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Toxic Dust Emission from Drought-Exposed Lakebeds – A New Air Pollution Threat from Dried Lakes

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Abstract

A large number of lakes worldwide are shrinking rapidly due to climate change and human activities. Pollutants accumulated in dried lakebed sediments may be released into the atmosphere as dust aerosols. However, whether lakebed dust carry sufficient toxic materials and exceeds threshold atmospheric concentrations to pose a significant health risk is currently unknown. Recently, Poyang Lake and Dongting Lake, largest lakes in East China, experienced record-breaking droughts with 99 % and 88 % areas exposed to the air. Here, we demonstrate, through field sampling, laboratory simulations, and model validation, that lakebed dust from these lakes could contribute maximum daily PM_{10} concentrations up to 637.5 μg/m^3. Critically, for the first time, we show that the dust generated from lakebeds exceeded regional thresholds for short-term non-carcinogenic risk (HQ=4.13) and Cr carcinogenic risk (~2.10×10^{-6}). These findings also suggest that lakebed dust could have a greater impact on human health as climate change leads to more extreme drought conditions in the future.

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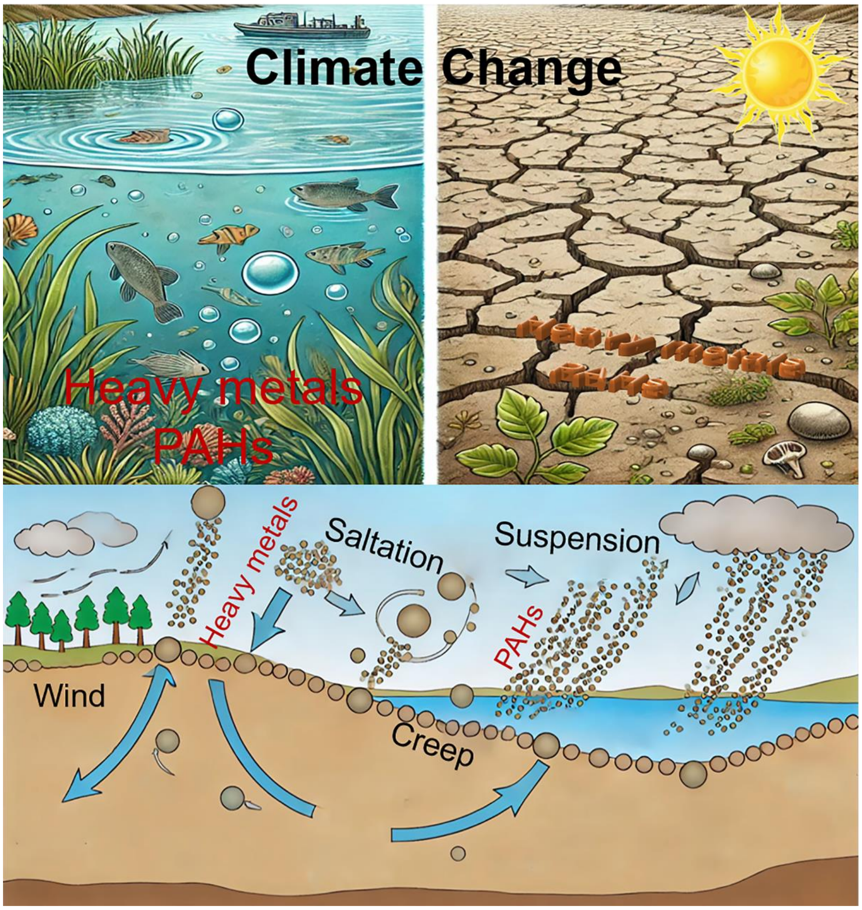
Graphical Abstract



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Short Summary

Numerous lakes are shrinking due to climate change and human activities, releasing pollutants from dried lakebeds as dust aerosols. The health risks remain unclear. Recently, Poyang and Dongting Lakes faced record droughts, exposing 99 % and 88 % of their areas. We show lakebed dust can raise PM10 to 637.5 $\mu g/m^3$ and exceed non-carcinogenic (HQ=4.13) and Cr carcinogenic ($\sim 2.10\times10^{-6}$) risk thresholds, posing growing health threats.

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1 Introduction

Global lakes possess a total volume of 181.9 × 10^3 km^3 and a total surface area of 2.67 × 10^6 km^2, representing 1.8 % of the Earth's terrestrial land area (Messager et al., 2016). Lakes serve as vital economic resources and are crucial for regional ecology, influencing the hydrological cycle and flood control (Williamson et al., 2009). Recently, many lakes worldwide have undergone rapid changes due to climate change and human activities (Beeton, 2002; Wurtsbaugh et al., 2017). For example, over the past 40 years, Lake Chad once the sixth largest lake in the world and located in the central Sahel sector at the southern edge of the Sahara, has become a symbol of the current global climate change in the region, having lost over 90% of its area due to persistent droughts (Gao et al., 2011). Agricultural water development in the Aral Sea watershed has led to a 74 % reduction in area and a 90 % decrease in volume (Micklin, 2007). Similarly, Lake Urmia, the world's second-largest hypersaline lake, plays a vital ecological and socio-economic role in northwest Iran, but has significantly shrunk by nearly 45 % in area and 85 % in volume in recent decades due to reduced inflows (Sima et al., 2021).

Exposed lakebeds serve as significant sources of dust in various regions. Owens Lake in California, which supplied drinking water to Los Angeles since 1913 and originally covered 280 km^2, was completely drained by 1926 (Reheis, 1997) and has since become the primary dust source in the continental U.S.(Gill and Gillette, 1991). Likewise, the Aral Sea's surface area was reduced by ~ 50 % between 1960 and 1992 due to water diversion for irrigation, leading to frequent dust storms from its exposed seabed (~27,000 km^2) (Micklin, 1988). In Australia, Lake Eyre contributes up to 3 % to the southeastern dust plume despite its considerable size (Farebrother et al., 2017). Similarly, Lake Urmia's exposed lakebed has become a new dust source, increasing aerosol optical depth, particularly in nearby regions (Hamzehpour et al., 2024; Alizade Govarchin Ghale et al., 2021). The evolution of lakebeds in the Chinese Loess Plateau has contributed to enhanced soluble-salt-bearing dust (Sun et al., 2023). In northern China, dry lakebeds, sandy grasslands, abandoned farmland, and mobile dunes significantly contribute to dust storms, with dry lakebeds containing high levels of fine particles (Yang et al., 2008). Notably, saline dust from these dried lakebeds contains high

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concentrations of sulfates and chlorides, leading to air pollution, soil salinization, vegetation degradation, and accelerated snow/ice melt (Liu et al., 2011). Recently, a study do find the Great Salt Lake, drying due to climate change, has exposed lakebed dust with high oxidative potential and elevated metal concentrations compared to nearby regions (Attah et al., 2024). These exposed dry lakebeds, driven by wind, have become source areas of dust emission (Tegen et al., 2002).

Poyang Lake, located in Jiangxi Province, is the largest freshwater lake in China. The watershed area of Poyang Lake covers 162,000 km^2 (Shankman et al., 2006). It is seasonal, exhibiting significant fluctuations in surface area (Guo et al., 2008). From April to September each year, during the wet season, the surface area of the lake can expand to over 4000 km^2. In contrast, during the relatively dry months from October to March, the lake's surface elevation drops by more than 10 m, causing the lake area to shrink dramatically to less than 1000 km^2 (Zhang et al., 2021). Dongting Lake ranks as the second-largest freshwater lake in China. During the flood season, substantial amounts of water from the Yangtze River pour into the lake, causing its area, which is usually under 500 km^2, to swell to 2,500 km^2. Conversely, in the dry season, from October to April, the lake discharges more water than it receives, leading to a drop in water level, and a significant portion of the lake area turns into dry land (Huang et al., 2012). Previous studies have shown that during extreme drought conditions, especially in 2022, the surface area of Poyang Lake and Dongting Lake drastically decreased to 322 km^2 and 311 km^2, respectively, with their water levels reaching their lowest in decades (Xu et al., 2024; Xia et al., 2024; Peng et al., 2024; Xue et al., 2023; Chen et al., 2023), exposing large portions of lakebed sediment. These exposed sediments are significant because aquatic environments tend to retain contaminants in sediments over extended periods. Sediments act as reservoirs for pollutants, such as polycyclic aromatic hydrocarbons (PAHs) and other organic pollutants, which strongly adhere to sediments due to their high hydrophobicity and resistance to degradation (He et al., 2014; Warren et al., 2003). Additionally, sediments reflect the historical accumulation of heavy metals from anthropogenic sources, and the burden of heavy metals in lake sediments is now increasing rather than declining (Yuan et al., 2011a). Furthermore, Poyang Lake and Dongting Lake have been reported to be contaminated by pollutants such as persistent organic pollutants (POPs) and heavy metals to a certain extent (Meng et al., 2019; Xu et al., 2018; He et al., 2018; Zhi et al., 2015; Li et al., 2013; Lu et al., 2012; Yuan et al., 2011b).

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Extreme droughts in East Asia, particularly affecting Poyang Lake and Dongting Lake, have become increasingly frequent and severe, as demonstrated by the unprecedented droughts of 2022. These droughts have severely impacted local hydrology and regional climate, raising concerns about the potential release of pollutants through dust aerosol generation from exposed lakebeds. Previous studies have primarily focused on salt lakes, investigating the inorganic ions and associated health risks related to heavy metals in lakebed soils and settled dust (Hosseinpoor et al., 2024; Grineski et al., 2024; Zucca et al., 2021; Ghale et al., 2021; Johnston et al., 2019). However, there remains uncertainty about whether the lakebed dust from freshwater lakes, such as Poyang and Dongting, contains sufficient toxic materials to exceed atmospheric thresholds and pose significant health risks. While exposed lakebeds are likely significant sources of dust aerosols, systematic studies on the health impacts of toxic substances, including polycyclic aromatic hydrocarbons (PAHs) and heavy metals, from freshwater lakes during extreme drought events remain sparse. To address this gap, this study utilizes field sampling, laboratory simulations, and model validation to analyze the composition of lakebed dust and its contribution to atmospheric particulate matter concentrations around these lakes. The analysis specifically focuses on the content of toxic materials (e.g., PAHs and heavy metals), the mass concentration of lakebed dust aerosols in the air. This comprehensive approach aims to quantify the potential health risks posed by lakebed dust emissions during the extreme drought conditions experienced in 2022 and in a possible long-term scenario, providing crucial insights into the environmental and public health implications of increasing dust emissions under a changing climate.

2 Materials and methods

2.1 Lakebed soil sampling

In October 2022, a total of 9 lakebed soil samples were collected during the dry season from the top 5 cm of the lakebed soil profile at Poyang Lake and Dongting Lake. The sampling sites around Poyang Lake include: Areas within one kilometer of the lake's surface, which are typically not covered by water year-round (A1 and A2); zones where conditions alternate between dry and wet in typical years (B1 and B2); and regions that are usually submerged but are currently exposed due to extreme drought (C1 and C2). Correspondingly, the three sampling points at Dongting Lake are designated as A3, B3, and C3, mirroring those at Poyang Lake. Details of the sample locations can be found in SI Table S1. The collected lakebed soils were air-dried, sifted through a 2 mm nylon sieve to remove debris, thoroughly mixed, and stored at 4 °C in the dark before analysis.

2.2 Laboratory dust aerosol generation and particle sample collection

We employed the GAMEL laboratory dust generator, as described by Lafon et al. (2014), to generate dust aerosols from lakebed soil samples. The GAMEL generator effectively simulates the natural sandblasting process and produce dust aerosols with realistic size distributions and chemical compositions. While wind tunnels are proficient in creating realistic dust aerosol conditions, they present several challenges, including the requirement for large volumes of parent soils and the significant expense of minimizing interference from ambient aerosols (Alfaro et al., 1997; Lafon et al., 2006; Alfaro, 2008). In our experiments using the GAMEL generator, we introduced 10 g of each lakebed soil sample into a PTFE flask. The flask was then vibrated to simulate the sandblasting process and produce dust aerosols. We maintained a steady flow of particle-free air through the setup (Gao et al., 2023a; Gao et al., 2023b). According to (Lafon et al., 2014), the shaker was optimally set to operate at 500 cycles/min, with an airflow rate of 8 L/min, controlled by a mass flow controller (MFC, Sevenstar, Beijing Sevenstar Flow Co., LTD). The aerosol stream was directed through a cyclone, with particles being captured on a 47 mm PVC film situated in a metal frame filter holder (Pall Gelman, Port Washington, NY, USA). Dust-$PM_{2.5}$ and dust-PM_{10} samples were collected by using an 8 LPM cyclone, or without it, respectively. The duration of the operation was set to 1 min. All the dust aerosol mass collected is shown in Table S2. The instrument setup is shown in Fig. S1.

2.3 Samples extraction and PAHs analysis

Approximately 0.5 g of lakebed soil samples containing generated dust-$PM_{2.5}$ and dust-PM_{10} were spiked with 10 ng of hexamethylbenzene (HMB) as a recovery surrogate. These samples were then separately extracted via ultrasonication using 10 mL of dichloromethane, n-hexane, and acetone for

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30 min. Following extraction, all liquids were combined and filtered through a 0.22 µm nylon membrane. The clarified extract was then concentrated to dryness using a rotary evaporator (IKA, Germany). The dry residue was reconstituted in dichloromethane and spiked with five internal standards: naphthalene-d8 (NAP-D8), acenaphthene-d10 (ANA-D10), phenanthrene-d10 (PHE-D10), chrysene-d12 (CHR-D12), and perylene-d12 (PERY-D12), each dissolved into 0.5 mL.

An analytical method for characterizing PAHs was developed utilizing a gas chromatograph-mass spectrometer (GC-MS; Thermo Fisher Trace ISQ 7900). This setup included a Thermo Fisher Trace 1300 GC equipped with a TG-5SILMS capillary column (30 m × 0.25 mm I.D., 0.25 µm film thickness). Quantification of the compounds was performed in selective ion monitoring mode (SIM) to improve sensitivity. For data processing, Chromeleon quantitative analysis software (version 7.2.9) was employed. The comprehensive procedures for the identification and quantification of PAHs are detailed in the SI.

2.4 Quality assurance and quality control

To mitigate potential contamination of target analytes by background levels, a procedural blank was prepared alongside each batch of samples using an identical protocol. Each batch included three replicates. If PAHs were detected in the procedural blank, the concentrations in the actual samples were adjusted by subtracting the amounts found in the blank. The recovery rates for spiked PAH components ranged from 70 % to 120 %.

2.5 Heavy metals analysis

The heavy metal content of Fe, Mg, Ti, Mn, Ba, V, Zn, Cr, Ni, Cu, As and Pb was conducted using X-ray fluorescence spectroscopy (XRF, S8 Tiger, Germany). The specific procedure involves passing the collected samples through a 200-mesh sieve, pressing them into pellets with boric acid (Analytical Reagent) as a backing material, and then gaining the elements of interest. Detailed information can be found in this study (Oyedotun, 2018).

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2.6 Enrichment Factors

Enrichment factors (EFs) were calculated to investigate the compositional variations between parent lakebed soil and the resulting dust aerosol. EFs are characterized as the ratio of the concentration of an individual PAH and heavy metal in the lakebed soil to that in the generated dust.

$$EF = \frac{C_{dust}}{C_{soil}} \quad (1)$$

Where C_{dust} is an individual PAH and the heavy metal concentration in dust-PM, and C_{soil} is the concentration of an individual PAH and heavy metal in the parent lakebed soil. EFs can indicate the redistribution of a compound from the lakebed soil to the dust aerosol. By analyzing the measured EFs along with PAH concentrations and heavy metals in lakebed soils and dust aerosol concentrations, the contribution of lakebed soil pollutants to atmospheric aerosols can be evaluated (Gao et al., 2023a; Gao et al., 2023b).

2.7 The Community Multiscale Air Quality (CMAQ) model configuration and data

The CMAQ model was used to simulate the total PM concentration and the contribution of exposed lakebeds dust of Poyang Lake and Dongting Lake, as well as their surrounding areas, during the extreme drought in October 2022. The modified source-oriented CMAQ model v5.0.2, incorporating an expanded Statewide Air Pollution Research Center (SAPRC-99) photochemical mechanism, was employed to simulate atmospheric $PM_{2.5}$ and PM_{10} levels, as well as the contribution of windblown dust from the exposed lakebed to $PM_{2.5}$ and PM_{10} concentrations. The CMAQ model was run from September 26 to October 30, 2022, with the first three days designated as a spin-up period to minimize the impact of initial conditions (Gao et al., 2023b; Ying et al., 2018).

Nested domains of 36 km and 12 km resolution were established in Central China, including the Dongting and Poyang Lakes, as shown in Fig. S2. The 36 km domain (127 × 197 grids) covers China and surrounding countries, while the 12 km domain (118 × 118 grids) covers Central China. Anthropogenic emissions are based on the Multi-resolution Emission Inventory for China (MEIC, v1.4, 0.25° × 0.25°, http://www.meicmodel.org, last access: July 26, 2024). Biogenic emissions were

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generated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1 (Guenther et al., 2012). Meteorological inputs were calculated using the Weather Research and Forecasting (WRF) model version 4.1.2, driven by FNL (Final) Operational Global Analysis data from the National Center for Environmental Prediction (NCEP) (https://rda.ucar.edu/datasets/ds083.2, last access: July 26, 2024).

For model validation, meteorological observations from the National Climate Data Center (NCDC; https://www.ncdc.noaa.gov/, last viewed on July 26, 2024) were used to validate the simulated WRF output. Hourly observations of air pollutants ($PM_{2.5}$ and PM_{10}) were collected from the China National Environmental Monitoring Centre (CNEMC; http://www.cnemc.cn/, last accessed on July 26, 2024) to assess the CMAQ simulation results.

2.8 Model scenario setting

We simulated two scenarios based on different land use types to identify the effects of the exposed lakebeds on dust PM due to extreme drought in 2022 (Table S3). CASE_unexposed represents an ordinary scenario without considering lake drying. It is driven by land use data from Moderate Resolution Imaging Spectroradiometer (MODIS) Land Cover Type product (MCD12Q1) (Sulla-Menashe and Friedl, 2018), which provides stable lake areas and represents the typical annual condition of the lake. CASE_exposed represents a real scenario considering the exposed lakebeds caused by the extreme drought, driven by modified MODIS land use data. In our study, Sentinel-2A Multispectral Instrument data were used to adjust the land use type. The differences between the two cases were regarded as the contribution of exposed lakebeds dust. We performed a median synthesis of the inversion results for the green band (B3) and near-infrared band (BB) of the sentinel-2 satellite from September to November, marking grid cells with an NDWI value greater than 0 as water bodies (Mcfeeters, 1996):

$$NDWI = \frac{(Green + NIR)}{(Green - NIR)} \quad (2)$$

where $Green$ is a band that encompasses reflected green light and NIR represents reflected near−infrared radiation. By comparing the water bodies detected by sentinel-2 and MODIS, we modified grid cells detected by MODIS as lakes but detected by sentinel-2 as non-water bodies to dust sources as exposed areas of lakes due to extreme drought. Other areas remained unchanged as the MODIS land use type (Fig. S2).

2.9 PAHs and heavy metal toxic equivalency in lakebed dust-PM

The Benzo[a]pyrene toxic equivalency factors (TEF) for individual species are provided in Table S4 obtained by these studies (Delistraty, 1997; Nisbet and Lagoy, 1992). The toxic equivalent concentration of PAHs ($C_{BaP_{eq}}$) is calculated using the following equation (Wu et al., 2022):

$$C_{BaP_{eq}} = \Sigma(C_i \times TEF_i) \quad (3)$$

where C_i represents the concentration of individual PAH species (ng/g), and TEF_i is the Benzo[a]pyrene toxic equivalency factor for each PAH species. BaP_{eq} indicates the toxic equivalent concentration of PAHs (ng/g). The BaP equivalent concentrations were calculated by multiplying each individual PAH concentration by its respective TEF.

The Cr toxic equivalency factors (TEF) for every heavy metal are provided in Table S5 obtained by the U.S.EPA. The toxic equivalent concentration of heavy metals ($C_{Cr_{eq}}$) is calculated using the following equation (Wu et al., 2022):

$$C_{Cr_{eq}} = \Sigma(C_i \times TEF_i) \quad (4)$$

2.10 Health risk assessment of modeling transport of lakebed dust aerosol

The non-carcinogenic and carcinogenic risks of PAHs and heavy metals in the modeled transport of lakebed dust (PM_{10}) for residents in the vicinity of lakes and surrounding cities were assessed through inhalation exposure. We derived estimates of short-term non-carcinogenic hazard for dust-PM using the following equation (Means, 1989):

$$HQ_{ST} = \frac{EC_{ST}}{AV} \quad (5)$$

Where HQ_{ST} represents the short-term hazard quotient (HQ) for an individual contaminant, EC_{ST} is the exposure concentration, equivalent to the contaminant concentration for short-term exposures

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(Epa, 2004), and AV is the corresponding acute dose-response value for that contaminant (from CalEPA OEHHA) (Monserrat, 2016). In this study, we assumed the maximum daily concentration of each contaminant as its short-term exposure concentration. A non-carcinogenic risk is indicated when HQ values exceed 1. It is acceptable to combine the individual HQs to calculate a multi-pollutant hazard index (HI) using the following formula:

$$HI_{ST} = \sum_{i} HQ_{STi} \tag{6}$$

Where HI_{ST} is short-term hazard index, and HQ_{STi} is short-term hazard quotient for the i^{th} contaminant.

Similar hazard indices of non-carcinogenic risk are used. The exposure concentration for chronic exposures (EC_C) was calculated as (Ren et al., 2021a; Epa, 2004; Means, 1989):

$$EC_C = C \times \frac{ET \times EF \times ED}{AT} \tag{7}$$

Where C is the concentration of individual contaminant concentration, ET is the exposure time, EF is the exposure frequency, ED is the exposure duration, and AT is the averaging time (for non-carcinogens, AT =ED ×365 d; for carcinogens, AT =70×365=25500 d). Specifically, given that chronic non-carcinogenic and carcinogenic risks generally require extended assessment over a typical 70-year period, we set parameters to estimate human exposure to lakebed dust. Based on guidelines from the Beijing Municipal Bureau (China) and supporting literature, resident outdoor exposure time was set as 8 hours per day over a 30-year period (Ren et al., 2021a; Bureau, 2009). The drought in Poyang Lake at Xingzi Station was identified below 7 m, representing an area of 600 km² (~4 % of the lake's total area) (Fig. S3a). Over a 15-year period (2000-2014), the average drought duration was estimated at 80 days per year (Table S6) (Qi et al., 2019). For Dongting Lake, the drought threshold was set at 700 km² (~28 % of lake's area) (Fig. S3b), with an average drought duration of 140 days per year observed over a 10-year period (2000-2009) (Fig. S4) (Huang et al., 2012). Detailed parameter information can be found in SI. The chronic non-carcinogenic hazard for dust-PM was calculated as:

$$HQ_C = \frac{EC_C}{RfC} \tag{8}$$

Where HQ_C is the chronic HQ for an individual contaminant, and RfC is the inhalation

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reference concentration for that contaminant (Ren et al., 2021a; Us Epa, 2015). There is a probability of non-carcinogenic risk with HQ values above 1. Chronic hazard index (HI_C) was calculated as:

$$HI_C = \sum_i HQ_{Ci} \quad (9)$$

The carcinogenic risk (CR) of each contaminant is calculated as (Ren et al., 2021a; Means, 1989):

$$CR = EC_C \times IUR \quad (10)$$

Where IUR is the inhalation unit risk for each contaminant. For regulatory purpose, a CR value lower than 10^{-6} is adopted negligible, and above 10^{-4} is not accepted by most international regulatory agencies (Gao et al., 2023b; Ren et al., 2021a). In this study, the World Health Organization (WHO) has provided a IUR_{BaP} estimate of 8.7×10^{-5} per ng/m³, derived from epidemiological studies on coke-oven workers (Organization, 2000).The inhalation unit risks of the selected metals for carcinogenic risk are 8.4×10^{-2}, 2.4×10^{-4}, 4.3×10^{-3}, 9.0×10^{-3}, 1.8×10^{-3} and 8×10^{-5} ((μg/m³)$^{-1}$) for Cr, Ni, As, Co, Cd and Pb, respectively (Ren et al., 2021a). The detailed information is found in SI Table S7.

3 Results and discussion

To evaluate the health effects of lakebed dust generated from Poyang Lake and Dongting Lake, three critical factors need to study: (1) the content of toxic materials, including PAHs and heavy metals, in the lakebed dust aerosols; (2) the mass concentration of lakebed dust aerosols in the air surrounding the two lakes; and (3) the duration of human exposure to air containing lakebed dust. We will analyze these factors in the following section.

3.1 PAH contents in the lakebed dust aerosols

To examine the PAH contents in the exposed lakebed dust from Poyang Lake and Dongting Lake, in October 2022, we collected a total of nine lakebed soil samples during the extreme drought conditions from the upper 5 cm of the lakebed soil layer at both Poyang Lake and Dongting Lake, including areas beyond the lake boundary, along the lakeshore, and within the lakebed. Sampling locations included regions typically not covered by water year-round (A1, A2 and A3), transitional zones that fluctuate between submerged and dry states (B1, B2 and B3), and areas usually submerged but exposed by drought (C1, C2 and C3) (Fig. 1). To study the chemical composition of lakebed dust aerosols, we opted for laboratory simulation experiments in a controlled environment to avoid contamination from other types of aerosols. Using the GAMEL (Générateur d'Aérosol Minéral En Laboratoire) laboratory dust generator, we replicated the natural sandblasting process, producing dust aerosols with realistic particle size distributions and chemical compositions (Lafon et al., 2014). In GAMEL's dust production system, 10 g of each soil sample was placed in a PTFE flask, which was agitated by a shaker to simulate the sandblasting process and produce dust aerosols. A detailed description is provided in the Materials and methods section.

The distribution of concentrations of PAHs in lakebed soil, dust-PM_{10}, and dust-$PM_{2.5}$ samples from Poyang Lake and Dongting Lake are shown in Fig. 1. In the Poyang Lake, the mean concentration of Σ_{16} PAHs was in the range of 122.7-2015.3 ng/g, 1331.8-3385.4 ng/g, and 1780.3-6683.9 ng/g in lakebed soil, dust-PM_{10} and dust-$PM_{2.5}$, respectively (Fig. 1a and Fig. 1b). In Dongting Lake, the PAHs concentration was between 1039.7-3357.9 ng/g, 5380.6-19748.5 ng/g and 8416.5-48473.0 ng/g in lakebed soil, dust-PM_{10} and dust-$PM_{2.5}$, respectively (Fig. 1c). It was found that the PAHs concentrations in the dry lakebed soils of Poyang Lake and Dongting Lake generally (A1, A2 and A3) are consistent to other studies (Zhang et al., 2021; Meng et al., 2019). Higher concentrations were generally observed in Dongting samples, and lower concentrations were in Poyang samples. This discrepancy may be due to the locations of the sampling points. The samples from Poyang Lake were collected from a nature reserve area, whereas the samples from Dongting Lake were collected from areas near human activity (0.25 km to a kindergarten and a judicial office).

Fig. 1d shows the precise locations of the sampling sites in Poyang and Dongting Lakes. Generally, among all sampling locations, the highest concentrations of PAHs were found in sites B1, B2, and B3, followed by sites C1, C2, and C3, with the lowest concentrations in sites A1, A2, and A3, indicating that areas along the lake, situated between the water and dry land, are more likely to absorb pollutants and become sinks for PAHs (Fig.1, S5 and S6). Lakeside areas are indeed prone to accumulating pollutants due to a combination of runoff, sedimentation, human activities, and hydrological changes (Zhang et al., 2010; O'sullivan and Reynolds, 2004). Fluctuations in water

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levels caused by weather, such as droughts or seasonal variations, can expose previously submerged sediments and release accumulated pollutants back into the environment via dust production.

Additionally, we observed that PAH emissions from the lakebed demonstrate enrichment effects. Enrichment factors (EFs) are calculated as the ratio of each toxic substance's concentration in lakebed soil to that in the generated dust aerosol, providing a basis for analyzing compositional shifts between the original lakebed soil and the dust aerosol. It is demonstrated that PAHs are highly enriched in fine dust aerosols ($PM_{2.5}$) (Fig. S7 and Fig. S8). Specifically, their absolute concentrations are significantly higher in fine dust particles compared to the parent lakebed soil (Fig. 1 a-c, Fig. S5 and Fig. S6). The total EFs ranged from ~60.4 to ~818.7 in dust-PM_{10} and from ~164.5 to ~1122.4 in dust-$PM_{2.5}$ (Fig. S7). Moreover, the EFs of total PAHs in dust-$PM_{2.5}$ were higher than those in dust-PM_{10}, especially NAP, ANY, and FLU showing consistently higher EFs in dust-$PM_{2.5}$ across most sites (Fig. S8), indicating that PAHs are likely to be enriched in finer particles during dust aerosol generation. This finding is consistent with previous studies showing that PAHs are mostly enriched in fine inhalable particles, with a high concentration in atmospheric particles (Gao et al., 2023a; Van Vaeck and Van Cauwenberghe, 1978).

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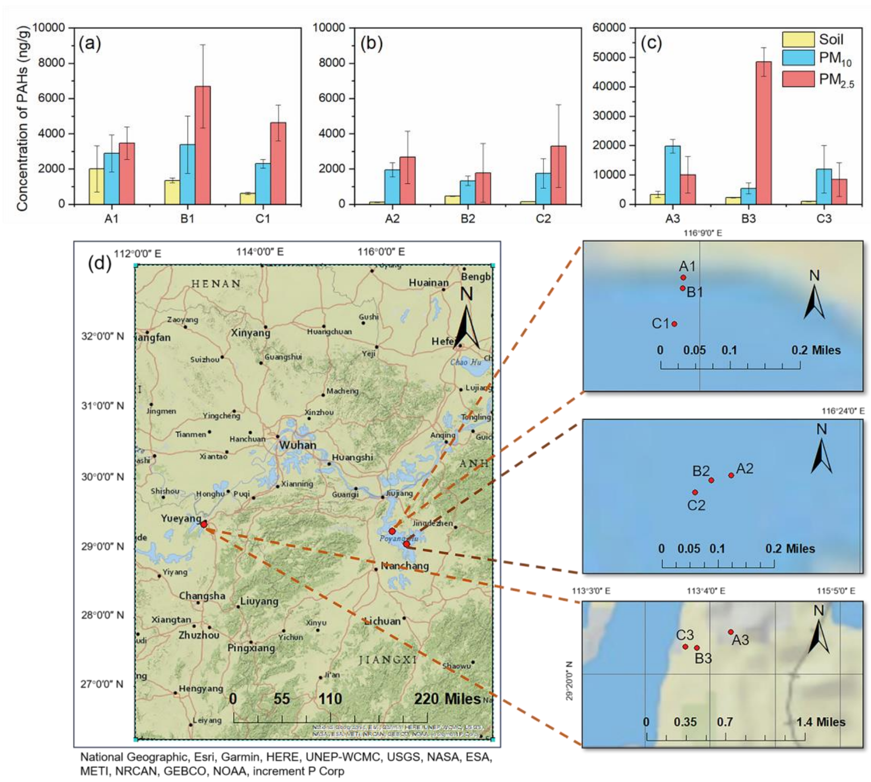

Figure 1. Comparison of total concentration of PAHs in the parent lakebed soils and dust aerosols. (a), (b), and (c) represent the concentration of PAHs. (d) indicates the sampling locations of Poyang Lake and Dongting Lake. A1, A2, and A3 are regions typically dry and exposed year-round. B1, B2, and B3 are transitional zones that fluctuate between submerged and dry states. C1, C2, and C3 are areas usually underwater but sometimes exposed due to extreme drought. A1, B1, and C1, along with A2, B2, and C2, are sampling sites from Poyang Lake, while A3, B3, and C3 are sampling sites from Dongting Lake. The base map in panel (d) is sourced from National Geographic, Esri, Garmin, HERE, UNEP-WCMC, USGS, NASA, ESA, METI, NRCAN, GEBCO, NOAA, and increment P Corp, as provided within the ArcGIS software.

3.2 Heavy metal contents in the lakebed dust

Apart from PAHs, heavy metals were analyzed by XRF to examine their spatial distribution during climate changes. Fig. 2 and Fig. S9 illustrate the comparative concentration of heavy metals in lakebed soil and dust-$PM_{2.5}$ samples across different sampling sites. In the Poyang Lake, Cr (ranging from ~0.006 % to ~0.0109 %), Ni (ranging from ~0.004 % to ~0.005 %), Cu (ranging from ~0.003 % to ~0.0071 %), Zn (ranging from ~0.0074 % to ~0.0184 %), As (ranging from ~0 % to ~0.003 %) and Pb (ranging from ~0.004 % to ~0.005 %) were detected. Xie et al. reported that mean values of Cr, Ni, Cu, Zn, As and Pb in Poyang Lake are 81.39, 30.47, 35.17, 104.17, 11.34 and 32.63 mg/kg, respectively (Xie et al., 2016), which are consistent with our findings but significantly higher than those in natural dust source region where Cr, Ni, Cu, Zn, As and Pb are from 13.4 to 51.2, 5.5 to 26.5, 5.1 to 27.0, 17.8 to 72.2, 2.6 to 11.6, and 4.9 to 41.1 mg/kg, respectively (Gao et al., 2023b).

Furthermore, the Fe, Mg, Ti, Mn, Ba, V, Zn, Cr, Ni, Cu, As and Pb in dust-$PM_{2.5}$ are higher than in the lakebed soil during the process of dust generation. Notably, Cr from B2 were ~0.0194 % and ~0.0184 % in dust-$PM_{2.5}$ and lakebed soil, respectively. Most dust-$PM_{2.5}$ likely originates from small colloids in the lakebed soil. These lakebed soil colloids typically carry large amounts of negative charges, aiding in the adsorption of many cations, including various heavy metal ions (Brady et al., 2008; Gao et al., 2023b). Consequently, heavy metals are enriched in small soil aggregates. During the sandblasting process, the smaller soil grains, which have higher heavy metal concentrations, are more likely to be ejected and form dust aerosols.

Similar to the spatial distribution of individual PAHs in the lake regions under drought conditions, heavy metals also exhibit their highest concentrations along the lakeshore, as shown in Fig. 2 and Fig. S9. Therefore, under some climate change conditions, extensive droughts expose lakebeds (Attah et al., 2024; Sun et al., 2023; Gao et al., 2011), resulting in the release of accumulated heavy metals from lakeshore areas into the environment, thereby posing risks to regional ecosystems and human health.

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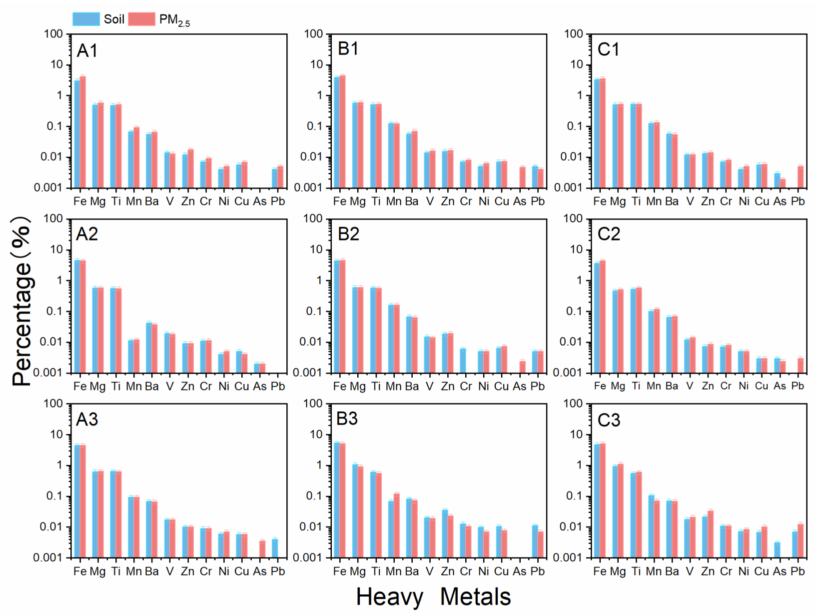

Figure 2. Comparison of heavy metal percentage between lakebed soil and generative dust-$PM_{2.5}$. A1, B1, C1, A2, B2 and C2 were obtained in Poyang Lake, and A3, B3 and C3 were obtained in Dong ting Lake. A1, A2, and A3 are regions typically dry and exposed year-round. B1, B2, and B3 are transitional zones that fluctuate between submerged and dry states. C1, C2, and C3 are areas usually underwater but sometimes exposed due to extreme drought.

3.3 Mass concentration of lakebed dust aerosols in the surrounding lake area

With the chemical compositions and concentrations of lakebed dust aerosols identified, the next step was to estimate the particulate matter (PM) contribution from exposed lakebed dust and calculate the PAH and heavy metal concentrations in the atmosphere around Poyang and Dongting Lakes based on dust composition profiles. Therefore, we modeled the mass concentration from exposed lakebed dust in Poyang Lake and Dongting Lake and their surroundings area during the extreme drought in October 2022, and validated the results using the observation data of local meteorology and air pollution.

To obtain the mass concentration of lakebed dust aerosols in the surrounding lake area, we designed two scenarios: CASE_unexposed excluded the lakebed dust exposure due to extreme drought, while

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CASE_exposed included it (Table S1). The differences between the two cases were considered the contribution of exposed lakebed dust. The validation results showed that CASE_exposed outperformed CASE_unexposed in terms of PM concentration after accounting for lakebed dust, particularly for PM_{10} (Table S8 and Table S9). For example, the model's mean fractional bias (MFB) and mean fractional error (MFE) for Nanchang of Jiangxi Province in China, a city near Poyang Lake, decreased from -40 % and 56 % in CASE_unexposed to -1 % and 39 % in CASE_exposed, respectively (Fig. S10). This indicates that the prediction met model performance goals (MFE ≤ ±30 % and MFE ≤ 50 %) after considering lakebed dust (Boylan and Russell, 2006), indicating its non-negligible impact in areas near the lakes.

The modelling results show that the monthly average PM_{10} concentrations of lakebed dust was 10.2 and 34.5 μg/m^3 in Dongting and Poyang Lakes, respectively, while the maximum daily levels could reach 119.9 and 420.1 μg/m^3 during the entire October in 2022 (Fig.3a and b). Considering transport effects, the maximum daily concentrations of lakebed dust PM_{10} could reach 17.8 ~ 302.3 and 12.2 ~ 637.5 μg/m^3 within 20 km of Dongting and Poyang Lakes, respectively. Furthermore, due to the influences of northeasterly winds, the impact of lakebed dust extended over 60 km, causing maximum daily concentrations exceeding 150 μg/m^3 in areas to the south of Dongting Lake and central Jiangxi Province.

We estimated the spatial distribution of PAHs and heavy metals using modelled PM concentrations based on the aforementioned dust composition profiles (Fig. 3c-f and Fig. S11-S13). Due to differences in composition, Dongting Lake dust has a higher fraction of PAHs and heavy metals than Poyang Lake dust for an equivalent mass of lakebed dust (Fig. S5 and Fig. S9). As a result, although Poyang Lake had a higher lakebed dust concentration, PAHs and heavy metals concentrations from lakebed dust displayed distributions around both lakes. For example, in Dongting and Poyang Lakes, the monthly average BaP concentrations in Dongting and Poyang Lakes were 0.010 and 0.014 ng/m^3, respectively, with maximum daily concentrations reaching 0.122 and 0.167 ng/m^3. For Cr, the monthly average concentrations were 0.001 and 0.003 μg/m^3 in Dongting and Poyang Lakes, while the maximum daily concentrations reached 0.013 and 0.031 μg/m^3, respectively.

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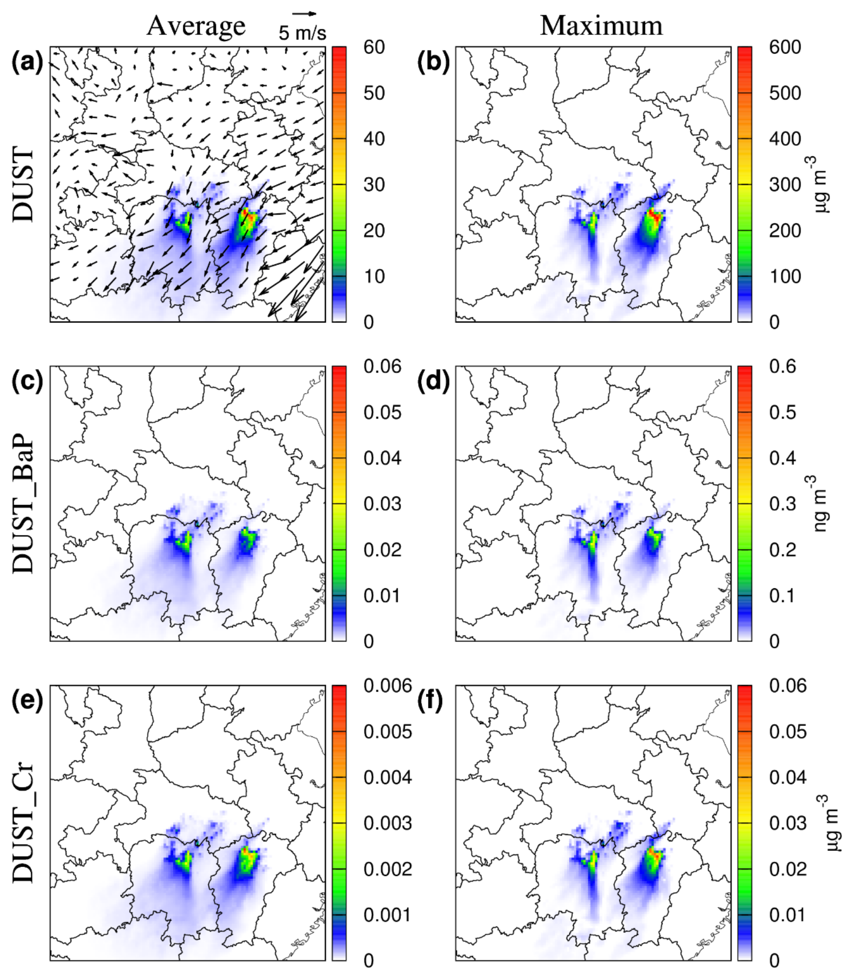

Figure 3. Spatial distribution of predicted monthly average and maximum daily concentrations of lakebed dust PM_{10}, along with its BaP and Cr concentrations. The left panels display the monthly average concentrations of (a) lakebed dust PM_{10}, (c) BaP in lakebed dust PM_{10}, and (e) Cr in lakebed dust PM_{10}. The right panels present the maximum daily concentrations of (b) lakebed dust PM_{10}, (d) BaP in lakebed dust PM_{10}, and (f) Cr in lakebed dust PM_{10}. The monthly average wind is overlaid in (A).

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3.4 Assessing health risks from toxic substances of lakebed dust emissions

To assess whether the lakebed dust aerosols pose a significant health risk to people living the lake shore areas, we analyze their toxicities, which are expressed using BaP_{eq} (Benzo[a]pyrene equivalents) for PAHs (Table S4) and Cr_{eq} (Cr equivalents) for heavy metals (Table S5), normalizing their toxic potential (Delistraty, 1997; Nisbet and Lagoy, 1992; Epa). This approach enables standardized comparisons of different compounds' toxicities under the same aerosol mass, helping evaluate their relative contributions to overall toxicity, as PAHs and heavy metals carry distinct health risks.

To provide a clearer comparison of BaP_{eq} and Cr_{eq} concentrations in dust aerosols and lakebed soils from Poyang and Dongting Lakes, Fig. S14 and Fig. S15present the BaP_{eq} concentrations of the total 16 PAHs and Cr_{eq} concentrations for heavy metals (ng/g) in lakebed dust aerosols. The geographical distributions of BaP_{eq} concentrations in lakebed soil, dust-$PM_{2.5}$ and dust-PM_{10} are shown in Fig. S14. Overall, the BaP_{eq} concentrations in Dongting Lake regions (A3, B3 and C3) were higher than those in the Poyang Lake regions (A1, B1, C1, A2, B2 and C2). Interestingly, in the Poyang Lake region, BaP_{eq} per unit mass of dust-$PM_{2.5}$ emitted from C1 was the highest (1470.1 ±115.3 ng/g) compared to A1 (296.0±131.9 ng/g) and B1 (865.3±450.0 ng/g). Additionally, A2, B2, and C2 showed a similar pattern, with the highest BaP_{eq} concentrations in C2 (1575.6±1058.9 ng/g). Furthermore, compared to this study (Wu et al., 2022), the BaP_{eq} per unit mass of dust-$PM_{2.5}$ from C1, C2, A3, and B3 were higher than those from coal-fired power plants (CFPPs) (1410±880 ng/g). The distribution of Cr_{eq} emissions from the two lake regions is displayed in Fig. S15. Compared with the results of Wu et al., Cr_{eq} emitted from dust-$PM_{2.5}$ were generally 2-3 times higher than Cr_{eq} from household coal burning (Wu et al., 2022).

To assess the inhalation risks posed by the transmission of lakebed dust emissions, we utilized the modeled lakebed dust PM_{10} and their contents of PAHs and heavy metals modeling results described above. Fig. 4 presents the non-carcinogenic and carcinogenic health risks associated with atmospheric PAHs and heavy metals in the vicinity of the lakes and several cities in Jiangxi and Hunan Provinces. It is important to note that we assumed the maximum daily concentration of each

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contaminant as its short-term exposure concentration for the non-carcinogenic risk assessment. Since chronic carcinogenic risks typically necessitate long-term evaluation, often assessed over a 70-year period. To estimate the duration of human exposure to air containing lakebed dust, we established health risk assessment parameters based on reports from the Beijing Municipal Bureau (China) and relevant literature. Outdoor exposure hours and exposure duration for residents was set to 8 hours per day and 30 years (Ren et al., 2021a; Bureau, 2009). The drought in Poyang Lake is defined as the water level at Xingzi Station was lower than 7 m, corresponding the lake surface area shrinking to 600 km² or ~4 % of the whole Poyang Lake's area (Table S6 and Fig. S3a). The average drought duration over a 15-year period (2000-2014) was 80 days per year (Qi et al., 2019). Similarly, for Dongting Lake, where the drought threshold is 700 km² (~28 % of Dongting's surface area) (Fig. S3b), the average drought duration over a 10-year period (2000-2009) was determined to be 140 days per year (Fig. S4) (Huang et al., 2012), as outlined in Table S7. Additionally, considering that atmospheric Cr (VI) and Cr (III) are typically present in a 1:6 ratio (Liu et al., 2018), Cr (VI) content was estimated 1/7 of total Cr for evaluating chronic non-carcinogenic and carcinogenic risks, while Cr (III) content was estimated 6/7 of total Cr to assess short-term non-carcinogenic risks. A Hazard Quotient (HQ) above 1 is defined as the threshold for non-carcinogenic risk, and individual HQs can be combined to calculate a cumulative hazard index (HI) for multiple pollutants (Ren et al., 2021b). For non-carcinogenic risk assessment, short-term exposure is assessed using the maximum daily concentrations of pollutants (As, Cr, Cu, Mn, Ni and V) from the modeled transport data (Fig. 4a), while chronic exposure is evaluated using the concentrations of pollutants (As, Ba, BaP_{eq}, Cr, Mn, Ni and V) derived from the modeled monthly average transport (Fig. 4b). In the areas around the lakes, short-term health risks, mainly driven by manganese (Mn) exposure, exceed the acceptable threshold, with HQ values of 1.67 near Dongting Lake and 4.13 near Poyang Lake. Additionally, the HI near Dongting Lake and Poyang Lake reached 2.55 and 5.69, respectively, while the average HI across Nanchang, a city near Poyang Lake, was also elevated at 1.65. By contrast, the calculated chronic risks remain below the EPA threshold (Means, 1989).

Moreover, we estimated the carcinogenic risk of BaP_{eq} and Cr in PM_{10} during lakebed dust aerosol transport (Fig. 4, c-d). The results show that the chronic carcinogenic risk associated with Cr, the areas surrounding the lakes exceed the acceptable threshold, with the highest values simulated at

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approximately 2.10×10^{-6} near Dongting Lake (28.896° N, 112.607° E) and 2.00×10^{-6} near Poyang Lake (29.256° N, 115.906° E), exceeding threshold limits (10^{-6}). However, all estimated carcinogenic risks of BaP_{eq}, with the highest value found in lake surroundings (~1.02×10^{-7}), remain below the established EPA threshold limits (Emergency and Response, 1989). These findings underscore the importance of addressing the health risks posed by dust aerosols originating from lakebed sources.

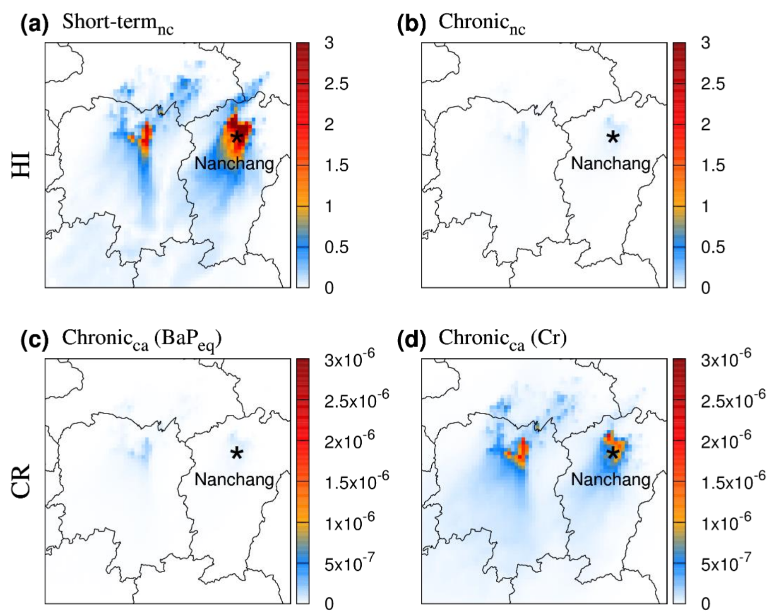

Figure 4. A comparison of short-term and long-term non-carcinogenic risks, along with the long-term carcinogenic risks associated with BaPeq and Cr, derived from modeled atmospheric PAHs and heavy metals in the vicinity of Poyang Lake, Dongting Lake, and surrounding cities, based on the dust-PM_{10} profile. Among these, (a) represents the short-term non-carcinogenic risk (nc: non-carcinogenic risk), (b) represents the chronic non-carcinogenic risk, (c) indicates the chronic carcinogenic risk of BaPeq, and (d) represents the chronic carcinogenic risk of Cr (ca: carcinogenic risk). An HQ value of 1 denotes the threshold for non-carcinogenic risk, suggesting potential health effects if exceeded, and individual HQs can be combined to calculate a cumulative HI for multiple pollutants. Similarly, a CR value of 1×10^{-6} denotes the carcinogenic risk threshold, beyond which chronic exposure may elevate the risk of cancer.

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4. Environmental implications and conclusions

Many of the world's lakes are shrinking at alarming rates due to the combined impacts of climate change and human activities, leading to widespread exposure of lakebeds as a significant dust source. In this study, we have used field sampling, laboratory simulations, and CMAQ modeling to demonstrate that the exposed lakebeds during extreme drought conditions can become a significant source of dust aerosols, with pollutants deposited in the lakebed re-entering the atmospheric cycle for Poyang and Dongting lakes, two of the arguably most important lakes in China.

Wind erosion produces dust aerosols by driving lakebed soil movements such as creeping, saltation, and suspension. Laboratory simulations confirm that pollutants become enriched in fine dust particles. Additionally, the concentrations are significantly elevated at the lakeshore and within the lake compared to the areas outside the lake. Hence, PAHs and heavy metals accumulated in the lakeshores and submerged regions of Poyang and Dongting Lakes are likely to be reintroduced into the atmosphere through wind-driven sandblasting processes.

The CMAQ simulations, when compared with meteorological and air quality observations, demonstrated that incorporating lakebed dust emissions significantly improved the alignment between observed and simulated results, particularly in predicting elevated PM_{10} levels (Fig. S10). However, uncertainties still exist in the simulated concentrations due to limitations of satellite-derived land use data and meteorological fields from Weather Research and Forecasting (WRF) simulations. These findings highlight that exposed lakebeds are substantial sources of dust aerosols, capable of reintroducing accumulated contaminants into the atmosphere and posing regional health risks. In this study, we quantified the health risks of the 16 priority PAHs and heavy metals identified by the EPA from lakebed dust. If emissions from other sources are considered, the inhalation risk from PAH transmission would exceed the threshold. Additionally, other persistent organic pollutants, such as halogenated compounds, were also detected in the lakebed sediments, though their health risks were not quantified in this analysis. This is particularly relevant for lake regions with high

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pollutant concentrations, especially in areas like Northeast China, which have historically experienced heavy industrial activity.

East Asia is one of the largest sources of dust aerosols globally (Song et al., 2019). These Asian dust aerosols travel over long distances and can be transported to regions such as eastern China, South Korea, Japan, and even across the Pacific Ocean to North America (Keyte et al., 2013; Ren et al., 2019). During the sampling period, dust events occurred in the regions of Poyang Lake and Dongting Lake. In conjunction with the CMAQ model and considering future global warming trends (Wang et al., 2017), we utilized the 2022 drought period in Poyang and Dongting Lakes as a baseline, along with predictive models, to evaluate the acute and long-term health risks associated with lakebed dust aerosol transport to surrounding areas. Notably, as carcinogenic risk is typically assessed over a 70-year period, we adjusted the exposure duration to 30 years, based on the duration of the 2022 drought and the projected drought scenarios in this study. It is important to mention that the short-term non-carcinogenic risk associated with Mn exposure in the regions surrounding Dongting and Poyang Lakes exceeds acceptable thresholds, while Cr presents a chronic carcinogenic risk, with concentrations surpassing permissible limits in certain areas. These insights are crucial for understanding a new source of air pollution. As climate change intensifies extreme drought conditions, lakebed dust events in East Asia may become more frequent, worsening pollution problems.

It is worth noting that extreme weather events leading to the drying of lakes on a global scale can result in significant emissions of lakebed dust aerosol, thereby increasing dust loads and atmospheric pollution at regional and global levels. This rise in dust emissions and associated pollutants in the atmospheric circulation requires further investigation to better understand its impact on air quality and climate systems.

Supplement

The supplement related to this article is available with attachment.

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Data availability

All data supporting this study and its findings will be available in an online data repository once this paper is accepted.

Author contributions

X.W. and H.Z. conceptualized the work and designed the experiments. Q.G. lead the experimental work of PAHs and heavy metals measurements. H.Z. and G.C. led the air quality modeling work. X.L. helped in experimental works. Q. G. and G. C. performed the analyses and wrote the manuscript, J.C. helped in writing. All authors contributed to the paper's writing.

Competing interests

The authors declare that they have no competing interest.

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