pC	p-Cresol	
Ph	Phenol	
PM _{0.2}	Particular Matter with mean particle diameter $< 0.2 \ \mu m$	
PM ₁₀	Particular Matter with mean particle diameter $< 10 \ \mu m$	
PM _{2.5}	Particular Matter with mean particle diameter $< 2.5 \ \mu m$	
PTEs	Potentially Toxic Elements	
Rn	Radon	
SO ₂	Sulfur dioxide	
Tol	Toluene	
VOCs	Volatile Organic Compounds	
Ху	Xylene	

type of environment for the research (indoor or outdoor) was included in the category named "Context of application".

All single pollutants or group of pollutants (e.g., VOC, Table 1) were included as individual variables as reported in the peerreviewed articles, and subsequently categorized as "Pollutants". However, some classes of pollutants (e.g., VOCs) inherently included some of the pollutants coded also as single variables in other studies. In such cases, the pollutants were rearranged to characterize as closely as possible the data in the original articles.

The variables included in the "Experimental method" category were determined based on a classification of the methodologies found in the analysis of the manuscripts. Methodologies that included the extraction of pollutants from the plant matrix, through solvents or washing, were labelled as "Washing/Extraction". These methodologies were mainly used for semi- or nonvolatiles pollutants. The methodologies "Exposure chambers" and "Models" included, respectively, the methodologies that used sealed chambers with pollutant-dispersion systems and mathematical models linking environmental variables and pollutant fluxes. Any other methodology that included laboratory experiments was labelled as "Laboratory measurement", and any methodology that included field surveys and experiments was labelled as "In-situ measurement".

Classification criteria for the variables in the "Results" category were determined by analyzing the results obtained in the peerreviewed articles. If the outcome of the study was based on a measurement, it was categorized as "Measured outcome". If they originated from predictive models, it was categorized as "Modelled outcome". An additional differentiation criterion of the results was the relevance of the results to address the aim of the work (naturebased solutions). If the study demonstrated an effective reduction of the pollutant concentration(s), it was classified as "Effective pollutant reduction". Alternatively, the study was classified as "Non-effective pollutant reduction", i.e., the results of the study did not show an effective reduction of the pollutant concentration(s).

In the "Purpose" category, depending on the aim of the work, the studies were divided among two classes. "Air phytoremediation" class included studies that looked at nature-based solutions to mitigate the air pollutant concentration. "Plant stress tolerance" class included studies that mainly dealt with air pollution stress tolerance of plants useful for the application of naturebased solutions. All the relevant variables have been summarized in Table 2.

2.3. Database analysis

The *zPatterns* function of *zComposition* R package (Palarea-Albaladejo and Martin-Fernandez, 2015) was used to find and display information of patterns (characteristic combinations of study variables) in the constructed dataset. This function investigates the patterns of observed/present (0) and unobserved/ missing (1) components in the dataset and returns a vector of pattern numbers. Summary statistics include the number of observed component frequency (%), and the statistics are also presented graphically. The function assigns an ID number to each identified patterns, ranked by their frequency. In brief, each pattern depicts a specific combination of variables/components (categorized as observed/present and unobserved/missing). In addition, the frequency of the peer-reviewed articles sharing the same features is also provided.

An illustrated workflow of the study, from the literature search to the analysis results, has been provided in Fig. 1.

Table 2

List of identified variables in the literature survey.

Category of variables	Variables	Description
Article information	(3) Title, authors, and year of publication	Bibliographic information about the peer-reviewed article
Context of application	(2) Indoor or Outdoor	Type of environment for the research
Pollutants	(31) PM ₁₀ , PM _{2.5} , PM _{0.2} , VOCs, BVOCs, NO, NO ₂ , CO, CO ₂ , SO ₂ , O ₃ , H ₂ S, NH ₃ , HNO ₃ , Isoprene, Monoterpenes, Formaldehyde, Ethylbenzene, Benzene, Xylene, Toluene, Bromotoluene, PTEs, PAHs, p-Cresol, Phenol, Rn, Ethyl acetate, Methane, Black carbon, DEHP	Pollutants included in studies of removal dynamics
Plant species	(5) single species, multiple species, plant life form (tree, shrub, herb)	Number of plant species and plant life form studied in each scientific publication
Experimental method	(5) Washing/Extraction, Exposure chambers, Models, In-situ measurements, Laboratory measurements	Methodologies used to achieve the postulated goal in each scientific publication
Results	(4) Measured or modelled outcomes, Effective pollutant reduction, Non-effective pollutant reduction	Type of achieved results
Purpose	(2) Air phytoremediation and Plant stress tolerance	Aim of the study

The numbers in brackets denote variables in each category.

3. Results and discussion

3.1. Database analysis

The main database included 143 rows (individual research papers) and 52 columns (variables) divided among seven variable categories (Table 2, Supplementary Table S1). The complete reference list corresponding to the data and the results showed in this paragraph have been reported in the Supplementary Materials.

In the last twenty years, the volume of the scientific literature in air phytoremediation shows a rising trend (Fig. 2 and Supplementary Table S1). According to our analysis, until 2002 there were no papers that matched the selected search criteria. Since 2003, the number of papers has increased, but not steadily, showing the maximum production of peer-reviewed articles in 2019, the last full year in the dataset.

This increase in article number in the given research area reflects the overall increase of the number of scientific publications across the disciplines, and could also be related to the growing awareness of environmental issues in urban environments and the well-being of citizens, as well as awareness of the impacts of air pollution on the environment. Our analysis also demonstrates that in the last twenty years, the majority of articles have focused on indoor phytoremediation (Fig. 2). This is certainly due to the interest in the indoor environment since people spend increasingly more time indoors, and in developed countries, people stay most of their time indoors (Soreanu et al., 2013; Brilli et al., 2018; Parseh

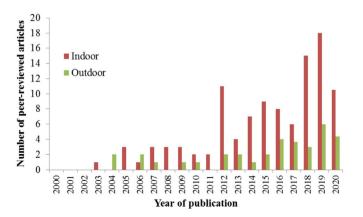


Fig. 2. Number of peer-reviewed articles dealing with air phytoremediation for the 2000–2020 time period separately shown for indoor and outdoor studies. The year 2020 only includes January–July.

et al., 2018). This could be explained by simpler approaches needed to analyze the effects of green systems in closed environments. The indoor environments represent semi-closed systems where the environmental and climatic factors have a weak influence on the flows of pollutants, and the setting and replication of the variables are facilitated in the experiments (Brilli et al., 2018; Pettit et al., 2018a). In outside conditions, environmental variables and climatic factors can vary over short distances and over short periods of time, and such variations might be difficult to predict and replicate, making outdoor studies complex (Garcia-Gomez et al., 2016; Hu et al., 2016; Baldacchini et al., 2019).

3.2. Pollutants investigated across studies

Our analysis demonstrated that the most widely studied pollutant was PM₁₀, analyzed in 44.1% of the inspected studies, followed by PM_{2.5}, studied in 41.3% of the papers (Fig. 3, Supplementary File 1 for the full list of patterns). Among the gaseous pollutants, NO₂ was the most frequently considered (25.9%), followed by O₃ surveyed in 22.4% of articles (Fig. 3). PM_{0.2}, VOCs, CO, and SO₂ were studied in 15-20% articles, and the other inspected pollutants were studied in less than 10% of articles (Fig. 3). The high scientific interest in the above pollutants shows that the research in air phytoremediation is largely guided by international treaties and regulatory targets concerning monitoring and mitigation of harmful air pollutants in urban environments (WHO, 2016; EEA, 2019). The data analysis presented in Fig. 3 shows the presence of a second group of pollutants (H₂S, NH₃, HNO₃, IsoP, MonoT, FA, EB, Ben, Xy, Tol, BT, PTEs, PAHs, pC, Ph, Rn, EA, CH₄, BC, DehP; Table 1 for acronyms) not frequently reported (frequencies lower than 10%) in the scientific literature of the last 20 years. These pollutants are not explicitly regulated by any monitoring agency (EEA, 2019). However, they have been recently recognized as a serious risk for human health (Beattie and Seibel, 2007; Wu et al., 2013; Li et al., 2015, 2018; García-Gómez et al., 2016; Shaheen et al., 2016; Deshmukh et al., 2019; Pace et al., 2018; Fooladi et al., 2019; Khalid et al., 2019; Sgrigna et al., 2020), and therefore are increasingly more considered in scientific studies (Supplementary Table S1). A large part of the compounds investigated in indoor air phytoremediation belong to this category of pollutants. The data highlighted in Figs. 2 and 3 show an apparent conflict as the indoor environment is the medium most frequently studied, but the pollutants characterizing it have a low frequency in the reviewed articles (Supplementary Table S1). This discrepancy can be interpreted as a fragmentation of efforts in the study of air pollutants occurring in indoor environments and the related phytoremediation strategies.

Our data analyses (Fig. 3) also highlight occurrence of 59 distinct

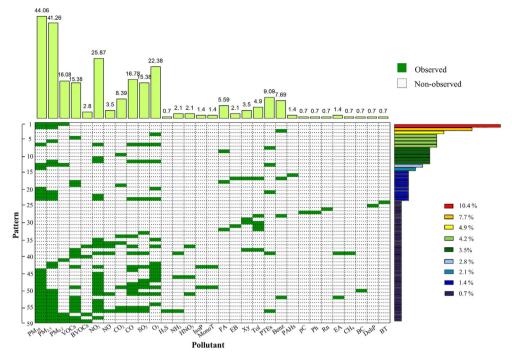


Fig. 3. Summary of the frequency of patterns (combinations of different key variables characterizing pollution in different studies) in the reviewed literature on air phytoremediation from 2000 to 2020 (143 peer-reviewed articles). Data are arranged by the type of investigated pollutant, and in the main part of the figure, given pollutant presence is shown by a filled cell and absence by an empty cell. The pollutants are shown on the lower horizontal axis along with the observed percentages in reviewed literature (upper part of the figure). The pattern codes are shown in the left y-axis, and their relative occurrence (%) is shown by the horizontal bars in the right. The acronyms are explained in Table 1 and the full list of the patterns is in Supplementary File 1.

combinations (patterns) of pollutants concurrently analyzed in peer-reviewed articles (Supplementary Table S1). The most significant patterns (frequency above 4%) are summarized as follows (Supplementary File 1 for the full list of the patterns):

- <u>Pattern 1</u> (10.4%) represents the peer-reviewed articles in which all the fractions of the particulate matter have been analyzed: PM₁₀, PM_{2.5}, PM_{0.2}.
- <u>Pattern 2</u> (7.7%) represents the peer-reviewed articles in which only the coarser fractions of particulate matter (PM_{10} , $PM_{2.5}$) have been analyzed.
- <u>Pattern 3</u> (4.9%) represents the studies in which only benzene has been analyzed.
- <u>Pattern 4</u> (4.2%) represents the peer-reviewed articles in which only ozone (O₃) has been analyzed.
- <u>Pattern 5</u> (4.2%) represents the studies in which VOCs have been analyzed.
- <u>Pattern 6</u> (4.2%) represents the studies in which the fine fraction of particulate matter (PM_{2.5}) has been studied
- <u>Pattern 7</u> (4.2%) represent the studies in which PM₁₀ have been analyzed together with other gaseous pollutants such as sulfur dioxide (SO₂), carbon monoxide (CO), nitrogen dioxide (NO₂) and ozone (O₃).

Thus, our analyses indicate that particulate matter in its various forms and with various levels of detail has been studied most thoroughly. On the other hand, the pattern 7 is the combination of factors included in the i-Tree Eco model (former Urban Forest Effects (UFORE) model) (Nowak and Crane, 2000; Nowak et al., 2006) widely used in different contexts with the results reported in a considerable number of articles (4.2%, see also Supplementary Table S1). The success of his model demonstrates that community-wide standards of the list of chemicals considered can

be achieved and this should be more widely encouraged. Except this pattern, analysis of the distribution of the frequencies of all patterns indicates that there is overall limited convergence among different lines of research in determining the pool of investigated air pollutants (Patterns 3, 4, 6, see also Supplementary Table S1). Furthermore, although the research has mainly focused on indoor environments, the pollutants typically belonging to the indoor pool are present with low percentages starting from the third pattern (Supplementary Table S1).

It is also evident, that most peer-reviewed articles have focused on a single pollutant or relatively small combinations of pollutants, while inclusion of a larger number of pollutants, more than three, is relatively rare (Fig. 3). This could reflect the obvious difficulties currently encountered in simultaneously studying and measuring interactions between plants and a wide range of pollutants, as happens in the real environment.

Our literature analysis indicates that in studies on NBS, the information of plant biogenic volatile organic compound (BVOC) emissions is limited. Plants, especially under stress, emit reactive BVOCs that in NO_x-polluted atmospheres can constitute an "ecosystem disservice", contributing to enhanced atmospheric reactivity, production of O₃ and fine particles, ultimatley leading to photochemical smog formation (Niinemets, 2011, 2018; Joutsensaari et al., 2015; Holopainen et al., 2017). Understanding how environmental drivers affect biogenic emissions, and species emission capacities is relevant for a targeted selection of plants (both for urban and suburban forests) that have minor BVOC emissions, and consequently can be used for urban landscaping without negative effects on air quality (Yuan et al., 2020).

3.3. Design of the phytoremediation studies

Among all studies considered, 62.2% studied more than one

pollutant, while the remaining 37.8% investigated a single pollutant (Fig. 4). Among the experimental methods used (Table 2), "exposure chamber" and "model" classes had the highest percentage of 30.1% (Fig. 4). The classes, "washing/extraction" and "laboratory measurements", had the same frequency of 20.3% (Fig. 4), and the last category "in situ measurements" had a frequency of 19.6% (Fig. 4). The sum of the frequencies exceeds 100% as some studies used multiple methods.

Experimental measurements ("measured outcomes") were used in 69.9% of studies, and modelling ("modelled outcomes") in almost 35.0% of studies (Fig. 4). Also in this situation, the sum of the frequencies exceeds 100% because several studies used both experimental measurements and modelling. Classification of studies based on the effective reductions in the concentration(s) of pollutants demonstrated that "effective pollutant reduction" was observed in 87.4%, and "non-effective pollutant reduction" in 11.9% of studies. This implies that the scientific research on air phytoremediation by measuring plant-pollutant interactions has provided, in majority, new knowledge on the subject, and useful data that can be translated into practical applications.

Nevertheless, there is still difficult to generalize because of large variation in effect sizes and lack of data of multiple relevant pollutants. The present uncertainties result from differences in experimental methods and their application protocols as there is currently no community accepted and standardized methodology for analysis. Although the majority of research outcomes in the field of plant-air pollutant interactions have been based on experimental measurements, extensive use is made of model estimations (Fig. 4). However, the parameterization of the models is often limited by availability of experimental information of environmental characteristics and pollutant physico-chemical properties alter plant pollutant uptake (Niinemets et al., 2014). leading to inherent uncertainties in model predictions.

On the basis of the analysis, 39 distinct patterns characterizing the study types were identified (Fig. 4). Each pattern represents

different combinations of inspected variables (Supplementary Table S1). Here we discuss patterns with a frequency above 5% in the pool of reviewed articles. The full list of the resulting patterns is provided in Supplementary File 2. In all the five most frequent patterns, the results suggested an effective pollutant reduction by plants.

- <u>Pattern 1</u> (13.9%) contains articles in which a single pollutant has been analyzed experimentally using exposure chambers, and the conclusions are based on measurements.
- <u>Pattern 2</u> (13.3%) represents studies looking at multiple pollutants have been analyzed using models.
- <u>Pattern 3</u> (12.6%) represents studies considering multiple pollutants have been analyzed using exposure chambers and the conclusions are based on measurements.
- <u>Pattern 4</u> (8.4%) includes articles using washing/extraction methods to analyze multiple air pollutants and the conclusions are based on measurements.
- <u>Pattern 5</u>: 6.2%, contains studies that use models to estimate the phytoremediation of single air pollutant.

Experiments in exposure chambers were most widely used in air-phytoremediation research (Fig. 4). In an exposure chamber, plant-single pollutant or plant-multiple pollutant interactions can be examined and measured. The possibility to alter/control the environmental variables and perform direct measures of pollutant absorption/emission represent the key advantages of this method (Maddalena et al., 2002; Niinemets et al., 2011). In addition, the exposure chambers allow selection and control of pollutant concentrations, distinguish various phases of plant physiological response over time, and measure plant pollutant absorption and its BVOC emissions (Rogers et al., 1977; Niinemets et al., 2011). It is well-known that, among different exposure chambers, the flowthrough dynamic chambers provide more realistic results than closed static chambers due to minimization of effects of diffusion

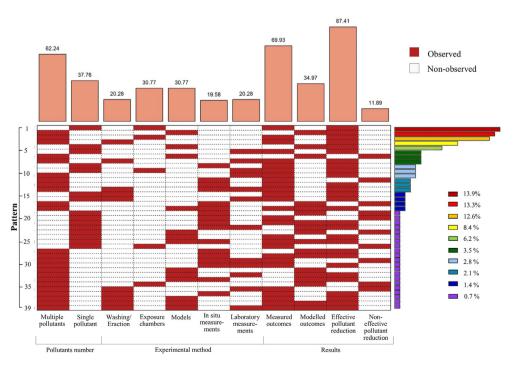


Fig. 4. Summary of the characteristics of the study design in the reviewed literature on air phytoremediation from 2000 to 2020 (143 peer-reviewed articles). The variables are shown on the horizontal axis (bottom side) along with the observed relative abundance (%) in reviewed literature (vertical bars on the top side). The patterns based on presence (filled cells) and absence (open cells) in the dataset are numbered on the left side. The frequency of occurrence (%) of different patterns is given on the right side.

leaks and better control of pollutant concentration (Maddalena et al., 2002; Niinemets et al., 2011). The limits of exposure chambers are mainly associated with their dimensions. They are typically relatively small to allow adequate air turnover for the given flow rate, and therefore do not allow the study of mature plants (Niinemets et al., 2011; Niinemets, 2012; Rao et al., 2014). Moreover, the gas-phase pollutant concentrations used in these studies are often of several orders of magnitude higher than environmental concentrations (Niinemets et al., 2011; Rao et al., 2014).

Our analysis further shows the extensive use of mathematical models to estimate the absorption of pollutants by urban green infrastructures. One of the most widely used models in urban and peri-urban environments is the i-Tree Eco model (Nowak and Crane, 2000; Nowak et al., 2006). This model requires meteorological data, pollutant concentrations drawn from local databases, and structural variables of the urban or peri-urban forest (Nowak and Crane, 2000; Nowak et al., 2006). The use of models such as i-Tree is convenient due to the relative ease of parameterization and interpretation of results, but they present limitations as they do not include the information about the physiology of plant species and species-specific relationships (Fowler et al., 2009; Sunderland et al., 2012; Wang and Lin, 2012; King et al., 2014; Rao et al., 2014; Pace et al., 2018; Parsa et al., 2019). These models currently in use reduce the complex eco-physiological mechanisms to an estimate of gaseous exchange based on physical parameters of the fluid (atmospheric air) and the individual pollutant (Nowak et al., 2006; Garcia de Jalòn et al., 2019). A review by Sicard et al. (2018) on the benefits of urban forests for O₃ mitigation showed that the average annual percentage of O₃ reduction levels estimated by i-Tree models is generally less than 2%, while studies based on passive samplers suggest O₃ reductions up to 40% in urban parks. Such discrepancies between model estimates and direct measurements suggest that dry deposition models are not yet able realistically incorporate the effects of vegetation on O3 reductions in all conditions. Our literature analyses also indicate that there is some disagreement between predicted and observed effects of urban forests on pollutant mitigation. Indeed, some in situ experimental studies have not observed an effective reduction in pollutant concentration(s) (Yli-Pelkonen et al., 2017a, b; Viippola et al., 2018; Yli-Pelkonen et al., 2020). These studies, performed with passive samplers, positioned either directly under the canopy of urban forests or far from the tree canopy, have concluded that there is no impact of the urban forests on the reduction of some air pollutants (Yli-Pelkonen et al., 2017a, 2017b; Viippola et al., 2018; Yli-Pelkonen et al., 2020). Despite this discrepancy, two thirds of the studies do demonstrate an effective reduction in pollutant concentration(s) (Supplementary Table S1). Clearly plant physiological status and pollutant physico-chemical characteristics need to be considered in numerical models predicting vegetation capacity to absorb air pollutants (Niinemets et al., 2014).

3.4. Analysis of the plant species used

All plant species present in the analyzed studies were recorded, resulting in a database of 413 species from 240 genera and 98 different families (Supplementary Table S2). Among the studies, 26.4% used a single species, whereas 70.1% used more than one species. In addition, the remaining 3.49% of articles did not specify the plant species and used a broad category, e.g. urban vegetation (Supplementary Table S1). The most widely considered species was *Chlorophytum comosum* (Thunb.) Jacques, a scapose herbaceous perennial used mainly in indoor air phytoremediation (10 citations in the database; Supplementary Table S2). The 80% of peerreviewed articles using *C. comosum* involved exposure chamber (Supplementary Table S1 and S2). These studies found a successful

ability of this species to take up VOCs and PM_x (Sriprapat et al., 2014; Torpy et al., 2016; Irga et al., 2017; Paull et al., 2018; Gong et al., 2019; Panyametheekul et al., 2019; Siswanto et al., 2020; Treesubsuntorn et al., 2020). Among tree species, Robinia pseudoacacia L. and Tilia cordata, Mill., both with 9 citations (Supplementary Table S2), were the two species most frequently used in outdoor air phytoremediation studies. We found that "washing/extraction" is the most adopted and performing method used to evaluate the air-remediation performance of the two above-mentioned tree species for PM_x, PTEs, and PAHs. (Dzierżanowski et al., 2011; Soudek et al., 2012; Przybysz et al., 2014a; Popek et al., 2015, 2017a; Sgrigna et al., 2020). At a genus level, 20 studies have used Ficus spp., followed by Quercus spp. with 16 citations and *Fraxinus* spp. and *Pinus* spp., both with 14 citations included in the database (Supplementary Table S2). Moraceae was the family comprising the greatest number of species used (25 citations), followed by Fabaceae (24 records, Supplementary Table S2). At the family level, there was a greater diversity in outdoor studies (75.3% families present in studies in outdoor environments) than in indoor studies (24.7%; Fig. 5). Regarding the share of individual families, most of them have been only used in outdoor studies, reflecting the circumstance that in indoor studies under more controlled conditions, researchers have mostly focused on a few widespread model species. In addition, most of the plant species used in indoor air-phytoremediation were ornamentals. This represents a limited but very important portion of the plant pool useful for purification of indoor air (Teiri et al., 2018a).

The data on species, genera and families confirm what has already been suggested in the previous analyzes. The studies on indoor air phytoremediation are limited to a small number of model species. Given the greater number of indoor studies, the species used in these studies are the most frequently employed in the pool of articles reviewed. Several of these species are widely accepted by the scientific community as useful for air phytoremediation, and due to data availability can be used for construction and parameterization of numeric models. The greater diversity of species used outdoors reflect a greater species number of urban forests, where plants from local species pool and exotics, often from different continents, are grown together (Yuan et al., 2020). However, limited experimental data on phytoremediation in different combination of environmental drivers and for different pollutants also complicates selection of promising taxa potentially useful for phytoremediation. Lack of information related to the physiology, genetics, and epigenetics of plants used outdoors and their capacity of uptake of different atmospheric pollutants implies that generalizations of urban vegetation phytoremediation are currently associated with significant uncertainties.

4. Conclusions

4.1. Findings

This review evaluated whether the current knowledge about Nature-Based Solutions offers a clear answer of the benefits of vegetation in air phytoremediation. The last twenty years of scientific literature (143 peer-reviewed articles) were reviewed and analyzed. First, we found that a majority of articles have focused on indoor phytoremediation probably due to the ease of working in closed environments both due to less variable pollutant concentrations, environment and lower plant diversity. As regards to the type of pollutant, we observed that the literature has primarily focused on the research of particulate matter (PM₁₀, PM_{2.5}, PM_{0.2}) probably due to due to its high importance for environmental policy. Particulate matter represents one of the main current and future challenges for air quality and climate change (Fuzzi et al.,

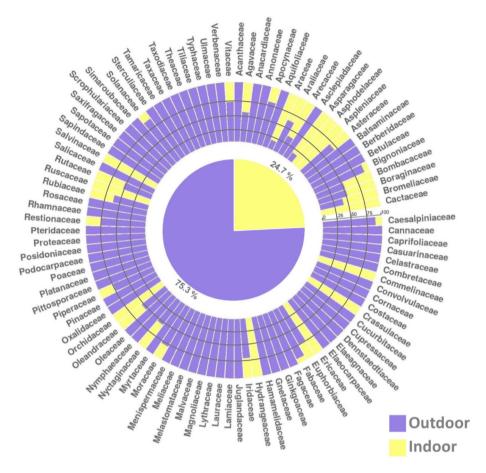


Fig. 5. Radial plot showing plant families and their respective percentages in indoor and outdoor experiments in the reviewed literature about air phytoremediation (2000–2020 time period). The total use in two different environments is shown in the center, and the frequency of use of each family is shown in the outer part of the circular histogram.

2015). The knowledge of particulate matter uptake by vegetation has increased considerably due to improved methodologies for extraction and quantification from the plant matrix (e.g., Dzierżanowski et al., 2011). These improved techniques are effective and reproducible in different environments and do not require overly sophisticated instrumentation (Baldacchini et al., 2019). Differently from particulate matter uptake, the stomatal and nonstomatal processes driving phytoremediation of gaseous pollutants are poorly understood and it is still challenging (Hu et al., 2016; Brilli et al., 2018). Uptake of the main gaseous pollutants is typically analyzed using exposure chambers, models, and in-situ surveys, often using passive samplers or other indirect techniques. This entails use of considerable approximations in bringing the data from the laboratory scale to the relevant indoor or outdoor scale (Yli-Pelkonen et al., 2017b). Some critical issues regarding the indoor environment had already been presented by Dela Cruz and Christensen (2014), which documented that moving from laboratory to real-life settings studies is necessary to clarify the plant's ability to take up VOCs in complex indoor environment. From our literature review, it is evident that there are few studies investigating a range of pollutants greater than six pollutant species, resulting in a major knowledge gap that hinders a reliable evaluation of NBS in air phytoremediation. Different experimental methodologies have been used across studies, and among the methodologies, the use of exposure chambers is most frequent (Fig. 4). Although exposure chambers do have certain limitations (Niinemets et al., 2011; Rao et al., 2014), it allows measurement of plant uptake in highly controlled manner and studies using exposure chambers have typically observed an effective reduction of the initial pollutant concentration(s) by plants.

Overall, we found that the current knowledge about NBS in air phytoremediation is heterogeneous. A huge number of pollutants has been analyzed, but only a few of these have been analyzed in individual studies. The overall heterogeneity is also reflected in the number of plant species tested for their potential in phytoremediation. As an alternative, modelling approaches have been used, but they have not always been parameterized by experimental data obtained through targeted and precise laboratory practices. Given this heterogeneity among the experimental studies and difficulties in model parameterization due to lack of experimental data for key pollutant uptake for many widespread species, we feel that the current knowledge of the contribution of NBS in air phytoremediation is insufficient.

4.2. Future directions

To compensate for the lack of information it would be necessary (i) to advance in targeted species screening, to evaluate the potential of NBS in air phytoremediation, and (ii) to establish a methodology that allows a full control of the experimental environment with the possibility to replicate the ambient environment. In addition, the experimental setup should allow continuous collection of data in real time on the modifications and performance of mature plants. Plants modulate their physiological responses based on the environment around them and the stresses to which they are subjected (Papazian and Blande, 2018). The use of engineered plants, to enhance the pollutants removal and also increase plant tolerance has been explored and need to be further researched (Soreanu et al., 2013; Weyens et al., 2015; Kim et al., 2018). To obtain realistic data of the potential of NBS, one cannot ignore the plant organism in its complexity, including genetic, epigenetic, and physiological components, and we cannot ignore the plant BVOC emissions (Niinemets et al., 2011). Due to the major gaps in knowledge of NBS, the search for the most suitable plants for phytoremediation is still ongoing. Especially for the outdoor environments, assessment of species potentials for air phytoremediation is difficult as this requires consideration of the trade-off between ecosystem services and disservices.

Moreover, understanding the interaction dynamics between plant organisms and the entire pool of pollutants present in each environment therefore represents a very important step in obtaining a reliable assessment of the performance of NBS (Papazian and Blande, 2018). Once these limitations have been addressed, a new NBS-research concept could be achieved. This would allow determination, in real time and in a natural environment, the balance between ecosystem services and disservices and estimation of NBS benefits in air phytoremediation.

Author contribution

All authors make substantial contributions to conception and design of the review. AP and DZ provided acquisition of data. ÜN and CG supervised data acquisition. AP structured the database and DZ performed the statistical data analysis. All authors jointly interpreted data. DZ and AP wrote the first draft of the manuscript. All authors contributed to manuscript critical evaluation, revision, read, and approved the submitted version. s.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.116817.

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