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Air–water exchange fluxes of phthalates and phenolics xenoestrogens on Xietang River and East Taihu Lake in Suzhou and the potential health risks†

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As endocrine disruptors, xenoestrogens are prevalent in inland lakes and are mainly attributed to atmospheric transportation. There is limited research on the air–water exchange of xenoestrogens in inland waters and the consequent health effects. This study investigated the air–water exchange process of selected xenoestrogens and associated health risks in adults by examining water and PM_{2.5} samples from the East Taihu Lake and Xietang River in Suzhou. The mean concentration of total xenoestrogens in surface water and PM_{2.5} were $40.30 \pm 7.19 \text{ ng L}^{-1}$ and $460.13 \pm 31.87 \text{ ng m}^{-3}$ (East Taihu Lake), $536.69 \pm 99.62 \text{ ng L}^{-1}$ and $63.93 \pm 3.82 \text{ ng m}^{-3}$ (Xietang River), respectively. Utilising a two-film model, the air–water exchange flux was calculated, with BPA exhibiting the most comprehensive range among all xenoestrogens from -8.88×10^9 to -1.01×10^{10} near East Taihu Lake and -1.32×10^{10} to -1.13×10^{10} ($\text{ng m}^{-2} \text{ d}^{-1}$) near Xietang River. Different xenoestrogens displayed various air–water exchange directions. DBP shows the highest dry deposition fluxes of 9373.26 ± 611.59 near East Taihu Lake and 478.97 ± 48.00 ($\text{ng m}^{-2} \text{ d}^{-1}$) near Xietang River. The study also assessed the non-dietary exposure risk of six xenoestrogens in PM_{2.5} for adults, concluding that no non-cancer risks were identified, with a hazard index below 1. DEHP concentration is within an acceptable level of carcinogenic risk (incremental lifetime cancer risk $<10^{-6}$). Results from this study underscore the importance of developing and implementing xenoestrogen management policies in the Taihu Lake Basin.

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Environmental significance

This research investigates the occurrence of xenoestrogens in freshwater systems in Suzhou, with a specific focus on the environmental mechanisms of air–water exchange. It also evaluates the potential health risks associated with inhalation of these substances, providing information on the effects of human actions on ecosystems and human health. The results emphasize the importance of implementing policies for the management of xenoestrogens, particularly in the Taihu Lake Basin, to reduce hazards and protect both the environment and public health.

1. Introduction

Xenoestrogens, such as phthalates (PAEs), bisphenols (BPs) and alkylphenols (APs), share structural similarities to estrogen and are commonly found in plastics, pesticides, chemicals, and water resources, contributing to pollution in the atmosphere

and surface water.^{1–4} These compounds are widely distributed in the environment and food, posing a global exposure risk to populations.⁵ Even at low concentrations in water, they disrupt the endocrine system in wildlife.⁶ Laboratory tests have revealed that certain xenoestrogens exhibit estrogen-like activity.⁷ Concerns regarding their impact on infants and children's endocrine function have prompted Bisphenol A (BPA) prohibition in baby bottle production in Europe, Canada and the United States.⁷ Exposure to xenoestrogens can lead to obesity, reproductive disorders, and genetic effects on sperm, which ultimately affects sperm quality and spermatogenesis.^{8,9}

Xenoestrogens primarily originate from human activities, including industry, transportation, commercial enterprises, and volatilisation from water sources.^{10–12} They enter water bodies through atmospheric deposition, direct releases, and urban runoff, making it essential to have a comprehensive understanding of their transport dynamics between the

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atmosphere and surface water, which significantly impacts the distribution and circulation of these compounds.^{13–15} Studying the air–water exchange of xenoestrogens is important as it plays a crucial role in comprehending and managing their transmission, distribution, and ultimate fate in the environment. As an emerging environmental pollutant, xenoestrogens can infiltrate the water system through various pathways, such as atmospheric deposition and river transport.¹⁶ These pollutants accumulate in water bodies and can be re-released into the atmosphere, impacting global biogeochemical cycles and ecosystem health.¹⁷

Xenoestrogen can be transported over vast distances through long-range atmospheric transport and water body flow, and studying air–water exchange aids in understanding the dispersion and redistribution of these pollutants globally.¹⁸ In the context of urban rivers, Chandra and Chakraborty.¹⁹ indicated that air–water exchange fluxes play an essential role in determining the fate of PAEs in a tropical urban riverine catchment. Moreover, atmospheric dry deposition should not be overlooked when studying the air–water transport in xenoestrogens.¹⁶ Historical data reveal that dry deposition is the primary source of organic pollutants in significant water bodies such as the Great Lakes, with dry deposition occurring 1.5 to 5.0 times higher than wet deposition.²⁰ Most studies examining the exchange of xenoestrogens at the air–water interface have focused on the ocean, with limited research on this process in inland surface water. Mi *et al.*²¹ demonstrated that seasonal meteorological variations influence the exchange process at the air–sea interface, leading to the long-distance migration of PAEs from land to the ocean, and different PAEs exhibit unique directions in air–sea exchange. Xie *et al.*¹² conducted a study of APs on the North Sea, highlighting the deposition, especially during winter, and the subsequent re-volatilisation into the atmosphere in warmer seasons with increasing temperatures and humidity.

Inland surface waters, such as rivers, lakes and wetlands, are inevitably impacted by human activities, including industrial discharges, agricultural water use, urban development and wastewater discharges.²² The input of pollutants from these anthropogenic activities directly affects water quality and disrupts the structure and function of aquatic ecosystems, exposing them to significant environmental stresses.^{23,24} In Suzhou, over 40% of the city's total urban area is covered by surface water, but the slow flow of these water bodies hinders the migration of non-point source pollutants in surface water and thus causes high concentrations of local pollutants.^{25,26} The Xietang River, located within a densely populated region, was chosen as the monitoring site for this study. Previous investigations by Wu *et al.*²⁷ and Yuan *et al.*²⁸ have detected the presence of organochlorine pesticides (OCPs) and polycyclic aromatic hydrocarbons (PAHs) in the sediments of this river. As a comparison, Taihu Lake, which is classified as a class IV drinking water source for Suzhou, was selected as the reference sampling site.²⁹

Previous studies have demonstrated that toxic pollutants entering surface water or the atmosphere through air–water exchange pose a significant threat to human health and the

environment in urban areas, thereby increasing health risks for residents.^{30,31} Kim *et al.*³² revealed that, particularly in non-metropolitan areas, the risk of cognitive decline associated with air pollution exposure is more pronounced in women. While there has been considerable epidemiological and toxicological research on xenoestrogens in China, many studies have predominantly concentrated on indoor air pollution.^{33–35} Moreover, existing literature has primarily targeted specific urban areas, such as Beijing, Nanjing, Tianjin, Shenzhen and Tibet, leaving cities such as Suzhou underexplored.^{36–40} Additionally, previous toxicological investigations have focused mainly on vulnerable and occupationally exposed populations, which may not adequately represent the exposure levels of the broader urban population.^{38,40,41}

In this study, we aim to explore the exchange flux of phthalates and phenolics xenoestrogens on Xietang River and East Taihu Lake in Suzhou, to understand the impact factors and evaluate the potential health risks to human. The main research objectives of this study were: (1) to monitor the concentration of xenoestrogens in atmospheric aerosols at the research site; (2) to identify the critical environmental factors which had impact on the exchange flux; (3) to assess the potential health risks to the population of different genders at the research site. This study fills the knowledge gap regarding the environmental behaviour and transport mechanisms of these xenoestrogens in inland surface waters, providing insights into pollution management and mitigation strategies. We also highlight the need for gender-specific risk assessments in environmental health studies. These findings are expected to promote theoretical research and provide references for practical applications in atmospheric pollution monitoring, environmental risk assessment, and policy making.

The main research contents of this study focused on (1) monitoring xenoestrogen concentration in air and water samples; (2) calculating the air–water exchange fluxes using the two-film model, with discussion on impact factors; (3) conducting health risk assessments and evaluating non-carcinogenic and cancer risks.

2. Experimental

2.1 Water and aerosol sampling

Two sampling areas in Suzhou were selected for this study: The Xietang River (A densely populated area) and the East Taihu Lake. The sampling points distribution is shown in Fig. 1 (detailed explanation is provided in ESI†). The East Taihu Lake represents the conditions of extensive inland shallow lakes influenced by both natural and anthropogenic factors^{42,43} In contrast, the Xietang River reflects the environmental changes occurring in urban aquatic ecosystems under significant human pressure.⁴⁴ Additionally, xenoestrogens are more prevalent in the atmosphere during summer, a pattern linked to the release of various materials, indoor ventilation, industrial activities, and atmospheric chemical processes, all of which significantly impact air–water exchange dynamics.^{45,46} Therefore, the surface water was sampled on the 22nd July and 5th August in the Xietang River and 26th June and 3rd July in the



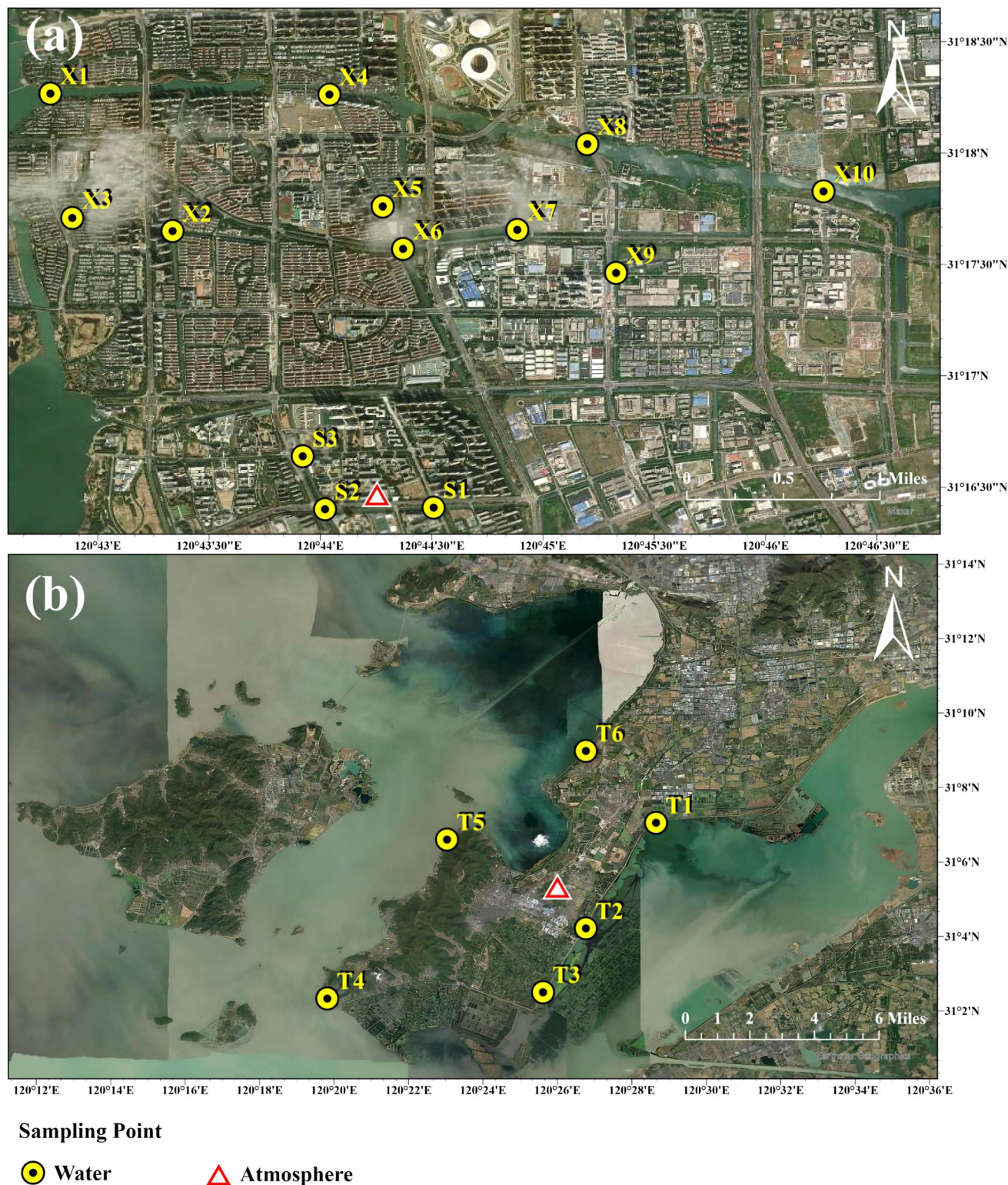


Fig. 1 Atmospheric and water sampling sites: (a) Xietang River (b) East Taihu Lake.

East Taihu Lake in 2022, respectively. Atmospheric particulate matter ($\text{PM}_{2.5}$, particles with an aerodynamic diameter of $\leq 2.5 \mu\text{m}$) was sampled from 22nd June to 10th August in Xietang River and from 21st June to 5th July 2022 in the East Taihu Lake, respectively (Detailed site information is provided in ESI, Table

S1†). The method for collecting water samples is closely similar to the methodology employed in our previous research.⁴⁷ A portable water quality analyser (YSI ProPlus, USA) was taken to measure *in situ* physical-chemical parameters, including dissolved oxygen (DO), conductivity, ammonia nitrogen ($\text{NH}_4^+ - \text{N}$),

nitrate nitrogen (NO_3^+-N) and pH.⁴⁸ Three parallel water samples were obtained from the near-shore zone and filtrated with microporous filtration membranes (Jinteng, China) by solvent filtration/degassing systems (Jinteng, China).

High-volume air samplers (TH1000CII, Tianhong, China) were applied to collect $\text{PM}_{2.5}$ aerosol samples with a sampling time of 24 hours. Aerosol was collected on preheated (450 °C for six hours) quartz fibre filters (203 mm × 254 mm, Whatman) at a 1.05 m³ min flow rate. All the filters were wrapped in pre-heated aluminium foil and stored in a dark freezing room at −20 °C before the analysis.

2.4 Sample pretreatment and instrumental analysis

2.4.1 Water sample pretreatment. The pretreatment procedure was optimised and implemented based on our previous research.⁴⁷ The collected samples were stored in a 500 mL brown glass bottle and filtered out to remove substances greater than 0.45 µm through microporous filtration membranes. The solid-phase extractor (waters, USA) combined with the C₁₈ solid-phase extraction cartridge (Sep-Pak 6cc, 500 mg, Waters, USA) for PAEs and HLB solid-phase extraction cartridges (Oasis HLB 3cc, 60 mg, Waters, USA) for BPA, and was activated with 6 mL of methanol (HPLC purity, Adamas, China) and 10 mL of ultrapure water sequentially, loaded with 500 mL of the above water samples at controlled flow rates of 10–15 mL min^{−1}, and 10 mL of *n*-hexane solution for PAEs analysis and 10 mL of methanol for phenolics were added to elute the cartridges. The eluate was collected with a 15 mL glass tube, concentrated to about 1 mL with a gentle nitrogen stream by using a nitrogen blowing instrument (Organomation, USA), filtered using a 0.22 µm syringe filter (Titan, China) and transferred to a 2 mL brown injection vial, concentrated to near dryness under a gentle nitrogen stream. Finally, 25 µL of methanol for phenolics analysis and 25 µL of *n*-hexane for PAEs were added for redissolve, vortexing and shaking for 1 min. Subsequently, the sample was subjected to analysis using gas chromatography-mass spectrometry (GC-MS) (Agilent, USA) and High-Performance Liquid Chromatography (HPLC) (1260, Agilent, USA).

2.4.2 Atmospheric sample pretreatment. The filter was fragmented using scissors before undergoing the solvent extraction procedure. In the analysis of PAEs, PAEs-*D*₄ internal standard solutions (10 µg mL^{−1}) were added, and 100 µL of 10 µg mL^{−1} standard solution was added to the spiked sample. Subsequently, 6 mL of deionised water and 10 mL of *n*-hexane were added and mixed with an IKA-KS501 stirrer (Germany). The resulting solution was then subjected to ultrasonic treatment for 30 minutes to facilitate extraction. Subsequently, the solution was centrifuged using an Allegra X-15R centrifuge (BECKMAN, USA), and the upper supernatant was collected for analysis using GC-MS (Thermo Fisher Scientific, USA). In the case of BPA and APs, a 100 ng mL^{−1} internal standard solution was employed. To determine the recovery rate, a 100 ng mL^{−1} standard solution was added and thoroughly mixed for spiked samples. The samples underwent two rounds of ultrasonic extraction with acetonitrile, and the upper acetonitrile phase

was collected after centrifugation. The resulting supernatant was transferred to a glass tube for nitrogen evaporation, followed by dissolution of the residue in 1 mL of methanol. The solution was then passed through a 0.22 µm hydrophobic PTFE membrane and analysed using Liquid Chromatography-Mass Spectrometry (LC-MS) (8050, Shimadzu, Japan).

2.4.3 Instrument analysis. The levels of PAEs were analysed using GC-MS with GC 7890B and MS 5977A (Agilent, USA) for the water sample. The chromatographic column was HP-5MS (30 m × 0.32 mm × 0.25 mm) quartz capillary column, the injection port temperature was 250 °C, the injection method was the splitless injection of 1 µL, the program temperature was 60 °C for 1 min. The temperature was raised to 220 °C at 20 °C min^{−1} for 1 min, and then the temperature was raised to 280 °C at 5 °C min^{−1} for 4 min. The carrier gas is 99.999% high-purity helium, and the flow rate is 1 mL min^{−1}. The mass spectrometry ion source was the EI source, the ion source temperature was 230 °C, the ionisation energy was 70 eV, the quadrupole temperature was 150 °C, and the solvent delay was 6 minutes. The scanning mode was ion selection mode, and the selected ion monitoring selected one quantitative ion and four qualitative ions for each compound, respectively: dimethyl phthalate (DMP) (163; 163 : 77 : 135 : 194), diethyl phthalate (DEP) (149; 149 : 177 : 121 : 222), dibutyl phthalate (DBP) (149; 149 : 223 : 205 : 121), benzyl butyl phthalate (BBP) (149; 149 : 91 : 206 : 238), di(2-ethylhexyl)phthalate (DEHP) (149; 149 : 167 : 219 : 113). As for detecting PAEs in atmospheric particulate matter samples, the Thermo Trace 1300 Series Gas Chromatograph with ISQ MS was used with the same scanning mode and selected ion, and the primary parameter setting is on ESI, Table S2.†

HPLC was used for the analysis of BPA in water using an ODS C₁₈ column (250 mm × 4.5 mm, 5 µm) with a column temperature set to 25 °C and an injection volume of 10 µL. The mobile phase was water-methanol (3 + 1), and the flow rate was set to 0.8 mL min^{−1}. A fluorescence detector was used, with an absorption wavelength of 227 nm and an emission wavelength of 313 nm. As for the BPA and APs in the particulate matter sample, LC-MS with acclaim polar Advantage II (50 mm × 3 mm, 3 µm) column was used for xenoestrogens analysis. Parameters are shown in ESI, Table S3.† The standard curves of all phenolic compounds reached $R^2 > 0.9999$.

2.5 Quality assurance/quality control

The target contaminants in this research are usually used as plasticisers, commonly present in most plastic products. Consequently, it is necessary to utilise glassware for all experimental procedures and to employ ultrapure water and *n*-hexane for ultrasonic cleaning before each use of experimental supplies. Three replicates of each water sample and atmosphere particulate matter sample were set up to ensure the accuracy of the results. Furthermore, the accuracy of the experiment is ensured by the recovery test and the blank test (including samples blank and solvents blank). The assay results showed that all solvents and samples blank were below the detection limits, and the sample recovery rate was within acceptable limits (73.8–123.0%). The Limits of Detection (LODs) ranges



from 0.88–2.17 ng L⁻¹ for PAEs and 0.09–0.12 ng L⁻¹ for phenolics. The Limit of Quantitation (LOQ) values are 2.94–7.22 ng L⁻¹ for PAEs and 0.30–0.40 ng L⁻¹ for phenolics. Detailed data for each target pollutants are shown in the ESI, Table S4.†

2.6 Air–water exchange fluxes

A two-film model was conducted to calculate plasticisers' air–water particulate exchange flux (F_{aw} , ng m⁻² d⁻¹). The exchange flux was defined by Whitman (1962).⁴⁹

$$F_{aw} = K_{ol} \times \left(C_w - \frac{C_a}{H'} \right) \quad (1)$$

K_{ol} is the overall mass transfer coefficient (m d⁻¹), and C_w and C_a are concentrations in the water (ng m⁻³) and atmosphere (ng m⁻³), respectively. H is the Henry's Law Constant, and H' is the dimensionless Henry's Law constant corrected by temperature and salinity, calculated as H/RT , where R is the gas constant (8.314 Pa m⁻³·(K mol⁻¹)⁻¹). T is the temperature (K). Typically, a positive flux indicates the transfer of xenoestrogens from surface water to the atmosphere, while a negative flux indicates the deposition of xenoestrogens from the atmosphere into surface water. Recently, fugacity fraction (ff) has been applied to evaluate the equilibrium state and direction of exchange between atmospheric particulate matter and water phases.⁵⁰

When applied the ff for air–water exchange analysis, the uncertainty (u) of ff was also calculated, and ff values in the range of $0.5 \times (1 \pm u)$ may be considered equilibrium.⁵⁰ In this study, fugacity fraction values falling below 0.26 indicate volatilisation, values between 0.26 and 0.74 signify air–water equilibrium, and values exceeding 0.74 represent deposition. Detailed formulas are shown in the ESI, Section 2.†

The dry deposition fluxes (F_{dd} , ng m⁻² d⁻¹) were calculated according to the following equation:⁵¹

$$F_{dd} = C_p \times V_d \quad (2)$$

V_d is the deposition velocity of atmospheric particles (m d⁻¹), and C_p (ng m⁻³) is the particle concentration of plasticisers. A value of 0.2 cm s⁻¹ for V_d was adopted to estimate plasticisers, which is recommended for semi-volatile organic compounds (SVOCs) and widely applied in China.^{52–54}

2.7 Health impact assessment

After determining the concentration of plasticisers in the atmosphere particulate matter, the probabilistic exposure model is used to evaluate the average daily dose (ADD) in mg per kg per day and incremental lifetime cancer risk (ILCR) of DEHP *via* inhalation and dermal contact.^{55,56} The formula for ADD of inhalation and dermal contact exposure was shown in the ESI, Section 3.†

The hazard index (HI) for non-dietary intake was determined by following the formula.⁵⁶ RfD is each pollutant's chronic exposure reference dose with the mg per kg per day unit. Humans are exposed to non-cancer risks if the values of HI are greater than 1.

$$HI_{\text{non-dietary}} = \frac{ADD_{\text{inhalation}}}{RfD_{\text{inhalation}}} + \frac{ADD_{\text{dermal}}}{RfD_{\text{dermal}}} \quad (3)$$

ILCR is the incremental lifetime cancer risk associated with inhalation and dermal contact, which was calculated by:^{55,57}

$$ILCR = \sum ADD_{\text{inhalation,dermal}} \times CSF \times \left(\frac{BW}{70} \right)^{\frac{1}{3}} \quad (4)$$

CSF is the cancer slope factor 0.014 per mg per kg per day for DEHP *via* inhalation or dermal contact.⁵⁸

2.8 Statistical analysis

Statistical analyses were performed utilising R Software Version 4.1.3. EPIWEB v4.1 was employed to model the parameters of target plasticisers. By quantifying the levels of xenoestrogens in particle phases and estimating absorbed fractions in airborne particulates (Φ), it becomes feasible to predict gaseous concentrations (ng m⁻³) and subsequently determine the overall transfer of xenoestrogens between air and water in the Taihu basin region. The relationship between the particle-bound fraction, gaseous concentrations, and particle-bound concentrations of xenoestrogens is explained by the subsequent eqn (5):⁵⁹

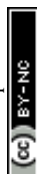
$$\Phi = \frac{C_p}{C_g + C_p} \quad (5)$$

where Φ is the particle-bound fraction, C_p and C_g are the particle-bound and gaseous concentrations of xenoestrogens. Adsorbed Fraction (Φ) can be calculated using the AEROWIN software, which utilises the Junge-Pankow partitioning model.⁶⁰ Additionally, Pearson correlation analysis was carried out using R Software. Statistical significance was considered for p -values less than 0.05.

3. Results and discussion

3.1 Concentration of xenoestrogen in surface water and atmosphere

The concentrations of specific xenoestrogens in the surface water are shown in Fig. 2. Among the six xenoestrogens analysed, DBP, DEHP and BPA were present in all samples from East Taihu Lake, as illustrated in Fig. 2a. The detection rates for DEP, DMP and BBP in East Taihu Lake were approximately 25%, 83% and 67%, respectively. Xenoestrogens typically exhibit concentrations ranging from several to several tens of nanograms per litre, with a total concentration between 12.75 and 93.86 ng L⁻¹ and an average concentration of 40.30 ± 7.19 ng L⁻¹. In the water bodies of East Taihu Lake, the mean levels of DBP and DEHP exceeded those of three other PAEs (DEP, DMP, BBP) during these two days. This finding is consistent with a previous study conducted by Gao *et al.*⁶¹ in Taihu Lake, where DBP and DEHP were identified as the predominant PAEs in surface water in 2014, with average concentrations exceeding 1000 ng L⁻¹, suggesting a positive trend towards decreased xenoestrogen levels in the Taihu Lake over time. In Fig. 2b, all



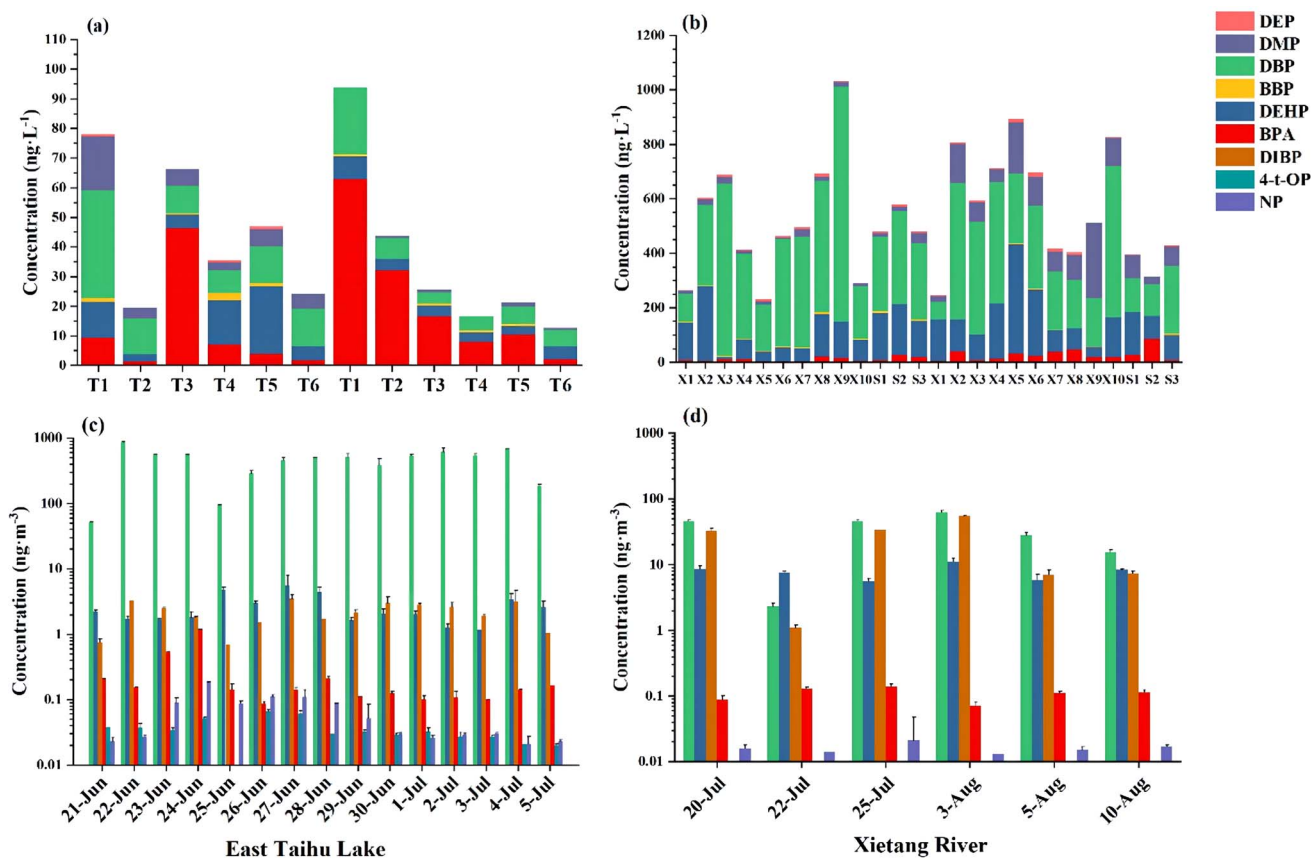


Fig. 2 Concentrations of Xenoestrogen in surface water samples in East Taihu Lake (a) and surface water samples in Xietang River (b), in Particulate Matters (PMs) at various sampling times near East Taihu Lake (c) and in PMs at various sampling times near Xietang River (d).

samples collected from Xietang River showed the presence of six targeted xenoestrogens, with the total xenoestrogens levels ranging from 231.82 to 1030.90 ng L⁻¹ and averaging at 536.69 ± 99.62 ng L⁻¹ in Xietang River. DEHP and DBP were also identified as the primary xenoestrogens. The combined concentrations of the five PAEs were similar to those observed in the Yangtze River (Jiangsu section) in research by Ren *et al.*,⁶² ranging from around 300 to 1000 ng L⁻¹. Zhao *et al.*⁶³ conducted a study that revealed a higher mean concentration of total PAEs in the Lanzhou Section of the Yellow River, measuring approximately 1900 ng L⁻¹. Yuan *et al.*⁶⁴ reported that BPA levels at the river in Suzhou were approximately 100 ng L⁻¹, predominantly originating from industrial discharges and higher than the concentrations observed in the present study. This difference indicates a decline in BPA concentration in surface water in Suzhou. Furthermore, the average concentration of BPA in the Xietang River is comparatively lower than that in the river Ganga (over 100 ng L⁻¹) and Xiangjiang River (around 37 ng L⁻¹) in 2022, which is resulted from the more stringent monitoring and regulation of domestic and industrial wastewater discharge in the Xietang River.^{65,66}

This study also conducted a comparative analysis of contamination levels of neighbouring compounds, specifically DBP, DEHP and BPA, in East Taihu Lake and Xietang River in China, with data from other inland surface waters globally (see

ESI, Table S5†). The results indicated that the concentrations of xenoestrogens in East Taihu Lake were relatively low. Conversely, the Xietang River exhibited elevated levels of DBP, ranging from 163 to 298 ng L⁻¹, and DEHP, ranging from 111 to 163 ng L⁻¹, which were comparable to concentrations observed in certain water bodies in Africa, specifically Nigeria and Uganda. In contrast, regions in Europe, such as the Netherlands and Italy, reported significantly higher levels of BPA pollution, with concentrations reaching up to 3620 ng L⁻¹, while the BPA concentrations in the study area were comparatively low, ranging from 12 to 39 ng L⁻¹. Overall, the pollution levels in the areas examined in this study are considered moderately low on a global scale. However, the contamination observed in the Xietang River necessitates further investigation. Therefore, it is recommended that future analysis focus on pollutant source identification and ecological risk assessments in the Xietang River to optimise pollution control strategies.

The presence of DEHP, di-isobutyl phthalate (DIBP), DBP, BPA, nonylphenol (NP), and 4-*tert*-octylphenol (4-*t*-OP) have been detected in all PM_{2.5} samples collected from East Taihu Lake and Xietang River (Fig. 2). The concentrations of total xenoestrogen (sum of DEHP, DIBP, DBP, BPA, NP and 4-*t*-OP) bound to PM_{2.5} ranged from 54.45 to 863.40 ng m⁻³ (mean = 460.13 ± 31.87 ng m⁻³) near the East Taihu Lake and 11.13 to 127.84 ng m⁻³ (mean = 63.93 ± 3.82 ng m⁻³) near Xietang



River. Higher concentrations of DBP (ng m^{-3}) were observed at East Taihu Lake (range: 51.16–858.17) compared to DIBP, DEHP, BPA, 4 *t*-OP and NP, which exhibited ranges of 0.70–3.49, 1.16–5.47, 0.09–1.19, below the limit of quantification (<LOQ)–0.07 and 0.02–0.18, respectively. Salgueiro-González *et al.*⁶⁷ revealed that DBP exhibits the most elevated levels within $\text{PM}_{2.5}$, particularly in suburban and industrial regions, surpassing other xenoestrogens by several orders of magnitude, which may be linked to industrial processes such as plastic production. Temporally, the Xietang River exhibited higher levels of DBP (range: 7.73–14.30 ng m^{-3}), DIBP (range: 21.05–48.05 ng m^{-3}) and DEHP (range: 4.56–9.10 ng m^{-3}) concentrations compared to BPA (range: 0.07–0.14 ng m^{-3}), 4 *t*-OP (<LOQ) and NP (0.01–0.02 ng m^{-3}). A study conducted in Guangdong Province by X. Liu *et al.*⁶⁸ observed a similar concentration distribution trend in $\text{PM}_{2.5}$ with DBP ranging from 0.78 to 11.20 ng m^{-3} , DIBP from 0.86 to 9.92 ng m^{-3} , DEHP from 1.21 to 82.60 ng m^{-3} and BPA from 0.01 to 1.65 ng m^{-3} .

The levels of xenoestrogens in $\text{PM}_{2.5}$ and water samples from both sampling exhibit an observable opposite trend (Fig. 2). Specifically, the concentration of xenoestrogens on $\text{PM}_{2.5}$ above East Taihu Lake was higher, whereas Xietang River showed a higher concentration in water samples of these compounds. Previous research by Liu *et al.*⁶⁹ indicated that urban rivers tend to have elevated concentrations of PAEs due to human activities, while suburban surface water shows significantly lower levels compared to urban and industrial areas. Similarly, Ma *et al.*⁷⁰ observed that the average concentration of $\text{PM}_{2.5}$ -bound PAEs was higher in suburban than urban areas. Furthermore, the average wind speed during the sampling period was 5.09 m s^{-1} for Xietang River and 1.10 m s^{-1} for East Taihu Lake in this study. Consequently, the high wind speeds were conducive to the dispersion and dilution of particulate matter and its associated xenoestrogens.⁷¹

3.2 Air–water exchange fluxes of xenoestrogen

DBP, DEHP and BPA were detected in water and aerosol samples in East Taihu Lake and Xietang River, and their air–water exchange fluxes were calculated. Eqn (5) was used to calculate the gaseous concentration of xenoestrogens based on the measured concentration of $\text{PM}_{2.5}$ particles (Table 1). The findings in Table 1 indicate that the estimated concentrations of DBP and BPA in the gaseous phase are higher than in the particulate phase, while DEHP shows an opposite trend. According to the findings of Teil *et al.*,⁷² most xenoestrogens exhibit a preference for the gaseous phase, particularly during summer months characterised by elevated outdoor temperature and reduced concentrations of PMs. In this context, DEHP, classified as a long-chain PAEs, possesses a higher octanol–air partition coefficient (K_{OA}), which enhances its distribution in the particulate phase. Conversely, DBP, classified as a short-chain PAE, is characterised by low K_{OA} values.^{73,74} Furthermore, Lee *et al.*⁷⁵ conducted a study investigating the presence of PAEs in both the water and atmosphere of Asan Lake in Korea. Their results indicated that the concentration of DBP in the water and atmosphere was measured at 30 000 ng m^{-3} and 3.62 ng m^{-3} ,

Table 1 Water and atmosphere concentration (gaseous and particulate phases ($\text{PM}_{2.5}$)) of each xenoestrogen on East Taihu Lake and Xietang River

			C_{a} (ng m^{-3})	
			C_{w} (ng m^{-3})	Particulate Gaseous
DBP				
East Taihu lake	26th June	15 085	290.25	7171.16
	3rd July	8243	542.43	13 401.88
Xietang river	22nd July	297 849	2.33	57.52
	5th August	163 196	27.72	684.83
DEHP				
East Taihu lake	26th June	10 235	2.97	0.52
	3rd July	4266	1.16	0.20
Xietang river	22nd July	163 485	7.57	1.32
	5th August	111 176	5.82	1.02
BPA				
East Taihu lake	26th June	11 572	0.09	0.49
	3rd July	22 000	0.10	0.56
Xietang river	22nd July	17 497	0.13	0.73
	5th August	39 255	0.11	0.63

respectively. In comparison, DEHP concentrations were found to be 110 000 ng m^{-3} in water and 6.52 ng m^{-3} in the atmosphere. Lee *et al.*⁷⁵ also noted that DBP predominantly exists in the gaseous phase, while DEHP is primarily associated with the particulate phase, which is similar with our results. It is noteworthy that, as an inland lake, the environmental concentrations of these compounds in East Taihu Lake are generally lower than those of Asan Lake, with the exception of the atmospheric concentration of DBP.

The analysis further demonstrates that the net flux of air–water exchange for gaseous xenoestrogens is approximately 1.1 to 35.6 times greater than that of $\text{PM}_{2.5}$ -bound xenoestrogens. This indicates that gaseous xenoestrogens have a more substantial influence on the total net air–water exchange process. The distribution of pollutants between gaseous and particle phases is crucial in determining their environmental fate and long-distance transportation.⁷⁶ Gaseous xenoestrogens significantly affect air–water exchange because high ambient temperatures and solar radiation in summer will promote the volatilisation of xenoestrogen from particles to the gas phase.⁷⁷ As PAEs with high vapour pressure and Henry's Law Constant, DBP tends to be distributed in the gas phase rather than bound to particles.^{78,79}

Fig. 3 and 4 illustrate the air–water exchange fluxes of DBP, DEHP, and BPA in East Taihu Lake and Xietang River, which all showed day-to-day variations, possibly because the difference in atmosphere concentration for DBP ($R = -0.99$, $p < 0.05$), DEHP ($R = 0.97$, $p < 0.05$) and BPA ($R = -0.99$, $p < 0.05$), and the concentration discrepancy of DEHP in water are also significant impact the daily air–water exchange fluxes ($R = 0.99$, $p < 0.05$).

The air–water exchange flux ($\text{ng m}^{-2} \text{d}^{-1}$) of individual xenoestrogens on 26th June and 3rd July in East Taihu Lake around -7.89×10^9 and -1.48×10^{10} for DBP (Fig. 3a) ranged from 1.84×10^4 to 9.93×10^5 and 9.64×10^4 to 3.39×10^5 for DEHP



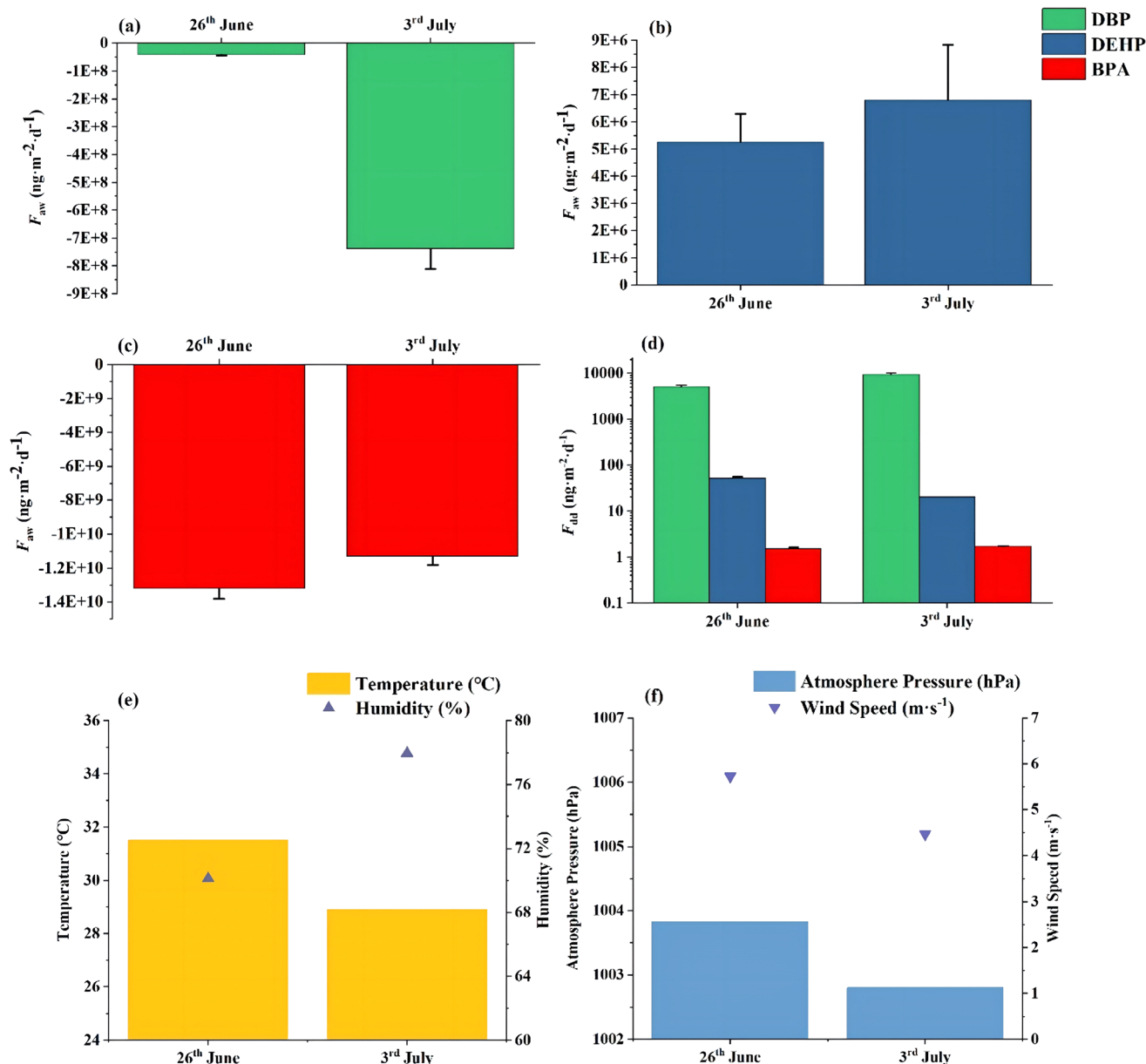


Fig. 3 Air–water exchange fluxes for DBP (a), DEHP (b) and BPA (c), their dry deposition fluxes (d) and meteorological condition (e and f) in East Taihu Lake.

(Fig. 3b), around -8.88×10^9 and -1.01×10^{10} for BPA (Fig. 3c), respectively. The net flux (ng m⁻² d⁻¹) of air–water exchange in Xietang River for DBP varied from -5.74×10^7 to -1.44×10^7 and -7.50×10^8 to -7.22×10^8 on 22nd July and 5th August, respectively (Fig. 4a). Furthermore, a net exchange flux range of 1.70×10^5 to 1.30×10^7 ng m⁻² d⁻¹ and 1.65×10^6 to 1.92×10^7 ng m⁻² d⁻¹ was observed for DEHP on 22nd July and 5th August (Fig. 4b). Regarding the exchange flux of BPA on the Xietang River (Fig. 4c), the flux is around -1.32×10^{10} ng m⁻² d⁻¹ and -1.13×10^{10} ng m⁻² d⁻¹. The air–water exchange primarily involves deposition from air to water and volatilisation from water to air. Generally, BPA exhibited net deposition from air to water ($ff < 0.26$) at both sites and during various sampling times. While there are currently no publicly available

data for the air–water exchange flux of BPA, Mukhopadhyay and Chakraborty (2021).⁸⁰ reported the exchange of BPA in water and sediment, which indicated that the high solubility of BPA in water could result in its predominant presence in the dissolved phase, with similar trends for DBP in all sites and sampling times.

There has been limited research focus on the air–water exchange flux of DBP and DEHP in surface waters globally. Xie *et al.*⁵⁰ concluded that net deposition was the primary mechanism for the air–water exchange of DBP in the North Sea, while volatilisation was dominant for DEHP. This aligns with our current findings, which indicate that the primary exchange process for DEHP in East Taihu Lake and Xietang River is net volatilisation from water to the atmosphere, whereas DBP

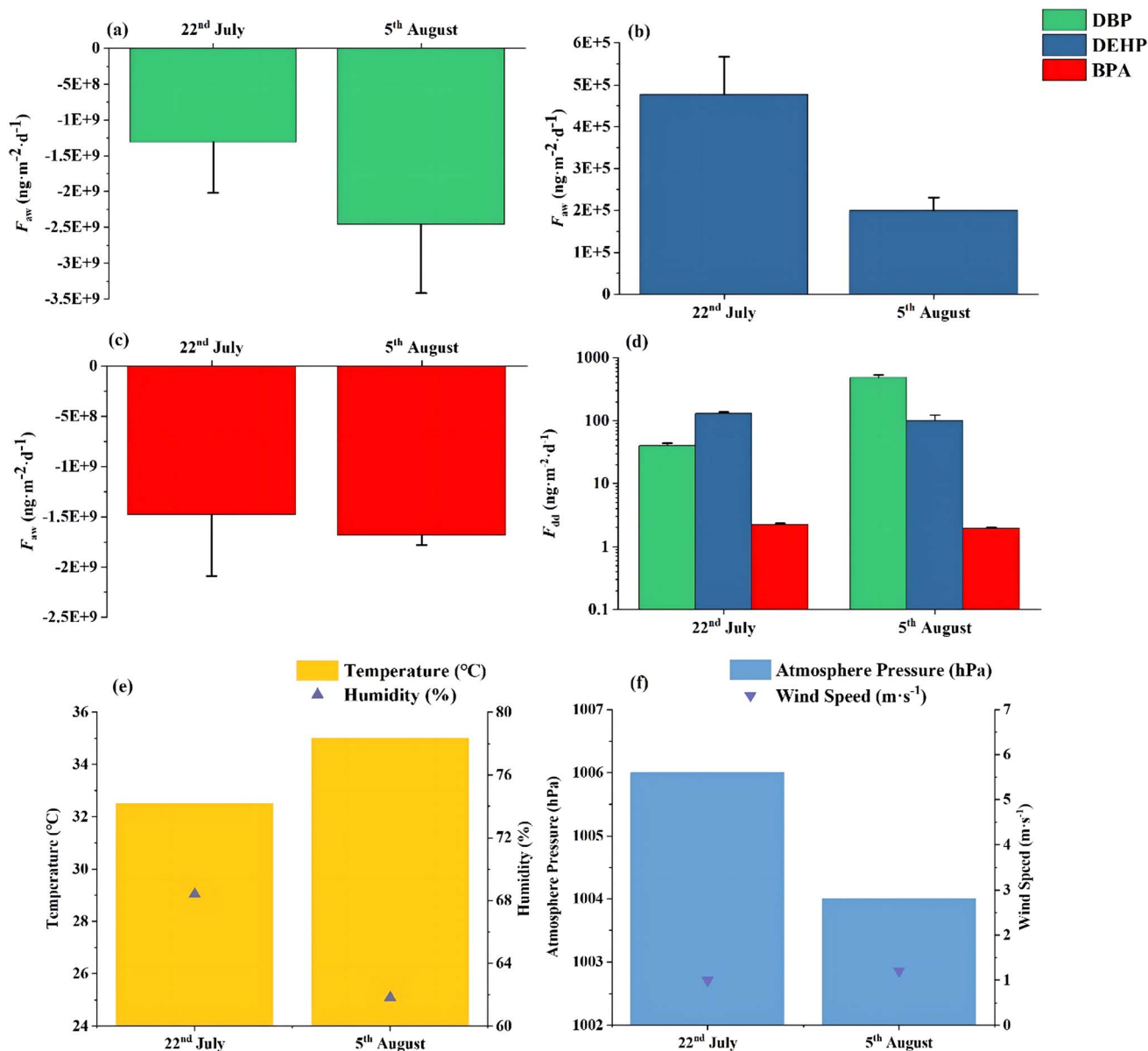


Fig. 4 Air–water exchange fluxes for DBP (a), DEHP (b) and BPA (c), their dry deposition fluxes (d) and meteorological condition (e and f) in Xietang River.

experience net deposition. A study conducted in the South China Sea by Mi *et al.*²¹ also found that DEHP predominately volatilises into the atmosphere, with average fluxes consistent with our observations. This phenomenon may be attributed to the elevated temperatures of the lake waters, which enhance the likelihood of DEHP migrating from water to air due to its high hydrophobicity and low water solubility.⁸¹ Furthermore, research in the Arctic by Xie *et al.*⁸² indicated that DEHP primarily performed net deposition for the air-sea gas exchange, which might result from long-distance transport from land-based sources in European countries.

In India, the air–water exchange of DEHP exhibited spatial variations, with volatilisation occurring from surface water in areas with open burning dumps, while deposition was observed

in industrial and residential zones, possibly related to using DEHP in masks during the COVID-19 pandemic.¹⁹ Consequently, regulatory authorities should consider developing appropriate standards for the incorporation of DEHP in masks.⁸³ Furthermore, considering the sustained demand for mask usage even in the post-pandemic era, it is essential to prioritise centralised and environmentally safe disposal methods for masks rather than relying on direct landfill or incineration practices.^{84–86}

Dry deposition fluxes ($\text{ng} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$) of xenoestrogens were calculated, with the trend indicating $F_{dd}(\text{DBP}) > F_{dd}(\text{DEHP}) > F_{dd}(\text{BPA})$ for both measurements in East Taihu Lake (Fig. 3d). The mean flux values were 5015.50 ± 450.87 and 9373.26 ± 611.59 for DBP, 51.29 ± 4.27 and 20.11 ± 0.02 for DEHP, and

1.51 ± 0.10 and 1.71 ± 0.02 for BPA, respectively. C. Wang *et al.*⁸⁷ and Guo *et al.*⁸⁸ both found that the dry deposition fluxes of DBP were lower than DEHP in the lake at Tianjin according to the model simulation results, identifying the atmosphere to be the primary source of DEHP. Furthermore, the deposition fluxes of DEHP measured in this study are significantly lower than the reported value from Zeng *et al.*⁸⁹ in Guangzhou's urban and suburban areas, possibly proving that DEHP is not the primary pollutant in $PM_{2.5}$ of Suzhou. The average dry deposition flux

($ng\ m^{-2}\ d^{-1}$) of xenoestrogens on the Xietang River were 40.23 ± 3.56 and 478.97 ± 48.00 for DBP, 130.74 ± 7.12 and 100.57 ± 21.34 for DEHP, 2.24 ± 0.10 and 1.93 ± 0.09 for BPA, respectively. The trend of the average flux of DBP, DEHP and BPA was inconsistent in Xietang River compared to East Taihu Lake, with a disparity of DBP and DEHP dry deposition fluxes at different monitoring dates (Fig. 4d). The atmospheric concentration of PAEs is a significant factor influencing the direction and magnitude of air–water exchange processes, as evidenced by

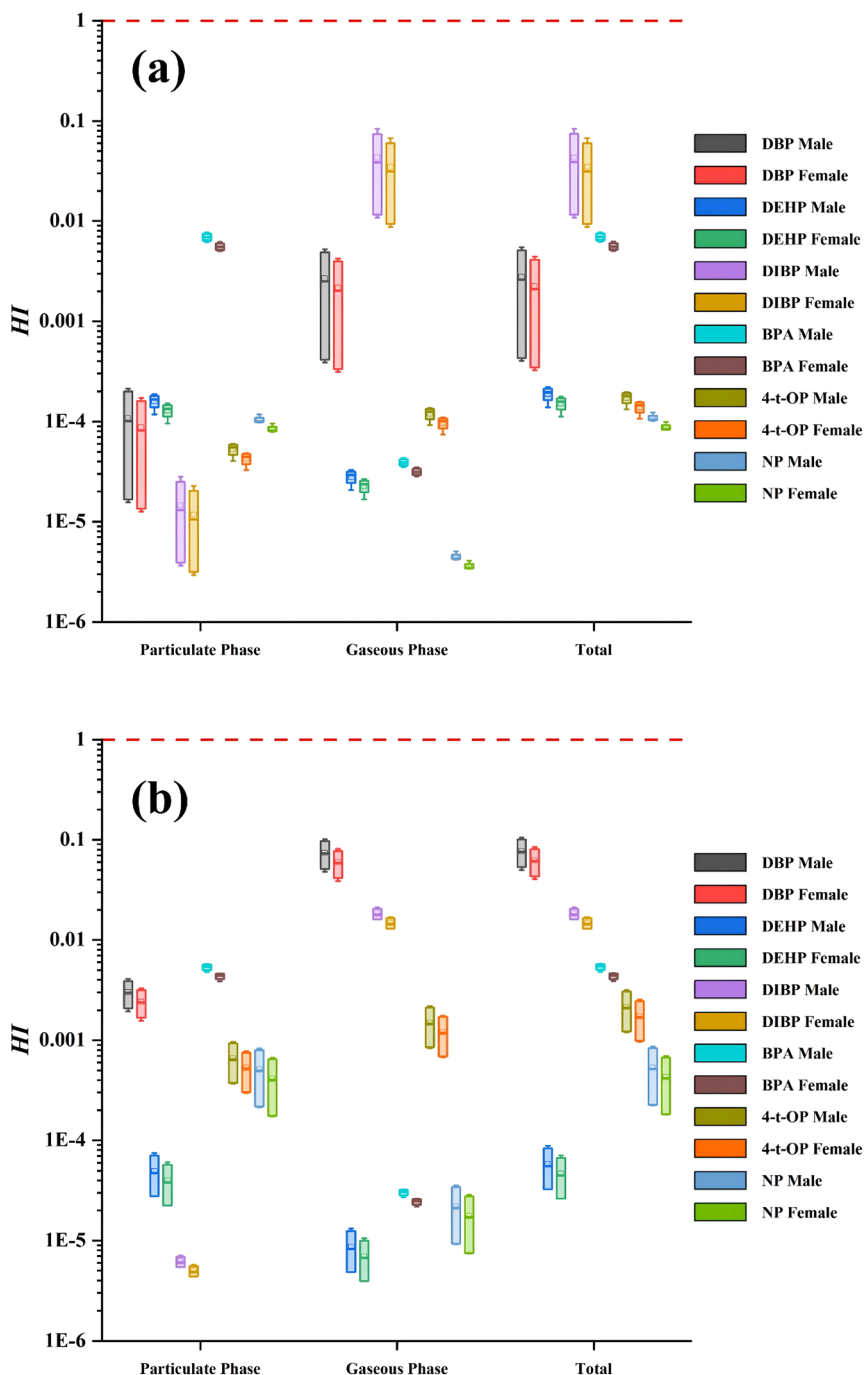


Fig. 5 Health risk assessment of non-dietary xenoestrogen intake near Xietang River (a) and East Taihu Lake (b). The red line is the risk control line ($HI = 1$). The box plot displays the interquartile range (25–75%). The whiskers indicate the range of data, and the hollow square denotes mean values.



variations in the net air–water exchange and dry deposition flux of DBP on different days.²¹ This discrepancy in flux can be attributed to the differing concentrations of DBP adsorbed onto PM_{2.5} at Xietang River (2.33 ng m⁻³ and 27.72 ng m⁻³ shown in Fig. 2d).

Meteorological elements, such as wind speed, temperature, humidity, and atmospheric pressure, could impact the exchange processes occurring at the interface of the atmosphere.⁹⁰ Changes in meteorological conditions could influence the distribution of xenoestrogens between the gaseous and particulate phases, thereby affecting their interface exchange processes.⁹¹ The partitioning of SVOCs between particle and gas is particularly sensitive to fluctuations in temperature.⁹² Furthermore, seasonal variations have also been shown to affect the particle/gas partitioning of SVOCs. However, these effects can vary among different compounds due to the distinct atmospheric reactions of various SVOCs and particle size distribution of their particle states, resulting in diverse behavior characteristics in the atmosphere.⁹³ No significant correlation has been observed between humidity and the partition coefficient of SVOCs.⁹⁴ Results of the Pearson correlation show that the air–water exchange flux of DEHP was significantly negatively correlated with the wind speed ($R = -0.94$, $p < 0.05$) (ESI, Fig. S1†). Zhu *et al.*⁹⁵ have found that wind speed is also negatively correlated with the dry deposition flux of pollutants because the wind speed influences the diffusion of pollutants, which will decrease the concentration of pollutants and then decrease the dry deposition flux.

Significant connections have been noted between the chemical properties of pollutants and the rates of interface exchange in previous research.⁹⁶ Disparities in dry deposition fluxes within the research area may be attributed to variations in the chemical properties of xenoestrogens. For example, in the Xietang River, air–water exchange fluxes were negatively correlated with water

solubility at 25 °C of xenoestrogens in July and August ($R = -0.99$, $p < 0.05$) (ESI, Fig. S2†). The Henry's Law Constant is greatly affected by the water solubility, thus affecting the flux from water to the atmosphere.⁹⁷ Other parameters, such as log K_{ow} , molecular weight, Henry's law constant, and vapour pressure, may also have an impact on the dry deposition fluxes (For detailed information in ESI, Section 5†).^{98–100}

3.3 Health risk assessment

According to the Seventh Census in 2020, the resident population in Suzhou is about 12.75 billion people with a 1:1.09 sex ratio (Female/Male).¹⁰¹ Analysis of the HI for non-carcinogenic health risks associated with xenoestrogens in male and female adults are presented in Fig. 5. The results suggest negligible chronic risks to the population in the study area from non-dietary exposure. Specifically, the non-cancer risk assessment on Xietang River (Fig. 5a) indicates that DIBP exhibits the highest non-cancer risks in the region, with maximum values of 0.08 for males and 0.07 for females. This finding suggests that DIBP poses the most significant non-cancer risks in the Xietang River, potentially due to variations in bioavailability and reference doses. According to Li *et al.*,¹⁰² although DIBP is found at lower concentrations in the air, it exhibits greater bioavailability compared to DBP and DEHP, resulting in a higher daily intake. Additionally, as an industrial substitute for DBP, DIBP is required to have a lower RfD than DBP, which consequently contributes to an elevated HI.¹⁰³ Conversely, DBP in the samples from East Taihu Lake shows higher HI values compared to other xenoestrogens for both genders, with peak values of 0.1 for males and 0.08 for females (Fig. 5b). This suggests that DBP is a more significant contributor to non-cancer risks in East Taihu Lake, as concentration is a critical factor in evaluating the HI associated with non-dietary exposure. The concentration of DBP

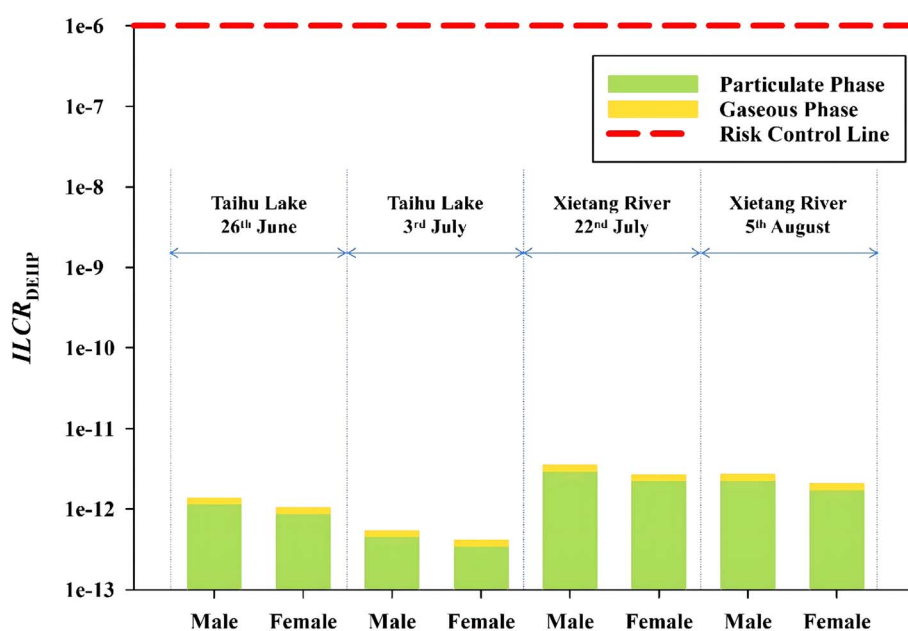


Fig. 6 Carcinogenic risk assessment for non-dietary exposure to DEHP. The risk control line is drawn in red ($ILCR_{DEHP} = 10^{-6}$).



in East Taihu Lake is orders of magnitude greater than that of other xenoestrogens.¹⁰⁴ While the trend of HI values in both Fig. 5a and b suggests that females in the study area may be less susceptible than males, these differences were not found to be statistically significant ($p > 0.05$). Results from Zhang *et al.*¹⁰⁵ have indicated that non-dietary exposure to outdoor dust would not induce non-cancer risk in adults on the Tibetan Plateau. Additionally, the HI resulting from gaseous and particle-phase xenoestrogen in Fig. 5 reveals that the gas-phase DBP, DIBP, and 4 *t*-OP are the primary contributors to the HI values in the Xietang River (Fig. 5a) and East Taihu Lake (Fig. 5b). Furthermore, the particle-phase DEHP, BPA, and NP in the two study regions constitute a significant portion of the non-cancer risk associated with total-phase (gas + particle phase) xenoestrogens.

Non-dietary cancer risk associated with exposure to DEHP is considered an acceptable threshold in health risk assessment at an ILCR_{DEHP} of 10^{-6} . This study presents a summary of ILCR values for males and females across various phases of DEHP, as summarised in Fig. 6. The calculated ILCR_{DEHP} were found to be below 1×10^{-11} for both times and across various sites, indicating very low cancer risks. The research suggests that gaseous DEHP in the study area poses a lower cancer risk than particle phase DEHP. They exhibited a trend of higher values in males than females, although this gender difference was not statistically significant ($p > 0.05$). Maceira *et al.*¹⁰⁶ found a low cancer risk from outdoor respiratory exposure to DEHP, from a respiratory exposure study in Tarragona in Southern Spain, which showed similar conclusions to this study. When assessing the carcinogenic risk of DEHP, the DEHP concentration, exposure frequency, and inhalation rate will significantly determine the evaluation results, while body weight will also affect the evaluation results to a certain extent.¹⁰⁷

4. Conclusion

This research has identified several key findings critical to understanding the behaviour of xenoestrogen in the exchange of air–water interfaces and their potential implications for public health. On the air–water interface, the exchange direction was deposition for DBP and BPA and volatilisation for DEHP. Additionally, the trend of dry deposition generally followed the order of DBP > DEHP > BPA, suggesting significant variations in the behaviour and fate of different xenoestrogens. This study also emphasises the significant impact of wind speed and specific chemical properties on the exchange of xenoestrogens between the atmosphere and water. There were non-carcinogenic and carcinogenic health risks of non-dietary intake routes.

5. Limitations and implications

Several limitations exist in the present study. The estimation of gaseous xenoestrogen concentrations was conducted using the gas/particle partitioning model. However, several sources of uncertainty are associated with the estimation of gaseous pollutant concentrations through the gas-particle partitioning

method. One significant assumption of the gas/particle partitioning model is that the gas and particle system is in a state of equilibrium. In field monitoring, this equilibrium may not be fully reached, which can result in either an overestimation or underestimation of gaseous pollutant concentrations.¹⁰⁸ Additionally, the partitioning of SVOCs between gas and particulate phases is highly sensitive to variations in meteorological conditions, which can further contribute to fluctuations in the estimates of gaseous concentrations.¹⁰⁹ Therefore, future research is essential to refer to actual monitoring data for a more comprehensive analysis of air–water exchange dynamics. The impact of gaseous pollutants on air–water interface exchange should also be emphasised in future, as they could significantly influence the transport and fate of xenoestrogens.

The partitioning of particles and gases is significantly influenced by temperature and seasonal variations. This study lacked seasonal monitoring data of air–water exchange fluxes. Therefore, the dynamic characteristics of those parameters could not be adequately captured, particularly given the differences in reaction characteristics and particle size distribution among various compounds. This may lead to a limited understanding of the behaviour of xenoestrogens under diverse environmental conditions. Furthermore, the lack of long-term monitoring and comparative studies regarding the effects of humidity constrains the interpretation of the results, as the relationship between other meteorological factors and the distribution coefficient of xenoestrogen remains ambiguous. Future research should aim to conduct long-term monitoring at different seasons and evaluate the impacts of various meteorological indicators. The expected results will help reveal the key factors affecting the allocation of xenoestrogens and their interactions. Although current assessments indicate that the health risks associated with inhalation of outdoor air and direct skin contact are below established risk thresholds for adults, it is necessary to consider the cumulative risks for diverse demographic groups and exposure pathways. Additionally, the existing research does not address the collective toxicity of various xenoestrogens present in the atmosphere, highlighting a critical gap that requires further investigation.

Data availability

The data of concentrations presented in the current study are available on reasonable request from the corresponding author. Other data are included in the ESI.†

Author contributions

Minhao Wang: writing – original draft, writing – review and editing, conceptualisation, data curation, formal analysis, investigation, methodology, software, validation, visualisation. Lei Han: writing – review and editing, conceptualisation, funding acquisition, methodology, project administration, resources, supervision, validation. Dongling Li and Ting Tong: data curation, investigation, validation. Ziyu Zhang, Yuwei Xia and Xinhui Shi: investigation, validation. Haifei Zhang: writing



– review and editing, supervision. Kui Chen: resource, validation. Fang Wang and Xiaowei Tie: validation.

Conflicts of interest

There are no conflicts to declare.

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