atmosphere

Supplementary Materials

1. Exposure experiments

We prepared environmentally relevant water-soluble brown carbon concentration (5 μgmL^{-1}) using humic acids and a mixture of amino acids (i.e., glycine, alanine, valine, leucine, isoleucine, proline, aspartate, glutamate, serine, and threonine). The humic acid solution was dissolved in 100 mL of sodium hydroxide with the use of 0.005 g of humic acid. Then, the solution was adjusted to pH 7.0 using 10 mM hydrochloric acid. The environmentally relevant water-soluble brown carbon solution was prepared with 5 μg mL^{-1} of humic acids and amino acids including glycine, alanine, valine, leucine, isoleucine, proline, aspartate, glutamate, serine, and threonine at a mass ratio of 1:1. We selected the exposure trees with similar heights (3-5 meters) and trunk diameters (30-50 centimeters), as well as similar environmental conditions (e.g., water, soil, and wind conditions). Three types of trees, including *Eucommia ulmoides*, *Koelreuteria paniculata*, and *Osmanthus fragrans* were selected in the exposure experiment. Before the exposure experiment, the leaves were washed with de-ionized water three times. Then, the concentration of water-soluble brown carbon at 5 μg mL^{-1} was sprayed on the leaves. The experiments were carried out for 10 min. Every 2 min, one leaf exposed to 5 μg mL^{-1}of water-soluble brown carbon from the exposure tree was collected for analysis.

2. Measurements of submicron water-soluble brown carbon

The standard solutions were prepared using 1-50 μg mL^{-1} of water-soluble brown carbon, which includes humic acids and a mixture of amino acids (i.e., glycine, alanine, valine, leucine, isoleucine, proline, aspartate, glutamate, serine, and threonine). Then, we recorded the u*l*traviolet-visible light absorbance of water-soluble brown carbon over a wavelength range of 400-410 nm at an interval of 0.1 nm. We determined the levels of water-soluble brown carbon in the ultraviolet-visible absorption spectra at 405 nm [1]. We plotted the ultraviolet-visible absorption spectra at 405 nm of water-soluble brown carbon against the concentration of water-soluble brown carbon from 1 to 50 μg mL^{-1}. Under optimal conditions, the coefficients of water-soluble brown carbon determination were found to be greater than 0.98 with the linear range between 1-50 μg mL^{-1}. The relative standard deviation (RSDs) measured at 5 μg mL^{-1} varied from 4.5 %, and the limit of detection was 0.5 μg mL^{-1}. We carried out the spiked experiments for the quality control of the established method. The average recoveries of water-soluble brown carbon in leaves were in the range of 85-95%, with a relative standard deviation within 5%.

3. The adsorption of water-soluble brown carbon by tree leaves

The concentration of water-soluble brown carbon versus sampling time is shown in Figure S1. Three types of leaves exhibited strong absorption ability of humic acids as well as a mixture of glycine, alanine, valine, leucine, isoleucine, proline, aspartate, glutamate, serine, and threonine at 5 μg mL^{-1}. The loss rates of water-soluble brown carbon were observed to be greater than 40% within 2 minutes for three types of leaves and exceeded 80% in 8 minutes. These results indicated that the humic acids and a mixture of amino acids could enter the leaf through the mechanical barrier of the stomata of the leaves. Several studies have documented that humic acids and a mixture of amino acids were used as fertilizer for leaves to alleviate the stress of the drought [2, 3]. On the other side, the levels of water-soluble brown carbon were on the order of magnitude in the range from 0.1-3 μg m^{-3}, which is lower than the concentration of water-soluble brown carbon in this exposure experiment [4]. The findings from this exposure experiment showed that the water-soluble brown carbon cannot be retained on the surfaces of tree leaves within 10 minutes. Our prior study also examined the interaction process between polycyclic aromatic hydrocarbons (PAHs) and tree leaves at high concentrations using whole-transcriptome analysis. From the analysis of the significant differentially expressed genes, seven main pathways, including flavone and flavonol biosynthesis, glyoxylate and dicarboxylate metabolism, RNA polymerase, ribosome biogenesis in eukaryotes, porphyrin metabolism, photosynthesis-antenna proteins, and photosynthesis were involved for absorbing high concentration of PAHs on the surface of plant leaves [5]. This study showed that tree leaves could metabolize humic-like substances.

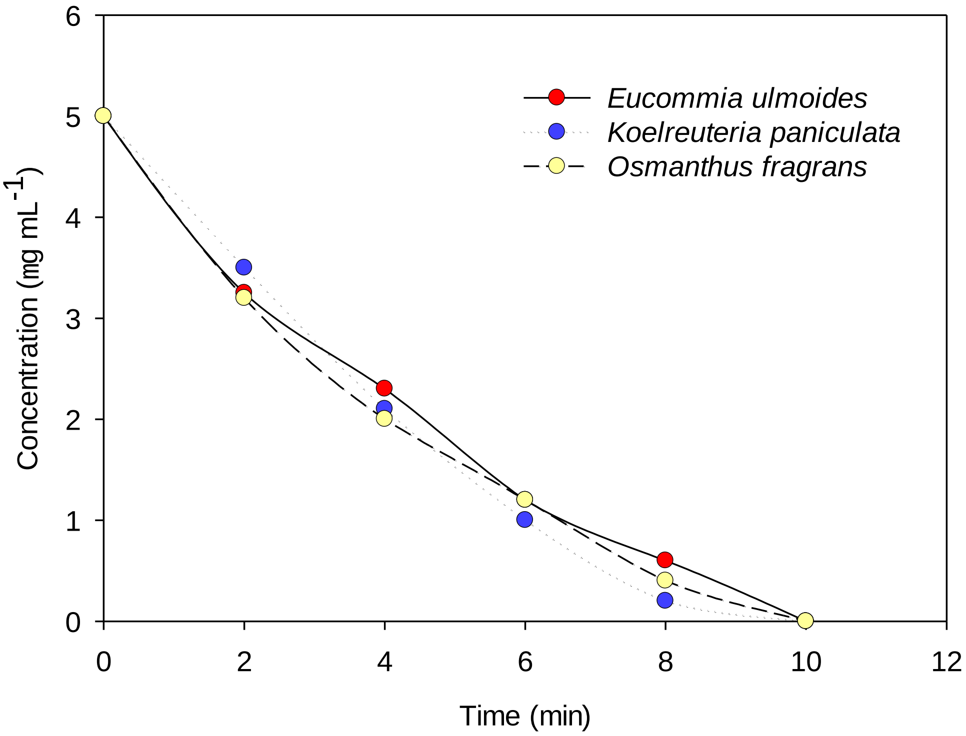

Figure S1. The concentration of humic acids and a mixture of amino acids (i.e., glycine, alanine, valine, leucine, isoleucine, proline, aspartate, glutamate, serine, and threonine) at 5 μg mL^{-1} against time in leaves of three trees.

References

1. Zhi, Y.; Li, X.; Lian, F.; Wang, C.; White, J. C.; Wang, Z.; Xing, B. Nanoscale iron trioxide catalyzes the synthesis of auxins analogs in artificial humic acids to enhance rice growth. *Sci. Total Environ.* **2022**, *848*,157536.
2. Khorasani, H.; Rajabzadeh, F.; Mozafari, H.; Pirbalouti, A. G. Water deficit stress impairment of morphophysiological and phytochemical traits of Stevia (Stevia rebaudiana Bertoni) buffered by humic acid application. *S. Afr. J. Bot.* **2023**, *154*,365–371.
3. Mutlu, A.; Tas, T. Foliar application of humic acid at heading improves physiological and agronomic characteristics of durum wheat (Triticum durum L.). *J. King Saud Univ. Sci.* **2022**, *34*,102320.
4. Wu, C.; He, C.; Brown, Z. E.; Miljevic, B.; Zhang, C.; Wang, H.; Wang, B.; Morawska, L.; Ristovski, Z. Light absorption properties of black and brown carbon during the prescribed burning season at an urban background site in Brisbane, Australia. *Atmos. Environ.* **2023**, *313*,120072.
5. Tian, S.; Liu, Q.; Qu, J.; Yang, M.; Ma, Q.; Liu, J.; Shao, P.; Liu, Y. Whole-Transcriptome Analysis on the Leaves of Rosa chinensis Jacq. under Exposure to Polycyclic Aromatic Hydrocarbons. *Toxics* **2023**, *11*,610.