Efficiency and mechanism of formaldehyde removal from air by two wild plants; *Plantago asiatica* L. and *Taraxacum mongolicum* Hand.-Mazz.

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Abstract

Indoor potted plants played an important role in the removal of air-borne VOCs. According to the difference between plant fresh extracts and boiled extracts on breakdown ability to the added formaldehyde, a simple quantitative evaluation method was used to identify the mechanisms of formaldehyde removal from the air by wild *Taraxacum mongolicum* Hand.-Mazz. and *Plantago asiatica* L.. After shoots exposure to formaldehyde (1.28 mg/m³ in the air) for 24 h, the formaldehyde removal rates of *P. asiatica* and *T. mongolicum* were 73.18 and 121.20 mg/h/kg FW (fresh weight), respectively. Formaldehyde can be transported from the air to the rhizosphere solution by plants, and the maximum rates of transmission by *T. mongolicum* and *P. asiatica* were 23.73 and 83.08 mg/h/kg FW, respectively. Although plant metabolism was responsible for formaldehyde loss in the air-plant-solution system, and the metabolic activity depended on the enzymatic and redox reactions in the plants, *P. asiatica* and *T. mongolicum* are still good candidate species for developing phyto-microbial technologies. The redox reaction was the main mechanism used by *P. asiatica* shoots to dissipate formaldehyde, while the enzymatic reaction was the main mechanism used by *T. mongolicum*. The higher oxidative potential and lower defensive enzyme activity in *P. asiatica* shoots led to its higher formaldehyde removal rate compared to *T. mongolicum*. Meanwhile, the stronger redox reaction ability in the *T. mongolicum* roots was partly responsible for its lower formaldehyde transmission rate. The results show two plants have strong tolerance to formaldehyde in the air and good formaldehyde removal ability.

Keywords Formaldehyde · Plant · Removal efficiency · Degradation mechanism · Enzyme · Phytoremediation

Introduction

Air pollution is an important threat to the health of urban residents [1, 2], as many illnesses are caused by air pollution, such as asthma [3, 4], lung cancer [5, 6] and cardiovascular disease [7]. It was reported that air pollution caused approximately 7 million premature deaths in 2012, of which 4.3 million were attributable to indoor air pollution [8]. Based on the consideration of the application prospects of individuals and limited space, most research on air purification technologies has focused on indoor air pollution control [9–11]. Outdoor air

management technology has mostly been addressed from the standpoint of governmental functions and environmental management, with a focus on emission control [12]. Meanwhile, pollutants in air that occur by accident or through discharges are mostly addressed using passive methods of waiting for natural purification or dilution; as a result, air pollution has plagued urban residents for a long time.

Photochemical oxidation is one of the most important mechanisms in the natural purification of organic pollutants in air, followed by phytoremediation [6, 10, 13, 14]. Plants can take up organic and inorganic pollutants in the air, break them down through assimilation or dissimilation through in vivo metabolism [15], accumulate them in tissues [16], or transport them to the rhizosphere via root release processes, and then the contaminants are eliminated through microbial degradation [17, 18]. Compared with photochemical oxidation, phytoremediation has the disadvantage of a low purification rate, and the purification efficiency is significantly affected by the plant species and environmental conditions [19, 20].

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Meanwhile, phytoremediation has the advantages of requiring no energy consumption, causing no secondary pollution, and easy application in large or small areas and at different light intensities [21, 22]. Phytoremediation is more suitable for the development of cheaper pollution control technologies for outdoor air than phytochemical oxidation because it is more easily controlled by humans.

Formaldehyde is the simplest aldehyde form and is widely found in the air due to emissions from factories and automobiles, the volatilization of building materials, and so on [23, 24]. The level of formaldehyde in polluted indoor air often exceeds the WHO limits by a dozen-fold (0.1 mg/m^3) [25]. Although the indoor air formaldehyde level is often higher than that of outdoor air, the formaldehyde in outdoor air is still an important source of indoor formaldehyde pollution because of indoor and outdoor ventilation. Before a pollution source can be completely eliminated, formaldehyde pollution in indoor and outdoor air should be simultaneously addressed to minimize the health risks for urban residents.

Formaldehyde in air can be removed by potted plants [26]. Formaldehyde dehydrogenase in plant tissue and microorganisms in the rhizosphere play important roles in the removal mechanisms [26, 27]. Thus, the ability of enzymes in plants to purify formaldehyde is one of the essential parameters for the assessment of the efficiency of plants in removing formaldehyde from air. Considering the limited area of contact between soils and closely planted plants, the rate of formaldehyde transmission from the air to the rhizosphere by plants should be considered as an important parameter for assessing the efficiency of phytoremediation [28]. Reactive oxygen species (ROS) in plants may play an important role in the purification of formaldehyde by plants [29]. Meanwhile, formaldehyde affects the activities of three defensive enzymes, i.e., peroxidase (POD), catalase (CAT), and superoxide dismutase (SOD), of which POD and CAT are the most sensitive to formaldehyde stress [30]. More evidence is needed to confirm the mechanism of formaldehyde removal. The variability in natural conditions makes wild plants more adaptable to environmental stress than those growing in anthropogenic environments, which may be beneficial for confirming the existence of redox mechanisms. Thus, wild *P. asiatica* and *T. mongolicum* were selected as the studied plant species in this paper. Both species naturally occur in the field and have medicinal value because of the existence of some active ingredients [31, 32].

The main aims of this study were to (i) quantitatively measure the rate of formaldehyde removal from the air by two plants, (ii) test the existence of a redox reaction mechanism in the breakdown of formaldehyde in plant tissues, and (iii) evaluate the potential use of the two plants for removing formaldehyde from the air based on their formaldehyde transmission rate, in vivo purification ability and antioxidant enzyme activity as parameters.

Materials and methods

Chemicals

All chemicals were analytical grade, including sodium dihydrogen phosphate ($\text{NaH}_2\text{PO}_4 \cdot 2\text{H}_2\text{O}$, 156 g/mol, purity >99.0%), disodium hydrogen phosphate ($\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$, 358 g/mol, purity >99.0%), ammonium acetate ($\text{CH}_3\text{COONH}_4$, 77 g/mol, purity >99.0%), hydrogen peroxide (H_2O_2 , 34 g/mol, 30%), guaiacol ($\text{C}_7\text{H}_8\text{O}_2$, 124 g/mol, purity >99.0%), and formaldehyde (HCHO , 30 g/mol, 37–40%). Formaldehyde solution was calibrated using the standard of the China Environmental Protection (HJ601–2011).

Preparation of plant seedlings

A number of wild plant seedlings growing in the same uncontaminated field were carefully separated from the soil, followed by thorough washing with deionized water. Then, uniform seedlings were individually selected and transplanted into 4-L containers filled with 1/5-strength Hoagland nutrient solution. The plants were grown hydroponically for 2 weeks before experimentation. The nutrient solution was continuously aerated and refreshed every two days.

Treatments with formaldehyde

There were three control treatments in the experiment, which are listed in Table 1. Based on the leaf area and biomass, uniform wild plant seedlings were selected and transplanted to glass bottles containing 20 mL of a modified Hoagland nutrient solution [28] added just before the experiment, with one *P. asiatica* (series-1) and *T. mongolicum* (series-2) plant per bottle (Table 1); the bottles were wrapped with dark paper, and the open area between the cap and plant shoots was sealed with a sponge wrapped in aluminium paper. The nutrient solution was aerated for 0.5 h before use in the experiment. Three glass bottles containing a plant and one glass bottle without plants (control A) (Table 1) were carefully placed into a transparent glass container ($50 \times 30 \times 35 \text{ cm}$) filled with 1000 mL of a solution containing varying formaldehyde contents as described in reference [28]. The device, which is schematically depicted, consisted of a static chamber and a sampling system (Fig. 1).

The container (pot) was sealed and placed into a plant growth room (with a 14-h light period) with temperatures of 25°C during the day and 20°C at night and 50–70% relative humidity. Two controls without formaldehyde were established at the same time. One control without plants in the glass container (control B,) was used to test the loss of formaldehyde in the container caused by other processes (such as gas leakage, photochemical reactions) and to study the relationship between the formaldehyde level in the air and the

Table 1 Treatments in the experiment

Series		1	2	3	4	5
Parameters	plants	<i>P. asiatica</i>	<i>T. mongolicum</i>	–	–	<i>P. asiatica</i> / <i>T. mongolicum</i>
	HCHO in containers	+	+	+	+	–
Sampling	Solution in containers	+	+	+	+	–
	Solution in bottles	+	+	+	–	–
	Plant tissues	+	+	–	–	<i>P. asiatica</i> / <i>T. mongolicum</i>
Brief description		HCHO removal in air-plant-water system		Control A, for series-1 or series-2	Control B, for series-1 and series-2	Control C, for series-1 or series-2
Objectives		(i),(ii),(iii)				

+/, with or without, –, no bottles in containers

content in the water under the experimental conditions. Another control (control C) consisted of glass bottles containing plant seedlings that were immersed in a vessel filled with 1000 mL of water without formaldehyde.

In treatment-1 (T1), the initial formaldehyde level in the air of the containers with *P. asiatica* was set at $0.65 \pm 0.04 \text{ mg/m}^3$, which was measured using a formaldehyde detector and verified based on the formaldehyde concentration in solution. In treatment-2 (T2), the initial formaldehyde level in the air of the containers with *P. asiatica* and *T. mongolicum* was set at $1.28 \pm 0.05 \text{ mg/m}^3$. In treatment-3 (T3), the initial formaldehyde level in the air of the containers with *P. asiatica* and *T. mongolicum* was set at $1.91 \pm 0.15 \text{ mg/m}^3$, and in treatment-4 (T4), the initial formaldehyde level in the air of the containers with *T. mongolicum* was set at $2.54 \pm 0.34 \text{ mg/m}^3$. At selected times (4, 9, and 24 h), a portion of the plant leaves in the three replicate bottles in treatment-1, treatment-2 and treatment-3 was removed for the analysis of contaminant concentrations and leaf oxidative capabilities. At the same time, two replicate samples of the solutions in the containers and bottles with or without plants were collected to determine

the contaminant concentrations. After sampling, equal volumes of the nutrient solution were immediately added to the bottles. After the foliar uptake of formaldehyde for 24 h, the bottles with the studied plants were taken out of the containers, the outer walls of the bottles were rinsed and blotted dry with a tissue paper, and the final volume of the rhizosphere solutions and fresh weights of the plants were measured. The formaldehyde solution was calibrated against the standard of the China Environmental Protection (HJ601–2011) before use in the experiment.

Analysis of formaldehyde concentrations

The analysis of formaldehyde was conducted as previously reported [28]. The leaf samples were weighed after being blotted dry with a tissue paper, homogenized using a mortar and pestle, and extracted using 2 mL of a cool phosphate buffer (phosphate-buffered saline, pH = 7.8). The extracted solution was transferred into a 10-mL centrifuge tube. The leaf samples were then extracted twice with 6 mL of fresh cool phosphate buffer. All extracted solutions were combined and centrifuged

Fig. 1 The experimental apparatus consisting of a static chamber and a sampling system

for 10 min. After centrifugation, 1.0 mL of the supernatant was transferred to a 10-mL glass colorimetric tube, and 0.5 mL of acetylacetone solution (prepared with ammonium acetate and glacial acetic acid) was added and diluted (and mixed) with distilled water to 5 mL. The mixtures in the colorimetric tubes were placed into a thermostated water bath fixed at 60 ± 2 °C for 15 min, subsequently cooled with cold water, and then filtered. The liquid phase was collected, and the concentration of formaldehyde was measured using spectrophotometry (UV-751, China).

The formaldehyde concentrations in the samples collected from the rhizosphere solution in control A and the absorbance of the solution in the containers were directly analysed using acetylacetone spectrophotometry with an ultraviolet-visible spectrophotometer at 414 nm. The detection limit of this method is 0.05 mg/L. The formaldehyde levels in the air were measured using an automatic formaldehyde detector (precision, $\pm 5\%$, detection limit, 0.01 mg/m³) (BGFM-05, China) and verified using the Antoine equation [33] based on the results of the formaldehyde concentrations in the solutions.

Abilities of the leaf extracts or rhizosphere solution to degrade formaldehyde

The leaf samples from the treatments and control C were extracted using 2 mL of phosphate buffer (pH = 7.8) four times, and the extracts were combined and centrifuged. A series of 0.5-mL clear liquids were separated from the extracts, and various volumes of 250 mg/L formaldehyde solution (50 μ L to 300 μ L) were added. Changes in the formaldehyde mass in the mixtures were quickly detected using acetylacetone spectrophotometry. In the control experiments, the same volumes of 250 mg/L formaldehyde solution were added to distilled water and analysed. The differences in the formaldehyde levels of the studied and control samples were computed to assess the capability of the leaf extracts to degrade the added formaldehyde. Boiled leaf extracts were prepared by placing glass bottles containing 10 mL of fresh leaf extracts into boiling water for ten minutes and then quickly cooling them. The capability of the boiled leaf extracts to dissipate the added formaldehyde was measured using the same method.

Accuracy and precision of the tests

In order to test the accuracy and precision of the method, the shoot and root samples collected from Control C were dried by a drier (XGQ-2000, China) at 80 °C, and then extracted by PBS buffer. The recoveries of the formaldehyde in the spiked shoot and root samples extracts with the method were $81.24\% \pm 0.43\%$ ($n = 12$) and $95.77\% \pm 0.78\%$ ($n = 12$), respectively.

Analysis of POD and catalase CAT activity

Samples (0.5 g fresh shoot and root) were separately collected and kept at -20 °C. The samples were ground in liquid nitrogen and extracted with 50 mM potassium phosphate buffer (pH = 7.8) containing 0.1 mM EDTA, 1% (w/v) polyvinyl pyrrolidone (PVP), 0.1 mM phenylmethanesulfonyl fluoride solution (PMSF) and 0.2% (v/v) Triton X-100 for the measurement of POD and CAT. The extracts were centrifuged at 4000 rpm and $0-4$ °C for 15 min. The supernatants were used for enzyme activity assays and gel electrophoresis. All procedures were performed at $0-4$ °C.

The CAT activity was detected at 240 nm in 2 mL of 50 mM potassium phosphate buffer (pH = 7.8) with 3 mM H₂O₂ and an enzyme extract containing 100 μ g proteins. The enzyme activity was calculated from the initial rate of the enzyme using the extinction coefficient of H₂O₂ of $40 \text{ mM}^{-1} \text{ cm}^{-1}$ at 240 nm [34].

The POD activity was assayed in 2 mL of 100 mM potassium phosphate buffer (pH = 6.5) containing 40 mM guaiacol, 10 mM H₂O₂ and an enzyme extract containing 100 μ g proteins at 25 °C using a spectrophotometer at 470 nm. The activity was based on the rate of tetra guaiacol production using an extinction coefficient of $25.5 \text{ mM}^{-1} \text{ cm}^{-1}$. The enzyme activity was recorded as the optical density (OD). All determinations were based on triple measurements of each sample.

Data analysis

Some experimental data, including the formaldehyde removal rates, formaldehyde transmission rates, breakdown coefficients, contributions of enzymatic reactions or oxidation reactions and activity of defensive enzymes (POD and CAT) were subjected to regression and one-way analysis of variance (SPSS, version 13.0).

Results and discussion

Formaldehyde removal rates

In control B, there was an approximate 4.31–7.22% decrease in the formaldehyde concentration in the container solution after 24 h, which was far lower than the decrease found in association with *P. asiatica* and *T. mongolicum*. Consistent with the Antoine equation, a linear relation existed between the formaldehyde levels in the air and the solution inside a container, namely, $C_{air} = 0.0211C_{solution} + 0.0137$, with $r = 0.9991$. Meanwhile, the formaldehyde concentration in control A was below the detection limit, indicating that most of the formaldehyde losses in the containers in the treatments were mainly caused by plants.

Fig. 2 Time-dependent curves of the formaldehyde levels in the air. Error bars represent standard deviations ($n = 3$)

The results show that both *P. asiatica* and *T. mongolicum* could efficiently remove formaldehyde from the air. The formaldehyde levels in the air in the treatments decreased with the shoot exposure time increasing (Fig. 2). Based on the plant biomass, the formaldehyde removal rates of *P. asiatica* reached 16.60, 73.18 and 50.62 mg/h/kg FW in 24 h when the initial formaldehyde concentration in the air was 0.65, 1.28 and 1.91 mg/m³, respectively, while the removal rates of *T. mongolicum* were 121.20, 38.39 and 238.81 mg/h/kg FW, corresponding to the initial formaldehyde concentrations of 1.28, 1.91 and 2.54 mg/m³ (Table 2). The removal rates were significantly influenced by the formaldehyde level in the air, which is consistent with the results of a previous report [35]. Due to the physiological differences among plants, their tolerances to formaldehyde also differ.

The experimental conditions are often limited (e.g. formaldehyde levels in the air or exposure time).

Formaldehyde transmission rates

The results show that formaldehyde can be transported from the air to the rhizosphere solution. The transmission rate of *P. asiatica* increased with an increase in the initial formaldehyde level in the air within 4 h of exposure time but sharply decreased with an increase in exposure time (Table 3). Under the same conditions, the transport ability of *T. mongolicum* was far greater than that of *P. asiatica*. When the initial formaldehyde level in the air was set at 1.28 and 1.91 mg/m³, the transmission rates of *T. mongolicum* were 2.50–4.17-fold those of *P. asiatica* within 24 h. Thus, these two plants could be used to develop phyto-microbial technologies for purifying air contaminated with formaldehyde, especially *T. mongolicum*. The formaldehyde removal rate was one thousand-fold the transmission rate, and accumulation in the plant tissue can be ignored; therefore, formaldehyde breakdown in plant tissues should be the main mechanism of formaldehyde removal by plants.

Capability of plant extracts to dissipate added formaldehyde

The ability of plant tissues to break down formaldehyde was investigated by measuring the capability of the plant extracts to dissipate the added formaldehyde. Meanwhile, boiled shoot and root extracts were prepared from control C to test whether a redox reaction existed and to distinguish the contributions of

Table 2 Rates of formaldehyde removal from the air by plant seedlings

Plants	Initial HCHO conc. in the air (mg/m ³)	Removal rate (mg/h/kg FW)						
		0~4 h	5~9 h	10~24 h	ANOVA	<i>r</i>	Correlation	0~24 h
<i>P. asiatica</i>	0.65	15.70±0.76	7.10±1.22	20.01±1.42	**	0.62	NC	16.60±1.27
	1.28	82.85±2.20	34.32±4.23	83.55±4.40	***	0.34	NC	73.18±4.00
	1.91	79.91±13.50	63.77±5.70	38.42±4.89	*	0.10	Negative	50.62±6.49
	ANOVA	**	***	***				***
	<i>r</i>	0.85	1.00	0.28				0.361
	Correlation	NC	Negative	NC				NC
<i>T. mongolicum</i>	1.28	79.40±2.70	128.59±5.73	129.88±6.72	**	0.67	NC	121.20±5.84
	1.91	75.39±9.76	57.58±4.40	22.13±6.18	**	0.70	NC	38.39±6.41
	2.54	138.85±16.08	116.79±7.43	306.14±4.77	***	0.96	Positive	238.81±7.21
	ANOVA	*	***	***				***
	<i>r</i>	0.999	0.90	0.90				0.584
	Correlation	Positive	Negative	Positive				NC

Data are listed as the average ± standard error for three replicates

NS, not significant; *r*, correlation coefficient; NC, No correlation

***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$

Table 3 Rates of formaldehyde transport from the air to the rhizosphere solution by plant seedlings

Plants	Initial HCHO conc. in the air (mg/m ³)	Transport rate (mg/h/kg FW)						
		0–4 h	5–9 h	10–24 h	ANOVA	<i>r</i>	Correlation	0–24 h
<i>P. asiatica</i>	0.65	32.02 ± 5.08	13.71 ± 0.81	0	**	0.94	Negative	8.19 ± 1.02
	1.28	63.08 ± 10.71	23.29 ± 2.82	3.78 ± 0.50	**	0.90	Negative	17.73 ± 2.69
	1.91	130.16 ± 14.84	9.75 ± 1.15	0	***	0.74	NC	23.72 ± 2.71
	ANOVA	**	***	***				***
	<i>r</i>	0.98	0.28	0				0.10
	Correlation	Positive	NC	NC				Positive
<i>T. mongolicum</i>	1.28	197.90 ± 19.12	75.16 ± 7.89	37.32 ± 3.09	***	0.84	NC	71.97 ± 6.76
	1.91	217.96 ± 2.94	99.33 ± 3.11	41.70 ± 3.25	***	0.90	Negative	83.08 ± 3.17
	2.54	130.60 ± 8.03	48.93 ± 5.75	26.19 ± 0.85	***	0.828	NC	48.33 ± 3.07
	ANOVA	***	***	**				***
	<i>r</i>	0.74	0.520	0.70				0.67
	Correlation	NC	NC	NC				NC

Data are listed as the average ± standard error for three replicates

NS, not significant; *r*, correlation coefficient; NC, No correlation

***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$

different mechanisms to formaldehyde removal. The results show that the formaldehyde in the boiled plant extracts could be quickly dissipated (Fig. 3), indicating the existence of other reactions in addition to enzymatic reactions. When the initial formaldehyde concentration in the extracts ranged from 18 to 125 mg/L, the amount broken down was positively related to the formaldehyde concentration in the extracts (Fig. 4). Considering the composition of the boiled plant extracts, a redox reaction between the reducing formaldehyde and the oxidizing ROS in plant tissues was likely to occur.

Maximum values for the breakdown capability of both the fresh and boiled extracts existed at certain formaldehyde levels (Fig. 3). Therefore, the contributions of the enzymatic reaction mechanism could be calculated as the difference in the ability of the fresh and boiled extracts to dissipate the added formaldehyde. The slope K was used to evaluate the ability of the plant extracts to dissipate formaldehyde, with K_T used as the parameter for the total breakdown ability of the fresh extracts and K_e and K_r used as parameters for the breakdown ability of the enzymatic reaction mechanism and redox reaction mechanism, respectively. The results show that the K_T value of the *T. mongolicum* shoot was 2.7-fold that of the *P. asiatica* shoot in control C (Table 4), while the K_T value of the *P. asiatica* root was only 45.6% of that of the *T. mongolicum* root (Table 5). These great differences were caused by the different contributions of the two breakdown mechanisms. The K_r value of the *P. asiatica* shoot extract was 6.2-fold that of the *T. mongolicum* shoot extract, while the K_e value of the *T. mongolicum* shoot extract was 1.6-fold that of the *P. asiatica* shoot extract. The redox reaction dominated the mechanism of formaldehyde breakdown in the *P. asiatica*

Fig. 3 Time-dependent curves of *T. mongolicum* (a) and *P. asiatica* (b) fresh leaf extracts or boiled leaf extracts in response to the added formaldehyde. Error bars represent standard deviations ($n = 3$)

Fig. 4 Concentration-dependent curve of the breakdown capability of leaf extracts. Error bars represent standard deviations ($n = 3$)

shoot extract, while the enzymatic reaction played a critical role in formaldehyde breakdown in the *T. mongolicum* shoot extract (Table 5). Conversely, only 2% of the breakdown ability of the *T. mongolicum* root extract was contributed by the enzymatic reaction mechanism, and 46% of that of the *P. asiatica* root extract was contributed by the enzymatic reaction. Considering the measured formaldehyde removal rate and transmission rate of the plants (Tables 2 and 3), it seems that the breakdown ability of the plant shoot was mainly responsible for the formaldehyde removal rate, while the formaldehyde transmission rate was mainly determined by the breakdown ability of the plant root. Therefore, the changes in the K_T values and the corresponding activity of the defensive enzymes POD and CAT in the plant shoots during the

formaldehyde treatments were investigated to further verify the existence and contribution of the redox reaction mechanism in formaldehyde removal by plants.

Activity of defensive enzymes

The activity of the antioxidant enzymes POD and CAT in the plant shoots collected from control C and the treatments was analysed to reflect the ROS level and oxidation potential in plant tissues [36]. The results show that the POD and CAT activities in the *T. mongolicum* shoot collected from control C were obviously stronger than those in the *P. asiatica* shoot ($p < 0.01$), corresponding to a low ROS level and oxidation potential and resulting in the low K_T value of *T. mongolicum* shoot extracts (Table 5).

When the treatments began, formaldehyde taken up by the plant shoots via gas exchange controlled by the stomata was rapidly dissipated by enzymatic and redox reactions in the plant tissues [15, 37]. With an increase in the initial formaldehyde level in the air, the foliar uptake rate of formaldehyde also rose because of an increase in the diffusion rate accompanied by elevated enzymatic reaction and redox reaction rates, resulting in a sharp increase in the rate of formaldehyde removal by the two plant species (Table 2). Meanwhile, a constant redox reaction led to a decrease in the ROS level accompanied by a decrease in the POD or CAT activity (Table 6), which was similar to results reported in the literature [31]. In addition, this constant redox reaction resulted in decreases in the breakdown ability of the plant shoot and the

Table 4 Breakdown coefficients (K_T , the ratio of the breakdown efficiency (mg/kg FW) to the initial concentration of HCHO in the extracts (mg/L) of plant shoot extracts for formaldehyde

Plants	Initial HCHO conc. in the air (mg/m ³)	K_T				ANOVA	r	Correlation
		Control	4 h	9 h	24 h			
<i>P. asiatica</i>	0.65	4.15 ± 0.15	4.24 ± 0.12	5.30 ± 0.25	3.40 ± 0.10	***	0.67	NC
	1.28	4.15 ± 0.15	3.41 ± 0.17	4.18 ± 0.19	4.25 ± 0.16	**	0.75	NC
	1.91	4.15 ± 0.15	3.67 ± 0.10	4.73 ± 0.22	4.20 ± 0.15	**	0.24	NC
	ANOVA		**	**	**			
	r		0.67	0.51	0.84			
	Correlation		NC	NC	NC			
<i>T. mongolicum</i>	1.28	11.04 ± 0.95	11.85 ± 1.18	12.22 ± 1.07	16.78 ± 1.14	11.85 ± 1.18	0.99	Positive
	1.91	11.04 ± 0.95	13.42 ± 1.03	9.86 ± 0.89	12.15 ± 0.84	13.42 ± 1.03	0.08	NC
	2.54	11.04 ± 0.95	13.26 ± 1.49	10.18 ± 0.93	11.45 ± 0.65	13.26 ± 1.49	0.34	NC
	ANOVA		NS	NS	**	NS		
	r		0.82	0.80	0.92			
	Correlation		NC	NC	Negative			

Data are listed as the average ± standard error for three replicates

NS, not significant; r , correlation coefficient; NC, No correlation

***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$

Table 5 Contributions of the enzymatic reaction (R_e , mg/kg) or oxidation reaction (R_r , mg/kg) to the formaldehyde breakdown efficiency of plant extracts (the initial concentration of formaldehyde was 13.5 mg/L). The parameters K_e and K_r represent the ratio of R_e or R_r to the initial formaldehyde concentration in the plant extracts

Plants	Parts	R_e , mg/kg FW	R_r , mg/kg FW	K_e	K_r	ANOVA (R or K)
<i>P. asiatica</i>	Shoot	21.38 ± 1.27	129.01 ± 1.65	1.70 ± 0.14	9.39 ± 0.13	***
	Root	43.77 ± 1.22	52.06 ± 0.17	3.23 ± 0.18	3.85 ± 0.18	*
	ANOVA	***	***	***	***	
<i>T. mongolicum</i>	Shoot	35.70 ± 0.50	20.50 ± 0.44	2.64 ± 0.08	1.51 ± 0.07	***
	Root	2.97 ± 0.39	205.94 ± 0.87	0.31 ± 0.02	15.21 ± 0.03	***
	ANOVA	***	***	***	***	

Data are listed as the average ± standard error for three replicates

NS, not significant; r , correlation coefficient; NC, No correlation

***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$

formaldehyde removal rate. With an increase in the formaldehyde level in the air, a constant increase in the rate of diffusion and a decrease in the rate of redox reactions eventually led to

the transient accumulation of formaldehyde in the plant shoot, resulting in an increase in downward transport from the shoot to root and a higher transmission rate (Table 3). At this time,

Table 6 Activity of POD and CAT in plant shoot extracts

Plants		Initial HCHO conc. in the air (mg/m ³)	Activity of defensive enzymes (U/g FW/min)					r	Correlation
			0 h	4 h	9 h	24 h	ANOVA		
<i>T. mongolicum</i>	POD	1.28	1305 ± 99	1150 ± 47	593 ± 30	1312 ± 130	***	0.52	NC
		1.91	1305 ± 99	962 ± 144	826 ± 55	738 ± 77	NS	0.90	Negative
		2.54	1305 ± 99	1311 ± 96	807 ± 39	1136 ± 124	**	0.016	NC
		ANOVA		NS	**	NS			
		r		0.46	0.83	0.30			
		Correlation		NC	NC	NC			
	CAT	1.28	3549 ± 147	4928 ± 155	3941 ± 207	3499 ± 657	NS	0.85	NC
		1.91	3549 ± 147	3553 ± 121	2996 ± 283	3004 ± 151	NS	0.65	NC
		2.54	3549 ± 147	4470 ± 158	2496 ± 165	5125 ± 198	***	0.54	NC
		ANOVA		***	**	**			
		r		0.33	0.99	0.73			
		Correlation		NC	Negative	NC			
<i>P. asiatica</i>	POD	0.65	569 ± 111	510 ± 55	639 ± 141	206 ± 26	*	0.90	Negative
		1.28	569 ± 111	847 ± 47	1010 ± 24	938 ± 41	NS	0.25	NC
		1.91	569 ± 111	916 ± 93	746 ± 69	967 ± 201	NS	0.53	NC
		ANOVA		**	NS	***			
		r		0.93	0.28	0.90			
		Correlation		Positive	NC	Positive			
	CAT	0.65	2005 ± 249	1968 ± 141	1932 ± 215	2456 ± 192	NS	0.97	Positive
		1.28	2005 ± 249	1930 ± 151	1978 ± 189	2167 ± 169	NS	1.00	Positive
		1.91	2005 ± 249	1025 ± 75	1320 ± 195	1234 ± 162	NS	0.41	NC
		ANOVA		***	NS	***			
		r		0.90	0.83	0.96			
		Correlation		Negative	NC	Negative			

Data are listed as the average ± standard error for three replicates

NS, not significant; r , correlation coefficient; NC, No correlation

***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$

the formaldehyde removal rate was reaction limited rather than diffusion limited, which differs from previous reports [26]. At the same time, stress caused by transient formaldehyde accumulation in plants leads to higher ROS levels, resulting in an increase in the redox reaction rate and enzyme activity until the accumulated formaldehyde is rapidly dissipated. This alternating process of transient accumulation and elimination led to the irregular changes in the activity of the antioxidant enzymes (CAT and POD) and the formaldehyde removal rates with an increase in uptake time (Tables 2 and 6).

When the uptake duration increased from 9 h to 24 h at a low formaldehyde concentration (0.65 mg/m^3), the low diffusion rate led to the weak transient accumulation of formaldehyde in the plant shoot and a limited increase in the ROS level, resulting in the 1.9-fold increase in the removal rate of *P. asiatica*, while no increase was found in that of *T. mongolicum* due to their different K_e values (Tables 2 and 5). When the shoots were exposed to a high concentration of formaldehyde (2.54 mg/m^3), the high formaldehyde diffusion rate led to elevated and rapid formaldehyde accumulation in the plant tissues, resulting in a ROS burst induced by formaldehyde stress and a high formaldehyde removal rate in *T. mongolicum* over 24 h.

The dominance of the redox reaction mechanism in the ability of the *P. asiatica* shoot to break down formaldehyde resulted in there being no significant differences in the K_T values of the *P. asiatica* shoot extracts collected in T1, T2, and T3 at different uptake times (4, 9, and 24 h) since the ROS level was regulated by the antioxidant enzymes (Table 3). In contrast, the ROS burst could lead to the inhibition of physiological activities in plants, resulting in changes in the K_T values of the *T. mongolicum* shoot extracts collected in T2, T3 and T4 at different uptake times (4, 9, and 24 h) due to the dominance of the enzymatic reaction mechanism in the ability of the *T. mongolicum* shoot to break down formaldehyde.

Conclusions

P. asiatica and *T. mongolicum* can efficiently remove formaldehyde from the air, and only a small amount was transported to the rhizosphere solution. The removal rate mainly depended on the ability of the plant shoot to break down formaldehyde, which was determined by enzymatic and redox reactions. The redox reaction mechanism dominated the ability of the *P. asiatica* shoot to break down formaldehyde, while the enzymatic reaction was the dominant mechanism for the *T. mongolicum* shoot to dissipate formaldehyde. The formaldehyde removal rate was diffusion limited rather reaction limited when foliar uptake began, while it gradually changed to reaction limited with an increase in the uptake time or the formaldehyde concentration in the air because of the

limitations of the enzymatic reaction and the regulation of ROS by antioxidant enzymes (POD and CAT).

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Compliance with ethical standards

Competing interests The authors declare that they have no competing interests.

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