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Wild and ruderal plants as bioindicators of global urban pollution by air, water and soil in Riyadh and Abha, Saudi Arabia

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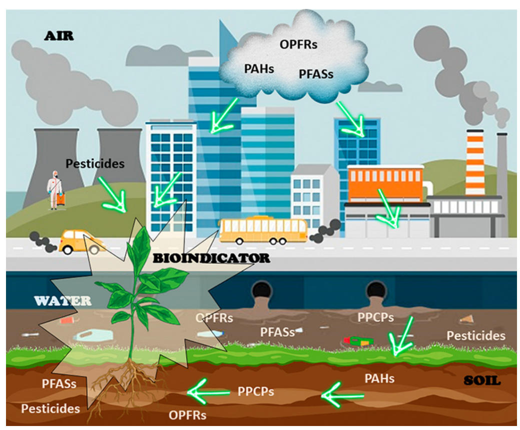
HIGHLIGHTS

- First wild plants assessment as bioindicators for many types of organic contaminants.
- PAHs were detected in all samples at highest concentration.
- PPCPs, PFASs, Pesticides and OPFRs were at lower concentration.
- Detection of contaminants of all groups indicates their potential as bioindicator.

GRAPHICAL ABSTRACT



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ABSTRACT

Recently, environment pollution around the globe has increased because of anthropogenic activities. As part of the biota, plants can assimilate the compounds present in air, water and soil and respond to changes in surround conditions, for that, they can be used as bioindicators of global pollution. However, urban plants' ability to monitor organic pollutants in air, soil, and water have not been profoundly studied yet. Anthropogenic contamination produced by five different types of pollutants [polycyclic aromatic hydrocarbons (PAHs), pharmaceuticals and personal care products (PPCPs), perfluoroalkyl substances (PFASs), pesticides and organophosphorus flame retardants (OPFRs)] has been studied in Riyadh and Abha areas (Saudi Arabia). In addition to the points in both cities, a control point located in the Asir National Park (close to Abha), which is little affected by human activity, was used. The 5 groups of contaminants were found with different but high detection frequencies from 85 % to 100 % in wild and ruderal plants. PAHs were detected in all the analyzed samples at the highest average sum of concentrations (ΣPAHs) 1486 ng·g^{-1} dry weight (d.w.). Statistically significant differences were obtained between Riyadh, Abha and the point located in the national park ($p < 0.05$). ΣPAHS in Riyadh >> ΣPAHs in Abha > ΣPAHs in the National Park. Values of the average sum of concentrations for the other groups of contaminants ΣPPCPs, ΣPFASs, Σpesticides and ΣOPFRs were 420.5, 171, 48 and 47 ng g^{-1} d.w., respectively. High values of PPCPs are due to the presence of salicylic acid. Differences in the average sum of each type of contaminant concentrations between cities were not statistically significant. The results of this assessment of wild and ruderal plants as bioindicators for 5 types of organic contaminants suggest that they can be used to monitor anthropogenic contaminants in the terrestrial environment.

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1. Introduction

Anthropogenic pollution can assume different forms and impact on all environmental compartments (Häder et al., 2020; Nabi et al., 2018). A high number of persistent organic compounds, such as perfluoroalkyl substances (PFASs), organophosphorus flame retardants (OPFRs) and polycyclic aromatic hydrocarbons (PAHs) as well as pseudopersistent, such as pharmaceuticals and personal care products (PPCPs), have been widely reported in the environment (Chaturvedi et al., 2021; Lee et al., 2021; Ripanda et al., 2022; Tong et al., 2022; Valdez-Carrillo et al., 2020; Yusuf et al., 2021). Their occurrence has been well-established in matrices, such as air (Enyoh et al., 2020), wastewater (Parida et al., 2021), seawater (MacKeown et al., 2022), lakes, rivers and wetlands (Galindo-Miranda et al., 2019), sediments (Kuznetsova and Timerbaev, 2022), aquatic biota (Świacka et al., 2022), soils (Vitale et al., 2023) and terrestrial organisms (Zhang et al., 2021). The negative effects of pollutants on different ecosystems were also described (Agathokleous et al., 2022; Nilsen et al., 2019; Yusuf et al., 2021) covering a wide biological range, from alterations and imbalances in ecological communities to nephrotoxic and hepatotoxic effects, including genotoxicity and cytotoxicity.

Densely populated urban areas are a source of pollutants and involve higher exposure of humans and other living organisms (Chaturvedi et al., 2021; Tong et al., 2022; Zhang et al., 2022). Most of the studies on pollution in urban areas are focused on the urban water cycle (Häder et al., 2020; Tong et al., 2022). However, soil and gardens, flower strips, field boundary and hedges vegetation accumulated low levels of pollutants caused by urban activity [air deposition of metals and other chemicals like PAHs, treatments with pesticides, irrigation with treated or untreated wastewater or infiltrations containing high levels of pharmaceuticals and other contaminants] (Yusuf et al., 2021; Zhang et al., 2022). These studies are in focus for further investigation due to the evidence of human exposure in addition to their environmental consequences. Tracking pollution through living organisms, such as plants (known as bioindicators), can be a useful approach to environmental monitoring in accordance with the One Health strategy, which encourages multi-sectorial risk assessments at the human-animal/biota-environment interface (Dao and Fu, 2021; Messmer, 2020; Ogunseitan, 2022). Plants can uptake pollutants after aerial deposition and those present in soil by deposition or irrigation, given for example a better idea of the living organisms' exposure and coming as an important approach for the one health system.

The use of several wild plants as bioindicators and/or to biomonitor pollutants backs to the 90's (Weinstein et al., 1990). Since then, plants have been widely used for identifying heavy metal and inorganic pollution, especially in interior and coastal wetlands, but also in other terrestrial ecosystems because they are easy to identify and widely available (Ali, 1993; Azzazy, 2020; Burger et al., 2007; Charles et al., 2011; Eid et al., 2021a; Eid et al., 2021b; Fidalgo Hijano et al., 2005; Gregson et al., 1994; Krizkova et al., 2008). Plants have also been reported to monitor organic pollution because their capacity to absorb contaminants from soil, water and after aerial deposition (Ernst, 2003; Klumpp et al., 1994). Regarding contaminants, PAHs have demonstrated to be ubiquitously present in soils and plants of urban areas (Chaplygin et al., 2022; Jia et al., 2021). Zhang et al. (2022) reported that the concentration of PAHs in 120 paired plant and soil samples, collected in the highly populated Yangtze River Delta (China), were 2407.92 ng g^{-1} and 546.64 ng g^{-1}, respectively, which were significantly higher than those in the rural areas. Going a step ahead, Wang et al. (2022) obtained information on the degradation constant for each PAH under investigation through the content of PAHs in *Abies holophylla* and *Pinus tabuliformis* needle samples. Pesticides are sprayed in row crops, they can contaminate wild plant because a certain amount of the pesticides reaches non-target areas (especially, soil or the ground-cover vegetation) due to spray drift and foliar runoff. This phenomenon has been widely reported for neonicotinoids insecticides (Botías et al., 2016; García-Valcárcel et al., 2022). Several studies, are focused on the uptake, bioaccumulation, metabolism and effects of PPCPs in plants and crops after reusing treated wastewater for their irrigation (Bigott et al., 2022; Bueno et al., 2022; Manasfi et al., 2021; Picó et al., 2019; Picó et al., 2018a). For example, Bueno et al. (2022) revealed the presence of 18 PPCPs in tomato at the average load of 5.8 μg kg^{-1} after three months to be irrigated with a mixture of PPCPs at approx. 1 μg L^{-1}. Picó et al. (2018a) identified 46 metabolites of ibuprofen in treated plants and in a further study detected few of them formed under real environmental conditions. To a lesser extent, bioaccumulation and distribution of PFASs in plants (Xu et al., 2022) and their potential as biosamplers of OPFRs (Wang et al., 2021) were also established.

The purpose of this study was to investigate and compare plants growing in Riyadh (large industrial city, 7,387,817 million inhabitants, intense traffic) and in Abha (small city, < 1,093,705 inhabitants, also intense traffic, near a national park), as bioindicators of anthropic pollution. To that end, an analytical approach based on several extraction protocols, followed by ultra-high-performance liquid-chromatography with tandem mass spectrometry (UHPLC-MS/MS) or gas chromatography–mass spectrometry (GC–MS) was applied to determine the presence of 30 PPCPS, 60 pesticides, 21 PFASs, 11 OFRs and 16 PAHs. The wild and ruderal plants growing on natural regions, roadsides, gardens and hedges from 5 and 3 different places of South Riyadh and Abha, respectively, were monitored from June to September 2019. Seventy-two samples from 25 species were analyzed to demonstrate if they were suitable indicators of pollution by the selected organic pollutants. This work is (up to our knowledge) a first attempt to make a comprehensive study on the capability of plants in urban areas to monitor a big diversity of organic pollutants in different media: air, soil and water. As far as we are aware, only some of the emerging contaminants studied here have been reported in very few samples of wild plants in Saudi Arabia (Picó et al., 2020; Picó et al., 2021).

2. Experimental

2.1. Reagents

The analytical standards of the target compounds (21 PFASs, 11 OPFRs, 60 pesticides, 40 PPCPs, 16 PAHs) were purchased from Sigma-Aldrich (Steinheim, Germany), Wellington Laboratories (Guelph, Ontario, Canada) and LCG standards (Barcelona, Spain) (detailed lists of unlabeled and labeled analytical standards –used as surrogated standards- are outlined in Tables S1 and S2, respectively). Pesticides and pharmaceuticals were acquired as analytical standards (>99.9 % purity), PFASs and OPFRs as analytical standard solutions and PAHs as mix solutions. Appropriate mixture and dilution of these compounds was performed for method's revalidation and to ensure quality assurance. Individual stock and working standard solutions were stored at −20 and 4 °C in the dark, respectively.

2.2. Characteristics of sampling sites and sample collection

Riyadh city (24°39′N 46°43′E) is in the country's center, on the eastern part of the Najd plateau at an average of 600 m above the sea level, the city had a population of ca. 7.4 million people in 2021, making it the most-populous city in Saudi Arabia. This city exhibits a larger temperature variation between winter (coldest month January with temperatures averaging 14.0 °C) and summer (warmest month is July, with an average temperature of 36.1 °C) in comparison to other cities in Saudi Arabia. Its deserted climate is characterized by arid and dry conditions with minimal humidity (from 0 to 10.3 %), particularly during summers, leading to a parched and dusty environment. The annual precipitation levels are low, averaging 6 mm, and occur mainly during a few intense rain events in early spring (mostly in April).

Abha (18°13′1″N 42°30′19″E) is located on the south-west in the hilly area at an elevation of 2270 m and surround by the Asir National Park. Most common vegetation are Acacia woodlands and thorn scrub. With a population of 1,093,705 in 2021, it has the 6th largest population in Saudi Arabia. The climate in Abha is both cooler and wetter, semi-arid and influenced by city's high elevation. Months with the largest precipitation are April, March, May with 6 mm precipitation. The average annual temperature is 26 °C. The warmest month of the year is June, with an average temperature of 31 °C. Usually, January is the coldest month in Abha, with an average temperature of 20 °C.

The study area was sampled between June and September in 2019. In Riyadh, the studied area correspond to the second industrial city, delimited by the southern ring road, the eastern ring road and the Alkharj road with a total area of >18,000,000 m^2 with around 1050 factories. This area was selected because of its heavy traffic, the presence of treated water from the city discharged to a recreational area and the high levels of pollution due to industry. The samples were taken at a cement factory (4), the South ring road (5), a plastic factory (7), the Eastern Ring Road (8) and Alkharj roadside (6). Samples in Abha were taken at two points (west and east) on King Abdul Aziz Road, a ring road that runs for about 8 km through the heart of the city and connects several neighborhoods (2 and 3) and at Wadi Bani Mazen (Asir National Park) (1) (Fig. 1). The points taken in the city ring were selected to compare the two cities, as both areas have the most intense traffic and are affected by various human activities. The point taken in the Asir National Park serves as a control, representing an area with minimal human influence. Sampling stands' geographic coordinates and description together the species collected at each sampling point are detailed in Table 1. Sample stands covered an area of at least 1800 m^2. Each selected stand was divided in 10 plots depending on the area to help to select the most abundant species. Before plant sampling, a visual evaluation was carried out in each stand to determine the species present. Wild and ruderal plant species were collected when the same species was present in at least 5 plots in each sampling stand. Fifty-three plant species (mean ± SD: 9.1 ± 5.0 species per stand) were collected in natural regions, roadsides, gardens, margins and hedges choosing a variety of species representing different plant types. The sample consists of 30 g of each plant species at each stand (from 2 to 5 plants depending on the species) stored in plastic polyethylene bags. The number and species of plants collected in each sampling stand were not always the same because it depends upon which species were available. Once in the laboratory, the wild and ruderal plant samples were cleaned with distilled water, air-dried, freeze-dried and stored in polyethylene bags to facilitate transport to the analytical laboratory in Spain without breaking the cold chain.

2.3. Extraction, determination and quality assurance

The extraction methods for emerging contaminants were already reported (Picó et al., 2020; Picó et al., 2021; Picó et al., 2018b). Briefly, pesticides were extracted using a citrate buffered QuEChERS, in which 2 g of dried samples were added with 8 mL of water, followed by dispersive solid-phase extraction (SPE) with a magnesium sulfate, primary-secondary amine (PSA) and octadecylsilice (C_{18}). PFASs, OPFRs and PPCPs were extracted using 5 mL of methanol and 5 mL of Mc. Ilvain buffer (pH 5.6) plus 0.2 M in ethylenediaminotetracetic acid (EDTA) followed by SPE with Strata-X cartridges and elution of the analytes retained in the cartridge with methanol.

The determination of pesticides, PFASs, OPFRs and PPCPs was carried out using an LC-MS/MS system equipped with a 1260 Infinity Ultra-High-Performance Liquid Chromatograph (UHPLC) and an Agilent 6410 Triple Quadrupole (QqQ) Mass Spectrometer (MS/MS) with an electrospray ionization (ESI) source (Agilent Technologies, Santa Clara, CA, USA). Compounds were separated using conventional reverse-phase chromatography and determined in either positive (pesticides, some PPCPs and OPFRs) and negative (PFASs and some PPCPs) ionization as previously detailed.

Methods' validation parameters were already detailed in the supplementary material of previous publications. Briefly, QuEChERS method was validated in three vegetable matrices apples, oranges and dates (similar to plants in this study) obtaining recoveries from 70 to 120 % for most of the compounds with RSDs <31 % with appropriate sensitivity as limits of detection (LODs) and quantification (LOQs) were from 1 to 10.5 ng g^{-1} d.w. and from 2.5 to 21 ng g^{-1} d.w., respectively. Matrix effect ranged from −55 to 19 % for apples, from −60 to 55 % for oranges and from −60 to 35 % for dates (Picó et al., 2018b). The method based on Mc. Ilvaine buffer and SPE provided recoveries ranging from 32 to 112 %, RSDs ranging from 6 to 19 % and LOQs ranging from 0.06 to 30 ng g^{-1}. Matrix effects were observed ranging from −80 to 141 %. For 70 % of the compounds these effects were between −30 % and +30 % that could be considered negligible (Picó et al., 2021).

The method to extract PAHs was developed and optimized for this study based on the method reported by Martinez et al. (2004). Briefly, lyophilized

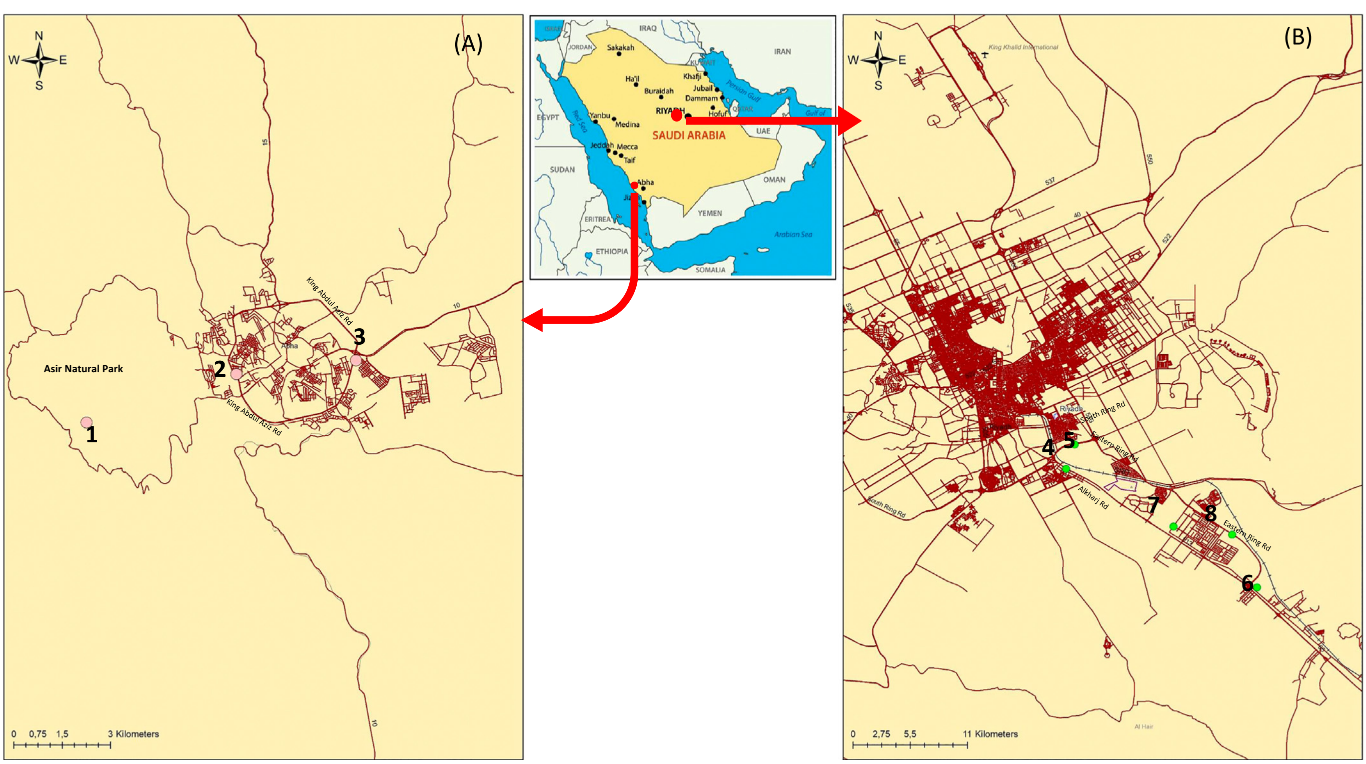

Fig. 1. Maps of the studied areas showing the stands in (A) Asir National Park and Abha city (from 1 to 3) and (B) South Riyadh (from 4 to 8).

Table 1
Plant species collected in the sampling stands.

City and sampling point	Species
Abha area–Date 19-6-2019	
Stand 1. Abha Wadi Bani Mazen	*Malva parviflora*
(N18°11′58.94″; E42°26′51.48″)	*Sencio* sp.
Asir Natural Park	*Juniperus phoenicea*
Slope vegetation in a wadi with little or not anthropic	*Asparagus officinalis*
disturbation (ca. 1000 × 1000 m)	*Dodonaea viscosa*
Irrigation: only rain	*Lavandula* sp.
	Organum vulari
Stand 2. Abdul Aziz Road	*Acacia laeta*
(N18°13′39.28″, E42°29′21.37″)	*Juniperus phoenicea*
Abha city	*Asparagus officinalis*
Garden in the city (Damside Park) (ca. 80 × 80 m)	*Barleria cristasa*
Irrigation: treated wastewater	*Psiadia punctulata*
	Rhmanus disperma
	Conyza sp.
	Solanum sp.
	Ipomoea aquatica
	Erigeron canadensis
	Dodonaea viscosa
	Moringa oleifera
	Bougainvillea glabra
	Catharanthus roseus
	Hibiscus rosa-sinensis
	Schefflera arboricola
Stand 3. Abdul aziz Road	*Dodonaea viscosa*
(N18°13′1.43″, E 42°31′22.26″)	*Cenchrus ciliaris*
Abha city	*Cadia purpurea*
Vacant lots (ca. 120 × 120 m)	*Ficus* sp.
Irrigation: no information	*Hibiscus rosa-sinensis*
	Grewia sp.
	Rosa abyssinica
	Segeratia thea
	Cucumus
	Conyza linifolia
	Solanum sp.
	Lichen on trees of
	Juniperus phoenicea
Riyadh city 6-9-2019	
Stand 4. Cement factory-south Riyadh city (N24° 36′ 16.34″ E 46° 46′ 41.7018″)	*Digitaria ciliaris*
	Ocimum bacilicum
Hedgerows in front of the cement factory (ca.	*Salsola imbricata*
300 m × 30 m).	*Conocarpus erectus*
Irrigation: treated wastewater	*Tamarix aphylla*
	Prosopis juliflora
	Typha domingensis
	Portuaca oleracea
	Calotropis procera
	Plachea sp.
	Ficus sp
	Alhagi graecorum Boiss
Stand 5. Heavy traffic –Riyadh city south ring road (N24° 37′ 32.568″ E46° 47′ 8.681″)	*Phramites australis*
	Atriplex leucoclada
Vacant lot (ca. 60 × 60 m)	*Prosopis juliflora*
Irrigation: no information	*Conocarpus erectus*
	Ficus sp.
Stand 6. Riyadh city Heavy traffic –Riyadh-Alkharj road (N24° 36′ 45.720″ E46° 49′ 19.548″)	*Conocarpus erectus*
Roadside hedges in front of a wastewater treatment plant (ca. 300 × 6 m)	
Irrigation: no information	
Stand 7. Riyadh city industrial area II (N24° 33′ 16.829″ E46° 52′ 17.040″)	*Prosopis juliflora*
	Phragmites australis
Sidewalk vegetation near of a plastic fabric facility (ca.	*Ecylptus australis*
500 × 5 m)	*Portuaca oleracea*
Irrigation: treated wastewater	*Zyzyphus spine-christi*
	Conocarpus erectus
	Atriplex leucoclada
	Pulicaria crispa
	Zygophillum coccinerum
	Fagonia indica
	Amaranthus grazineus
	Trichodesma africanum
	Sesuvium verrucosum
Stand 8. Riyadh city. Riyadh city industrial area II (N24° 32′ 51.947″ E46° 55′ 19.601″)	*Conocarpus erectus*
	Hibiscus tiliscum
Roadside hedges of the Eastern Ring Road (ca. 500 × 4 m)	*Vitex agnus-castus*
Irrigation: no information	*Nerium oleander*
	Calotropis procera
	Salsola imbricata

plants (0.5 g) were extracted by 20 min ultrasonication with 3 × 20 mL of a mixture of hexane-dichloromethane (1:1, v/v). The resulting extracts were cleaned-up by solid phase extraction (SPE) using neutral alumina (Supelclean™ LC-Alumina-N SPE Tube bed wt. 2 g, volume 6 mL), reconstituted in 250 μL hexane. PAHs were eluted with 40 mL of n-hexane/dichloromethane (1:1, v/v), and then, the eluate was converted to hexane and concentrated to approximately 250 μL.

PAHs were analyzed using GC–MS JASCO Series YL6900. PAHs separation was achieved using a capillary column Zebron ZB-5MS of 30 m × 0.25 mm i.d. with 0.25 μm film thickness from Phenomenex (Torrance, CA, USA). The mass spectrometer was operated in the electron ionization mode with an ionizing energy of 70 eV acquiring 3 characteristic ions per compound (GC–MS conditions and ions selected are summarized in Table S3). Quantitation, based on peak areas, was performed by the internal standard method using surrogate deuterated standards. This method allows the determination of the 16 PAHs at ng g^{-1} d.w. levels (LODs between 0.5 and 5 ng g^{-1}) with acceptable precision (RSDs calculated from 3 replicates below 30 %) and accuracy (recovery percentage above 45 % for all compounds except naphthalene ca. 22 %, results shown in Table S4).

In order to minimize laboratory contamination, only glass and metal equipment was used. All glassware was cleaned by ultrasonic agitation in water containing detergent and rinsed with ultrapure water and high-purity methanol. The solvents were purchased from vwr, Germany. Analytical and procedural blanks were tested periodically to elude false positive whereas addition of isotopically labeled internal standards and periodic analysis of control samples (spiked at know concentrations) was implemented to avoid false negatives.

2.4. Statistical analysis

As most wild plant species were not present in all the sampling sites, the influence of the specie was not statistically analyzed. Instead, concentrations were compared to the location –Riyadh, Abha, Wadi Bani Marzen (Asir National Park)– and with the sampling stand. The Kolmogorov-Smirnov ($n > 50$) or Shapiro–Wilk ($n < 50$) tests were used to test for normality and Levene's test for homogeneity of variances. Given the lack of normality of the parametric analysis, differences in concentrations were established through the tests of Mann–Whitney U (M–W) or Kruskal–Wallis (K—W), considering places (Riyadh, Abha and Wadi bani Marzen) or sampling stands (1 to 8) and compound's family (OPFRs, Pesticides, PFASs, PPCPs, PAHs) as factors. Results were statistically significant at $p < 0.05$. All statistical analyses were carried out using IBM SPSS Statistics 26®.

3. Results and discussion

Table 2 shows minimum, maximum and average concentration of each contaminant in Riyadh South, Abha and the Wadi Bani Marzen (Asir National Park). Fig. 2 summarizes these results as the average of the sum of concentrations for each type of contaminant at each sampling stand. Fig. 3 illustrates the spatial distribution of the contaminants and Fig. 4, the pattern of each type of contaminant (grouped according to their physicochemical properties as shown in the legend and detailed in Table S5). The influence of plant species on contaminant content cannot be assessed because the plant species are very diverse and the differences in the content can be attributed to other many factors not controlled in this study (the mean value of each type of pollutant in plant species taken from the same city is given in Table S6). The most notable similarity, in terms of plant species, is that *Conocarpus erectus* is present in all five stands in Riyadh, while

Table 2
Minimum, Maximum and average concentrations of each contaminant as well as frequency in Riyadh South, Abha and Asir National Park.

	Riyadh (n = 37)				Abha city (n = 28)				Asir National Park (n = 5)			
	Min	Max	Average	Frequency	Min	Max	Average	Frequency	Min	Max	Average	Frequency
PAHS												
Naphthalene (Naph)	245.6	384.2	305.0	37.0	89.4	152.1	117.4	28.0	26.2	51.5	38.2	7.0
Acenaphtylene (Acy)	9.4	23.4	16.3	37.0	<LOD	9.2	4.5	20.0	<LOD	5.0	1.7	4.0
Acenaphthene (Ace)	2.3	9.2	6.1	37.0	<LOD	5.4	1.9	21.0	<LOD	1.5	0.5	3.0
Fluorene (Flu)	97.2	192.4	131.8	37.0	38.2	89.4	53.9	28.0	12.4	21.5	18.4	7.0
Phenanthrene (Phe)	448.2	612.8	563.9	37.0	201.4	301.4	276.0	28.0	89.5	132.5	112.5	7.0
Anthracene (Ant)	126.5	202.5	162.4	37.0	72.4	125.6	96.5	28.0	40.2	57.5	48.8	7.0
Fluoranthene (Fla)	325.2	589.2	481.3	37.0	167.6	289.4	198.4	28.0	78.3	96.5	86.9	7.0
Pyrene (Pyr)	256.8	362.9	291.3	37.0	98.4	202.1	134.5	28.0	40.2	60.2	49.5	7.0
Benzo(*a*)anthracene (BaA)	<LOD	4.2	0.9	19.0	<LOD	3.8	1.3	21.0	<LOD	1.5	0.4	2.0
Chrysene (Chr)	13.4	75.9	59.5	37.0	31.2	85.2	47.3	28.0	2.6	10.0	5.7	7.0
Benzo(*b*)fluoranthene (BbF)	<LOD	2.8	0.9	22.0	–	–	–	–	–	–	–	–
Benzo(*a*)pyrene (BaP)	36.2	102.5	74.0	37.0	17.6	42.9	27.5	28.0	<LOD	8.2	3.9	6.0
Dibenz[*a*,*h*]anthracene (DahA)	1.4	7.2	3.5	37.0	<LOD	3.0	0.1	1.0	<LOD	0.5	0.1	2.0
Benzo[*g*,*h*,*i*]perilene (BghiP)	<LOD	1.4	0.3	11.0	–	–	–	–	–	–	–	–
ΣPAHs	1759.2	2294.2	2097.2	37.0	847.1	1148.0	959.1	28.0	332.9	443.9	366.6	7.0
Pesticides												
Imidacloprid	<LOD	1.2	0.0	1.0	–	–	–	–	<LOD	1.9	0.9	4.0
Atrazine-Deisopropyl	<LOD	12.9	5.3	23.0	<LOD	12.9	8.4	13.0	<LOD	5.3	2.7	1.0
Clothianidin	<LOD	5.6	0.2	1.0	–	–	–	–	–	–	–	–
DMPF	<LOD	152.7	16.2	16.0	<LOD	14.6	2.7	5.0	–	–	–	–
Carbofuran-3-OH	<LOD	7.8	0.5	2.0	–	–	–	–	–	–	–	–
DMA	<LOD	36.6	1.9	2.0	<LOD	37.9	2.5	1.0	–	–	–	–
Azinphos-Methyl	–	–	–	–	<LOD	32.1	2.5	4.0	–	–	–	–
Parathion-methyl	<LOD	2.9	0.1	1.0	–	–	–	–	–	–	–	–
Alachlor	<LOD	383.9	11.9	2.0	–	–	–	–	<LOD	6.8	3.4	1.0
Fipronil	<LOD	5.9	0.4	3.0	–	–	–	–	–	–	–	–
Chlorfenvinphos	–	–	–	–	<LOD	5.0	0.3	1.0	–	–	–	–
Coumaphos	–	–	–	–	<LOD	4.4	0.8	3.0	–	–	–	–
Tolclofos-methyl	<LOD	1002.7	29.5	1.0	<LOD	175.9	40.9	9.0	4.5	10.5	7.5	2.0
Spinosad C	–	–	–	–	<LOD	7.5	0.5	1.0	–	–	–	–
Buprofezine	<LOD	62.4	1.8	1.0	–	–	–	–	<LOD	1.2	0.6	1.0
Pyriproxyfen	<LOD	7.2	0.2	1.0	<LOD	3.1	0.2	1.0	–	–	–	–
Dichlofenthion	–	–	–	–	<LOD	3.2	0.2	1.0	–	–	–	–
Ethion	–	–	–	–	<LOD	13.5	0.9	1.0	–	–	–	–
Cyhalothrin	<LOD	6.5	0.3	2.0	–	–	–	–	–	–	–	–
Acrinathrin	–	–	–	–	<LOD	8.1	0.9	2.0	–	–	–	–
Fluvalinate	<LOD	4.1	0.2	2.0	<LOD	10.1	0.7	1.0	–	–	–	–
ΣPesticides	<LOD	1508.6	68.6	30.0	10.2	195.3	61.6	15.0	9.8	20.4	15.1	2.0
OPFRs												
TCEP	<LOD	31.6	4.5	16.0	<LOD	94.5	17.5	14.0	<LOD	48.3	10.5	2.0
TPP	<LOD	19.0	0.6	2.0	<LOD	87.8	10.6	14.0	<LOD	29.1	4.2	1.0
TClPP	<LOD	70.6	18.4	23.0	<LOD	145.1	6.8	3.0	–	–	–	–
TDClPP	–	–	–	–	<LOD	32.2	4.2	13.0	–	–	–	–
TPhP	<LOD	8.9	0.2	1.0	<LOD	67.8	5.8	8.0	<LOD	19.7	2.8	1.0
TDBPP	<LOD	37.3	1.1	2.0	<LOD	184.2	14.3	3.0	–	–	–	–
TnBP	<LOD	25.4	3.1	20.0	<LOD	110.8	6.6	7.0	–	–	–	–
CDP	<LOD	22.1	4.0	16.0	<LOD	187.9	11.1	4.0	–	–	–	–
TMPP	–	–	–	–	<LOD	356.6	13.3	2.0	–	–	–	–
ΣOPFRs	<LOD	106.0	32.0	35.0	<LOD	653.0	90.2	25.0	0.0	97.1	17.5	2.0
PFASs												
PFBA	<LOD	341.2	12.3	8.0	<LOD	9.6	0.7	3.0	<LOD	0.0	0.0	0.0
PFPeA	<LOD	1887.9	133.3	22.0	<LOD	887.3	76.7	18.0	<LOD	120.4	20.0	3.0
PFBS	<LOD	9.1	0.4	5.0	<LOD	8.7	0.7	4.0	<LOD	0.0	0.0	0.0
PFHxA	<LOD	17.1	0.9	4.0	<LOD	76.0	9.0	14.0	<LOD	113.6	33.1	3.0
PFHpA	<LOD	21.3	1.4	11.0	<LOD	3204.7	197.0	12.0	<LOD	110.7	16.9	2.0
PFHxS	<LOD	14.8	0.5	8.0	<LOD	1.6	0.1	3.0	–	–	–	–
PFOA	<LOD	15.3	0.5	13.0	<LOD	9.4	0.8	7.0	–	–	–	–
PFHpS	<LOD	21.7	3.4	18.0	<LOD	18.9	0.9	2.0	–	–	–	–
PFOS	<LOD	4.9	0.8	12.0	<LOD	1.7	0.1	3.0	<LOD	0.7	0.1	1.0
ipPFNA	–	–	–	–	<LOD	2.1	0.1	1.0	–	–	–	–
PFNA	–	–	–	–	<LOD	0.9	0.0	1.0	–	–	–	–
FOUEA	–	–	–	–	<LOD	2.6	0.1	1.0	–	–	–	–
ipPFNS	<LOD	78.5	2.2	5.0	<LOD	0.4	0.0	3.0	–	–	–	–
PFDA	<LOD	0.7	0.0	2.0	<LOD	4.8	0.2	1.0	–	–	–	–
ΣPFASs	<LOD	1892.7	155.7	35.0	0.0	3227.6	286.5	23.0	<LOD	215.1	70.0	3.0
PPCPs												
Salicylic acid	<LOD	6604.8	670.3	35.0	<LOD	9954.1	2030.3	22.0	302.1	2204.8	1147.9	7.0
Thiamphenicol	<LOD	109.1	4.0	6.0	<LOD	0.0	0.0	0.0	–	–	–	–
Furosemide	<LOD	655.4	33.5	11.0	<LOD	168.7	21.8	6.0	<LOD	90.1	26.6	3.0
Chloramphenicol	<LOD	20.0	0.5	1.0	<LOD	0.0	0.0	0.0	–	–	–	–
Methylparaben	1.3	1271.4	109.4	37.0	<LOD	953.8	110.8	19.0	14.4	147.4	73.9	7.0
Ethylparaben	<LOD	21.3	3.9	15.0	<LOD	8.1	1.1	6.0	<LOD	3.0	0.4	1.0

(continued on next page)

Table 2 (*continued*)

	Riyadh (n = 37)				Abha city (n = 28)				Asir National Park (n = 5)			
	Min	Max	Average	Frequency	Min	Max	Average	Frequency	Min	Max	Average	Frequency
Naproxen	<LOD	2.9	0.1	1.0	–	–	–	–	–	–	–	–
Benzafibrate	<LOD	5.9	0.2	1.0	<LOD	42.9	6.5	7.0	–	–	–	–
Propyparaben	<LOD	4.6	0.5	6.0	<LOD	6.5	1.2	8.0	<LOD	7.1	2.7	4.0
Warfarin	<LOD	2.6	0.1	1.0	<LOD	13.6	0.6	2.0	–	–	–	–
BPA	<LOD	1608.5	94.7	6.0	<LOD	200.8	9.3	7.0	<LOD	5.2	0.7	1.0
Diclofenac	<LOD	686.8	92.1	17.0	<LOD	287.7	20.9	6.0	<LOD	129.7	20.6	3.0
Indomethacin	–	–	–	–	<LOD	3.3	0.1	1.0	–	–	–	–
Ibuprofen	<LOD	1214.1	59.5	9.0	–	–	–	–	–	–	–	–
Metformin	<LOD	28.9	3.9	13.0	–	–	–	–	–	–	–	–
Atenolol	<LOD	1010.1	142.3	9.0	<LOD	2069.1	235.9	8.0	–	–	–	–
Codeine	<LOD	21.6	0.6	1.0	<LOD	13.0	0.5	1.0	–	–	–	–
Ofloxacin	<LOD	387.0	10.5	1.0	<LOD	0.0	0.0	0.0	–	–	–	–
Paracetamol	<LOD	14.0	2.5	12.0	<LOD	25.6	1.8	3.0	–	–	–	–
Tramadol	<LOD	233.6	18.0	5.0	<LOD	2.9	0.1	1.0	–	–	–	–
Caffeine	<LOD	42.4	8.2	32.0	<LOD	4128.2	192.4	11.0	–	–	–	–
Etoricoxib	<LOD	8.3	0.2	1.0	–	–	–	–	–	–	–	–
Enelapril	<LOD	203.7	5.5	1.0	–	–	–	–	–	–	–	–
Lorazepam	<LOD	194.7	5.3	1.0	–	–	–	–	–	–	–	–
Simvastatin	<LOD	543.6	14.7	1.0	<LOD	279.2	10.0	1.0	–	–	–	–
ΣPPCPs	21.5	2363.0	572.1	37.0	<LOD	5496.0	591.2	25.0	19.6	189.6	98.4	7.0

Dondonea viscosa is found in the Abha stands and in the Asir National Park stand (for comparison of the results see Tables S7 and S8). Although some trends in the case of PAHs and PPCPs can be envisaged, the low sample size prevent to perform a statistical treatment and then, further studies would be needed.

The five types of contaminants were found in wild and ruderal plants. Their sum of concentrations (Σ) indicate that ΣPAHs>>ΣPPCPs> ΣPFASs>ΣPesticides>ΣOPFRs.

PAHs are ubiquitous in the environment because they originate mostly from combustion processes and are released into the atmosphere. PAHs reach plants mostly by atmospheric deposition, although they can also reach them through roots that absorb those deposited in the soil. The ΣPAHs concentrations in the plants ranged from 333 to 2294 ng g^{-1} dry weight (d.w.) with an average of 1486 ng g^{-1} d.w. Contaminants levels in plants were higher in industrial and traffic heavily areas such as Riyadh South (average ΣPAHs of the 5 sampling points 2097 ng g^{-1} d.w.) compared to those growing less populated and less industrialized cities, such as Abha (average ΣPAHs of the 2 sampling points 959 ng g^{-1} d.w.) ($p < 0.05$). However, PAH levels are also higher in these smaller cities than in natural areas little altered by humans, such as Asir National Park (average ΣPAHs of the sampling point 367 ng g^{-1} d.w.) ($p < 0.05$). The levels reported in Riyadh agree with those reported by Zhang et al. (2022) in urban and periurban crops (2407.92 ng g^{-1} d.w.) growing in the Yangtze River Delta urban agglomeration in China. The authors also

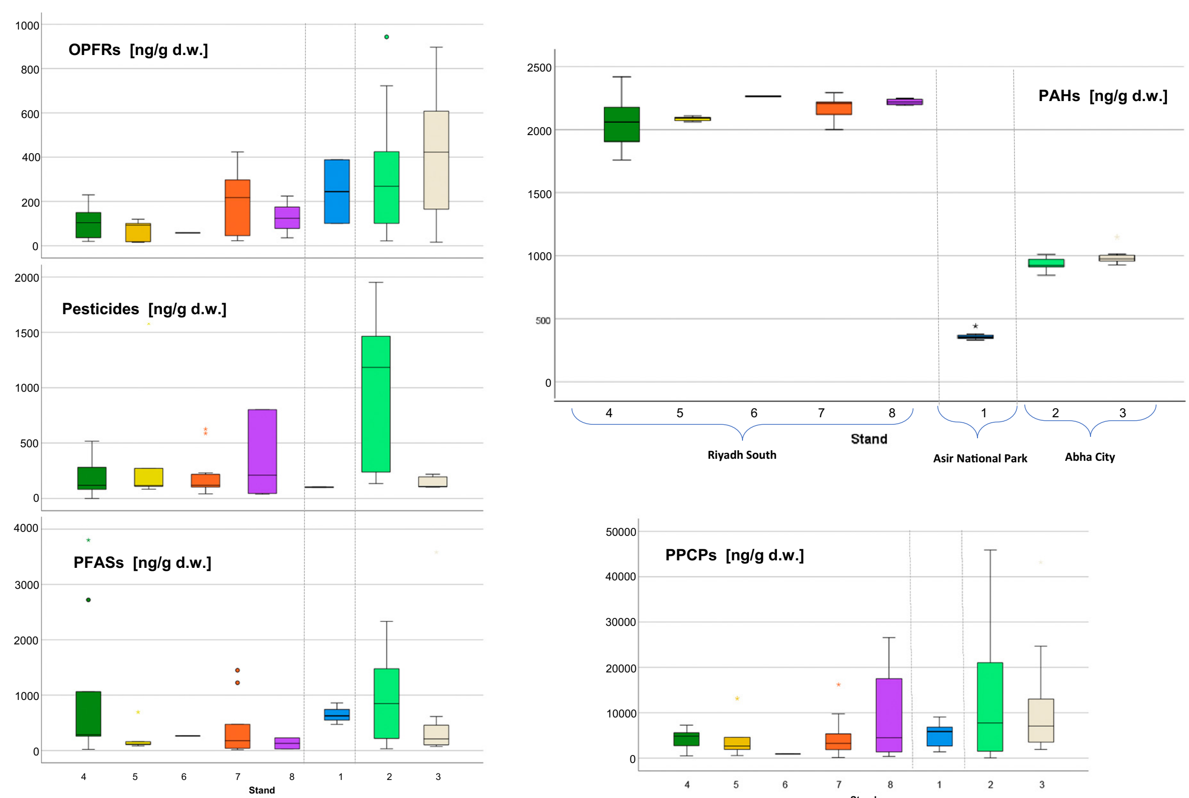

Fig. 2. Average sum of concentrations of each type of contaminant found in each stand at Riyadh, Abha and Asir National Park.

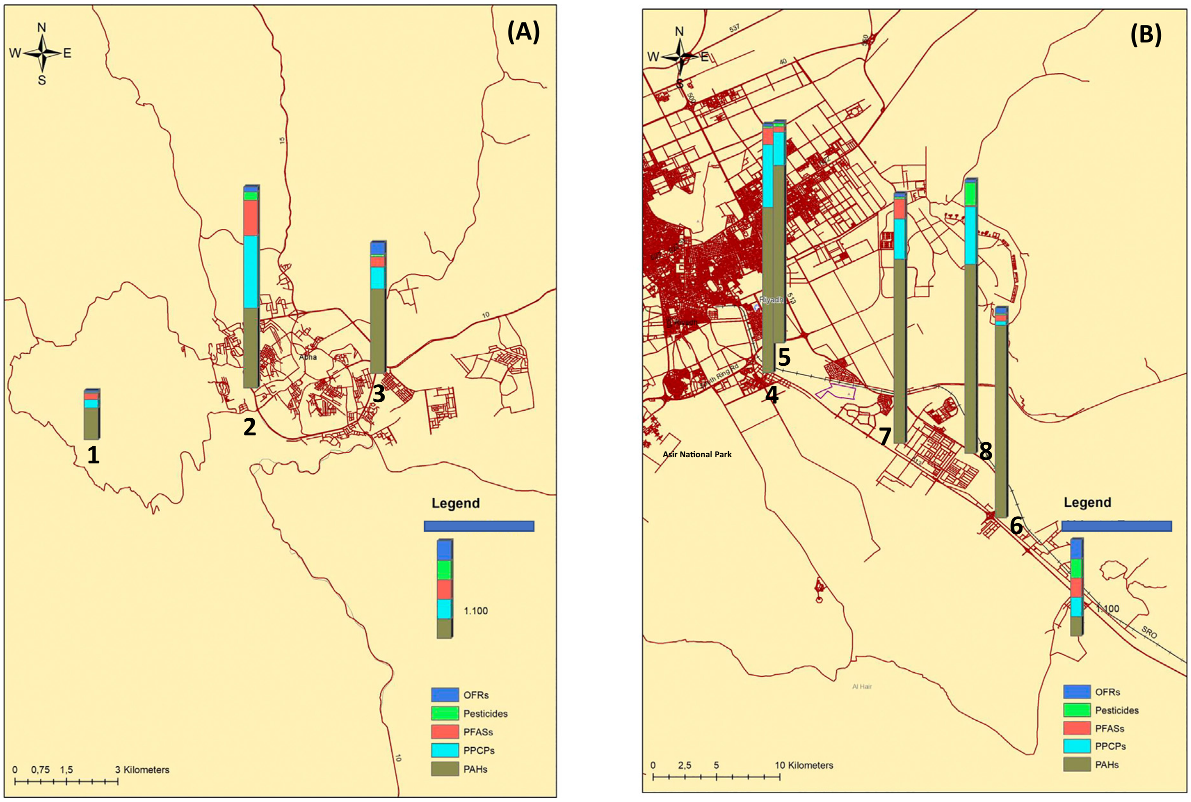

Fig. 3. Spatial distribution of the contaminants in plants at each stand of (A) Asir National Park and Abha city and (B) Riyadh.

concluded that concentrations in urban areas are higher than those in the rural areas. Comparison of levels considering only *C. erectus* found in the Riyadh sampling points showed a high number of PAHs and higher concentrations detected in stand 4 located in a cement factory whereas in the other points showed similar concentration. Contrarily, comparison of levels considering only *D. viscosa* showed more individual PAHs and at higher concentrations in the control point at the Asir Natural Park than at those points located in Abha city. However, the global results indicated a different and more rational behavior.

Fig. 3 shows the spatial distribution of the contaminants at each sampling point. It is noticeable that PAHs were found at the highest levels among the contaminants, with Riyadh exhibiting the greatest concentrations of these substances among the areas that were studied. The statistically analysis also pointed out that the three points located closed to the Al-Kharj road had levels significantly higher of PAHs (see Fig. 2) ($p < 0.05$). As shown in Table 2, naphthalene, fluorene, phenanthrene, anthracene, fluoranthene and benzo(a)pyrene are the prominent PAHs in wild and ruderal plants. These compounds are characterized to be among those with higher water solubility and vapor pressure. These properties are related to the PAHs molecular weight, which in turn is contingent upon the number of rings each molecule encompasses. Specifically, molecules containing between 2 and 3 rings are considered to have low molecular weight, those containing 4 rings are categorized as medium molecular weight, and those with 5 or more rings are regarded as having high molecular weight. This agrees with several studies establishing that PAHs with log $K_{OW} < 5$ were more easily accumulated in plants (Jia et al., 2021; Zhang et al., 2022). PAH-profiles of the plant samples showed a higher contribution of those of three (average 46 %) and four (ca. 40 %) rings followed by those with two rings (ca. 12.5 %) and finally the less abundant are those with five ring or more (3 %) (see Fig. 4). To establish the origin of PAHs, various ratios have been proposed, including the low molecular weight (LMW)/high molecular weight (HMW) ratio, Flu/(Flu + Pyr) ratio, Ant/(Ant + Phe) ratio, and IcdP/(IcdP + BghiP) ratio. According to these reports, a ratio of LMW/MHW <1 denotes a pyrogenic source whereas a ratio >1 a petrogenic one (Zhang et al., 2022). A Fla/(Fla + Pyr) ratio between 0.4 and 0.5 indicates fossil fuel combustion, < 0.4 petrogens and >0.5 grass, wood or coal combustion (De La Torre-Roche et al., 2009). An Ant/(Ant + Phe) ratio value <0.1 indicates a petrogenic source, and one >0.1 indicates a pyrogenic one (Pies et al., 2008). Finally, an IcdP/(IcdP + BghiP) ratio between 0.2 and 0.5 suggested fossil fuel combustion, < 0.2 a petrogenic source and >0.5 grass, wood or coal combustion (Yunker et al., 2002). The ration IcdP/(IcdP + BghiP) was not calculated since IcdP was not detected in the samples. The mean values of LMW/HMW, Ant/(Ant + Phe), and Flu/(Flu + Pyr) were 1.3, 0.2, and 0.6, respectively, in plants from Riyadh; 1.3, 0.3, and 0.6 in plants from Abha; and 1.6, 0.35 and 0.6 in plants from Asir National Park. These results suggest that PAHs in plants may have originated from petroleum combustion and from grass, wood, or coal combustion. Nevertheless, it is important to be cautious when interpreting these ratios because the results can be influenced not only by the PAHs present on the plant and the adjacent soil but also by the plant's ability to absorb these substances.

Wastewater, compost, and manure application release PPCPs into the soil systems (Bueno et al., 2022; Picó et al., 2019). Plants can take up these compounds (Bigott et al., 2022; Manasfi et al., 2021). In this study, PPCPs was found to be the second-highest group of contaminants in wild and ruderal plants. The ΣPPCPs concentration in the wild plants ranged from <LODs to 5496.0 ng g^{-1} d.w. with an average of 63 ng g^{-1} d.w. (see Table 2 and Fig. 3) The highest values are due to salicylic acid that reached averages >2000 ng g^{-1} d.w. This can be a natural product from plants produced specifically due to hydric stress. Then, salicylic acid could not be the best PPCP to be included in environmental analysis. In addition to this, 23 PPCPs were detected in Riyadh, 17 in Abha and 7 in the stand at the Asir National Park. Abha had the highest average concentration of ΣPPCPs (excluding salicylic acid) (25 ng g^{-1} d.w.), followed by Riyadh (18 ng g^{-1} d.w.), while Asir National Park had the lowest concentration (4 ng g^{-1} d.w.). However, these differences between ΣPPCPs concentration were not statistically significant. The differences in ΣPPCPs concentrations between stands in each city (5 for Riyadh and 2 for Abha) were not statistically significant. However, comparison of *C. erectus* in Riyadh stands shows

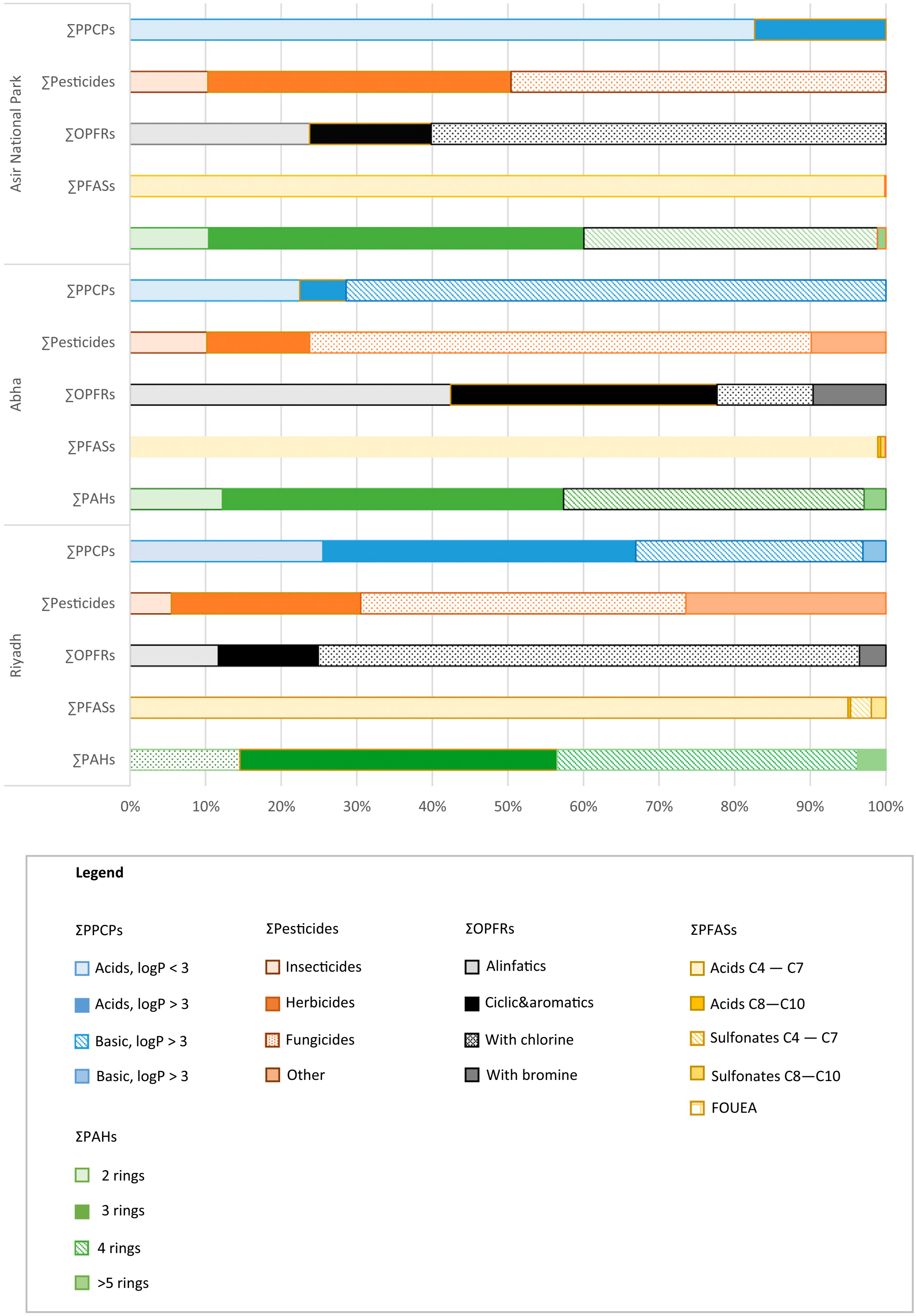

Fig. 4. Congener composition of PAH, OPFRs, Pesticides and PPCPs found in plats taken in Riyadh South, Abha and Asir National Park (Wadi Bani Marzem).

the presence in stand 4 of high concentrations of diclofenac and ibuprofen not present in the other stands. This could be related to the irrigation with treated wastewater.

The environmental behavior of PPCPs depends mostly on their electrostatic interactions (marked by the acid or basic character of the compounds) and their hydrophobicity (compounds with log P > 3 are considered hydrophobic) (Bigott et al., 2022; Manasfi et al., 2021). PPCPs were separated into acidic and basic/neutral compounds in Fig. 4 based on their charge at ambient pH. Within each group, those with log P values above or below 3 were further distinguished. In Riyadh, acidic PPCPs with log P < 3 make up 26 % of the detected compounds, while acidic PPCPs with log P > 3 account for 41 %. Basic/neutral PPCPs with log P < 3 represent 30 % of the total, while basic/neutral PPCPs with log P > 3 make up only 4 %. In Abha, only acidic PPCPs with log P < 3 (22 %) and basic/neutral PPCPs with log P < 3 (71 %) were detected. Acidic PPCPs with log P > 3 represented only 6 % of the detected compounds in Abha. In contrast, the stand located in the National Park showed a higher proportion of acidic PPCPs with log P < 3, at 82 %, and a lower proportion of acidic PPCPs with log P > 3, at 18 %. Our previous study (Picó et al., 2018a) already suggested that some ionic compounds theoretically not absorbable by roots, are somehow susceptible to be taken up by the plant and translocated to different parts of the plant, as is the case of ibuprofen. This was further confirmed under uncontrolled real environmental conditions since the presence of 7 pharmaceutical products at concentrations ranging from 25 to 96 ng g^{-1} in the soil samples, and from 35 to125 ng g^{-1} in the plants was established (Picó et al., 2019). Our results also agree in part with those reported by Bueno et al. (2022) who observed a negative correlation between the distribution of PPCPs within the plants and the log P and reported high occurrence of diclofenac and caffeine. Most of the detected PPCPs in this study have also log P < 3.

According to Xu et al. (2022), PFASs in soils were taken up by the plant root and could translocate and accumulate throughout the plant through various processes influenced by factors such as soil property and PFAS species and plants. The ΣPFASs concentrations in the plants ranged from <LOD to 3227.6 ng g^{-1} d.w. with an average of 171 ng g^{-1} d.w. (Table 2). Average ΣPFASs concentrations at each sampling stand were higher in Abha (ΣPFASs = 286.5 ng g^{-1} d.w.) than in Riyadh (ΣPFASs = 155.7 ng g^{-1} d.w.) and in this city higher than in the stand located in the National Park (ΣPFASs = 70.0 ng g^{-1} d.w.) (Figs. 2 and 3). These differences were not statistically significant ($p > 0.05$). Most prominent PFASs were the short-chain perfluorocarboxylic acids (from C4 to C7) that as can be observed in Fig. 4 appeared as the unequivocal major compounds in plants. These agree with the study of Xu et al. (2022) who reviewed the recent literature about the uptake, bioaccumulation and translocation of PFASs in plants and concluded that following uptake by root, only short-chain PFASs (C ≤ 7 for PFCA) were distributed across plant tissues.

The Σpesticides concentration in the wild plants range from <LODs to 1059 ng g^{-1} d.w. with an average of 63 ng g^{-1} d.w. Pesticides impact wild plants usually by spray drift that is carried by the wind to nearby areas. Also, in the case of gardens and public green areas, these plants can be directly treated. Other source could be wastewater since the treatments does not eliminate pesticides. No great differences are observed between the concentrations in Riyadh South and Abha, but differences are observed between these cities and the point located in the Asir National Park. The most relevant pesticides were DMPF and DMA (degradation products of amitraz), alachlor, tolclofos methyl, buprofezin and atrazine deisopropyl. Few studies covered the semi-natural habitats of wild plants were focused on those that mark the edges of agricultural fields and in neonicotinoids insecticides (Botías et al., 2016; García-Valcárcel et al., 2022). Both studies reported levels of neonicotinoid in non-target wild vegetation (range: ≤ 0.02–106 ng g^{-1} or to < method quantification limit (MQL) to 52.9 ng g^{-1} for thiametoxan and from < MQL to 98.6 ng g^{-1} for imidacloprid, depending on the study) that can be a hazard for beneficial insects. However, in this study, neonicotinoids (imidacloprid and clothianidin) represent a minor percentage of the compounds found (5 % in Riyadh and 10 % in Abha and Asir National Park) and are at low concentrations (max. 1.9 ng g^{-1} d.w. for imidacloprid and 5.6 ng g^{-1} d.w. for clothianidin) (see Fig. 4). Herbicides and fungicides are the most common pesticides (Fig. 4) but there is no information to compare their effects. This pointed out the need of additional studies to enlarge the information about the non-target pesticides.

The widespread presence of OPFRs in different matrices across the globe is attributed to transport mechanisms such as emission of OPFR-containing materials, wastewater discharge, long-range atmospheric transport and aerial deposition. In this study, tris(2-chloroisopropyl) phosphate (TClPP), tributyl phosphate (TnBP) and cresyl-diphenyl phosphate (CDP) were the most relevant OPFRs with concentrations ranging from <LOD to 520 ng g^{-1} d.w. The ΣOPFRs concentrations in wild plants ranged from <LOD to 653 ng g^{-1} d.w. with an average of 46.5 ng g^{-1} d.w. (Table 2). Average ΣOPFRs concentrations were higher in Abha (ΣOPFRs = 90.2 ng g^{-1} d.w.) than in Riyadh (ΣOPFRs = 32.0 ng g^{-1} d.w.) and in this city higher than in the stand located in the National Park (ΣPFASs = 17.0 ng g^{-1} d.w.) (Figs. 2 and 3). The differences in ΣOPFRs between Riyadh, Abha and the stand at the National Park were not statistically significant. Fig. 4 classifies the OPFRs according to their lateral chain alkyl, aryl, chlorinated and brominated. Chlorinated OPFRs are the dominant in Riyadh and Asir National Park. However, Abha showed a different pattern dominated by a higher percentage of alkyl and aryl OPFRs. The high presence of TClPP in Riyadh and Asir National Park indicate that these compounds come from a diffuse source (Wei et al., 2015) difficult to explain. There is almost no information on the behavior of these compounds in plants. Then, this study sheds light on the presence and levels of these OPFRs in wild and ruderal plants that can help interpret results from other parts of the world.

The effect of rapid industrialization, population growth and heavy traffic on human health is a major public concern. In this study, concentrations of a wide range of pollutants are found in wild and ruderal plants, even those growing in natural areas far from human impact. The concentrations found independently of species-specific characteristics and their correlation with the characteristics of the sampling site indicate that plants can be an exceptional indicator of the presence of such compounds in the environment, and their monitoring can become an early warning system for exposure control.

4. Conclusions

The results of this study demonstrate that plants can be a good bioindicator of the contamination of the surrounding abiotic compartments. The five types of contaminants studied were detected in the samples indicating that plants are capable of absorbing pollutants by aerial deposition, from water used for irrigation and soil where they grow. The concentrations of PAHs in wild and ruderal plants in the urban areas with an important traffic and industrial impact are generally higher than those with less traffic and industry and than in the natural areas with less anthropogenic impact.

For the other pollutants, the results do not show such a significant difference between larger and smaller cities, between traffic intensity or between the levels of industrialization, but there is a significant difference between the levels found in Riyadh and Abha and those found at the point located in the Asir National Park.

An important finding of the study is wide and ruderal plants could be good indicators of the environmental pollution despite differences in environmental conditions. The local wild and ruderal plants can be an excellent complementary indicator to measure the pollution in Saudi Arabia and other regions. The results suggest that determination of contaminants in wild plants can become a one-health approach to establish the impact of environmental pollution and exposure levels on plants, animals and ultimately humans.

CRediT authorship contribution statement

Yolanda Picó: Methodology, Conceptualization, Resources, Investigation, Writing – original draft, Visualization. **Julian Campo:** Conceptualization, Methodology, Validation, Formal analysis, Writing – review & editing. **Ahmed H. Alfarhan:** Writing – review & editing, Supervision, Project administration. **Mohamed A. El-Sheikh:** Methodology, Validation, Resources, Writing – review & editing. **Damià Barceló:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

Data availability

Data will be made available on request.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.164166.

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