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Insight into the size-resolved markers and eco-health significance of microplastics from typical sources in northwest China

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Abstract

Research on atmospheric microplastics (MPs) from typical sources is limited, constraining the targeted management of pollution. Here, the characteristics and source profiles of eight types of common MPs and three classes of plasticizers (phthalates, benzothiazole and its derivatives, bisphenol A) emitted from plastic burning (PB), fruit bag burning (FB), road traffic (RT), agricultural film (AF) and livestock breeding (LB) sources were determined in $PM_{2.5}$ and PM_{10} in the Guanzhong Plain, northern China. PB features high proportions of poly(methyl methacrylate) and 2-hydroxy benzothiazole, with poly(methyl methacrylate) being more abundant in coarse particles (PM_{coarse}). FB exhibits the higher proportion of di-n-octyl phthalate in PM_{coarse} than $PM_{2.5}$. RT shows a distinguishable profile with high abundances of rubber. The abundance of 2-benzothiazolyl-N-morpholinosulfide in PM_{coarse} was twice that in $PM_{2.5}$ for RT. Polystyrene is the most abundant MP in AF. LB shows the distinguishing feature of benzothiazoles, especially 2-benzothiazolyl-N-morpholinosulfide and N-cyclohexyl-2-benzothiazolesulfenamide. The eco-health risk assessments reveal combustion-derived MPs (PB and FB) pose the highest ecological risk (Level III). Elevated hazard indices to human health were observed in LB and PB, primarily attributed to bis(2-ethylhexyl) phthalate. Notably, poly(methyl methacrylate, polyethylene terephthalate, polyethylene, bisphenol A and phthalates emerged as key drivers of oxidative stress. This study advances the understanding of atmospheric MPs, offering critical insights for source tracking and risk assessment to mitigate their eco-health effects.

Keywords: Microplastic and plasticizer emission source; size distribution; Phthalates (PAEs); eco-health risk; ROS

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1. Introduction

Global plastic production has gradually increased since the 1950s, resulting in serious environmental contamination (Klein et al., 2023). Waste plastics have accumulated in the environment to be degraded into plastic debris under the influences of UV-radiation, mechanic abrasion and temperature changes (Peeken et al., 2018; Akhbarizadeh et al., 2021). Microplastics (MPs) are plastic particles with 1 μm-5 mm in size (Can-Guven, 2021). Current research on MPs pollution sources have primarily focused on aquatic and terrestrial ecosystems (Allen et al., 2020). Understanding the sources of atmospheric MPs can assist develop efficient MPs management strategy.

Common atmospheric MPs sources include waste incineration, agricultural activities and road traffic (Panko et al., 2013; Luo et al., 2022; Yang et al., 2024; Chen et al., 2024). Incineration activities can lead to the fragmentation of plastics, accelerating the release of MPs (Luo et al., 2022; Luo et al., 2024b). Yang et al. (2021) have estimated that per metric ton of plastic can produce 360 to 102,000 MPs, primarily composed of polypropylene (PP) and polystyrene (PS). Agricultural activities also a significant contributor to atmospheric MPs (Jin et al., 2022). The large consumption of plastic film combined with short life cycle results in a number of films being left in farming soil, then transforming into MPs via degradation or fragmentation (Brahney et al., 2021; Wang et al., 2022a; Aini et al., 2023). Furthermore, tire and road wear microplastics (TRWMPs), producing from the interaction between tires and the road surface, is a significant source of atmospheric MPs (Panko et al., 2013; Liu et al., 2023). Evangeliou et al. (2020) have estimated that annual total global tire wear particle emissions were 2907 kt y^{-1}, with 29 and 288 kt y^{-1} for $PM_{2.5}$ and PM_{10}, respectively. Liu et al. (2023) have showed rubbers were the dominant compounds of TRWMPs in $PM_{2.5}$ in

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tunnels, including natural rubber (NR), styrene-butadiene rubber (SBR), and butadiene rubber (BR) polymers.

Plasticizers are widely used in the production of plastics in order to achieve the desired material properties (Demir and Ulutan, 2013). Since plasticizers are not chemically bound to the plastic products, they can easily diffuse into the surrounding environment during the life-time (Demir and Ulutan, 2013; Yadav et al., 2017). Simoneit et al. (2005) illustrated that the major plasticizers detected in particulate matters (PMs) from open-burning of plastics were dibutyl phthalate (DBP), diethylhexyl adipate (DEHA) and diethylhexyl phthalate (DEHP). Zeng et al. (2020) reported phthalate concentrations in greenhouses air were higher than that in ambient air. Liu et al. (2023) found that phthalates were the most dominant plasticizer compositions in tunnel $PM_{2.5}$, accounting for 64.8% of the detected plasticizers. Zhang et al. (2018) demonstrated that tire material-related compounds, benzothiazole (BT) and 2-hydroxybenzothiazole (2-OH-BT) were the major compounds in both tire and road dust samples. The majority of existing studies on atmospheric MPs and plasticizers have focused on analyzing the emission characteristics of individual source and lacked a comprehensive and comparative analysis of the MPs emission profiles of various sources.

MPs and plasticizers can remain suspended and spread to other areas when they emitted from the sources into the air (Gasperi et al., 2018). Airborne MPs can easily enter the human body directly via respiration compared to other environmental media, posing a serious health concern (Liao et al., 2021). Recent studies suggest that these inhaled pollutants can promote reactive oxygen species (ROS) generation (Wang et al., 2024). This ROS overproduction acts as a central driver of oxidative stress, which can damage biomolecules and disrupt cellular functions (Bates et al., 2019; Jiang et al., 2019). Oxidative potential (OP) is a metric reflecting the ability of inhaled pollutants to produce

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ROS, serving as a critical indicator of PM toxicity (Jiang et al., 2019; Bates et al., 2019; Luo et al., 2024c). Previous studies have demonstrated that metals and organic compounds can affect the OP of PMs (Ghanem et al., 2021; Luo et al., 2023). However, studies focusing on health risk assessments of MPs and plasticizers emitted from atmospheric pollution sources remain scarce.

The Guanzhong Plain located in the central of Shaanxi Province, northwestern China, inevitably consumes a large number of plastics with a developed economy and a large population (Chen et al., 2022; Wang et al., 2022b; Xu et al., 2024). The environmental conditions of strong wind and ultraviolet ray in this area exacerbate the problem of atmospheric MPs pollution. There is a notable absence of systematic comparative analyses examining the emission profiles across various emission sources, which is the key to controlling MPs pollution. The primary aims of this research are to (i) characterize the distributions of MPs and plasticizers in dual-size PMs ($PM_{2.5}$, PM_{coarse}) from typical MP sources in the Guanzhong Plain, (ii) obtain MPs and plasticizers tracers for the five typical MP sources, and (iii) evaluate the health risks of MPs and plasticizers in $PM_{2.5}$ and PM_{10}. This study could provide valuable scientific support for the development of targeted pollution control strategies, as well as sustainable improvement of the regional environment and public health protection.

2. Methods

2.1 Sample collection and gravimetric method

This study selected five typical emission sources of MPs from the Guanzhong Plain, including plastic burning (PB), fruit bag burning (FB), road traffic (RT), agricultural film (AF) and livestock breeding (LB). It should be noted that PB burned plastics including plastic bags, bottles, disposable tableware, foam boxes and other plastic daily necessities. Fruit bags are designed to enhance fruit

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quality by shielding them from pests, diseases, and direct pesticide contact, also containing some plastic components (Ali et al., 2021). The Guanzhong Plain is an important fruit production base in China, with the highest consumption of fruit bags. Local residents often use the above-mentioned plastic products to ignite solid fuels for indoor heating or cooking. Table 1 provides a summary of the essential details for each source.

During January and February 2024, $PM_{2.5}$ and PM_{10} samples were collected simultaneously from five distinct sources in three key cities of the Guanzhong Plain: Xi'an, Tongchuan, and Xianyang. The specimens were gathered using pre-fired quartz-fiber filters (QM/A, PALL, Ann Arbor, MI, USA) with a diameter of 47 mm, which had been subjected to a temperature of 780 °C for 3 hours (Wang et al., 2022b). MiniVOL samplers (Airmetrics, Springfield, OR, USA) were employed for collection, operating at a steady flow rate of 5 L min^{-1} (Wang et al., 2022b) (Figure S1). Sampling durations for each source ranged from 2 to 24 hours, depending on the emission amount. In AF and LB, the sampler was set at about 1.5 m height, corresponding to the human breathing height. For PB, FB and RT, the sampling heights were related to the height of the chimney and flyover, about 3-4 m above the ground. The field blank of each type of source was synchronously collected.

Table 1 Basic sampling information of target emission sources

Emission source	Sampling duration (h)	Sampling height	Sample No.	Sampling location
Plastic burning (PB)	2.0	3-4 m above the ground	5	Open space, about 1 m downwind of chimney of rural household stove in rural Xianyang
Fruit bag burning (FB)	2.0	3-4 m above the ground	5	
Road Traffic	13.6-14.1	3 m above	5	Open space, flyovers on traffic

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(RT)		the ground		arteries in downtown Xi'an
Agricultural film (AF)	24	1.5 m above the ground	5	Open space, about 2 m away from the greenhouse in farmland in rural Tongchuan
Livestock breeding (LB)	2.5-3.5	1.5 m above the ground	5	About 1 m from the feed trough in a cow shed of approximately 8 m^2 in rural Tongchuan

Filters were transferred using stainless steel tweezers into pre-labeled clean-air cassettes after collection and frozen at-20°C until chemical analyses. An electronic microbalance (± 1 μg sensitivity, ME 5-F, Sartorius, Germany) was used to weigh the filters before and after sampling (Wang et al., 2022b). Field blanks were processed identically to the samples. Additionally, cotton lab coats and nitrile gloves were utilized during sampling, while the use of plastic materials was minimized (Bogdanowicz et al., 2021).

2.2 Chemical analysis

This study quantified eight kinds of microplastics and three classes of plasticizers (phthalates, benzothiazole and its derivatives, bisphenol A) in $PM_{2.5}$ and PM_{10} samples.

Microplastics (MPs): To quantify the contents of the MPs, a setup was employed where a Curie-point pyrolyzer (JHS-3, Japan Analytical Industry Co., Ltd) was connected to a gas chromatography-mass spectrometry (GC/MS) system (7890GC/5975MS, Agilent Technology, USA) (Liu et al., 2023). The pyrolysates of polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), poly(methyl methacrylate) (PMMA), natural rubber (NR), styrene-butadiene rubber (SBR), and butadiene rubber (BR) were identified using mass spectrum fragments, retention times, and target product intensities compared to plastic standards. The quantified markers for the pyrolyzed compounds are shown in Table S1. Details regarding preparation of samples, instrument configurations, and QA/QC protocols are available in Sun et

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al. (2022).

Phthalates (PAEs): A thermal desorber (TD) coupled with a GC/MS system (7890GC/5975MS, Agilent Technology, USA) was utilized to analyze phthalates, including dimethylphthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DBP), butyl benzyl phthalate (BBP), bis(2-ethylhexyl)phthalate (DEHP), and di-n-octyl phthalate (DnOP). Aliquots of the filters (1.5 cm^2) were diced into smaller fragments, augmented with the internal standard Chrysene-d12, and then inserted into TD tubes for analyses (Liu et al., 2023). The sample tube was inserted directly into a GC injection port set to an initial temperature of 50 °C (Wang et al., 2016). Analytical procedure details are detailed in Ho et al. (2019) and Liu et al. (2023).

Benzothiazole and its derivatives (BTs): Nine types of benzothiazole related compounds were quantified, including benzothiazole (BT), 2-hydroxy benzothiazole (HOBT), 2-mercaptobenzothiazole (MBT), 2-aminobenzothiazole (2-NH2-BT), 2-(methylthio)benzothiazole (MTBT), 2-(4-morpholinyl)benzothiazole (24MoBT), N-cyclohexyl-2-benzothiazolamine (NCBA), 2-benzothiazolyl-N-morpholinosulfide (OBS), N-cyclohexyl-2-benzothiazolesulfenamide (CBS). The appropriate filter sample was cut and spiked with an IS of benzothiazole-d4. After a series of extraction and concentration procedures, the target analytes were washed out with 5 mL of methanol. Before the analysis, the eluates were then dried to 1 mL under a stream of nitrogen (Zhang et al., 2018). Target analytes were separated using an ultra-performance liquid chromatography system (UPLC; ACQUITY, Waters, USA) and subsequently identified with a triple quadrupole mass spectrometer (ESI-MS/MS; Xevo TQ-S, Waters, USA). Analytical details are provided in Zhang et al. (2018).

Bisphenol A (BPA): Quantification of total BPA and separation from the matrix components

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were carried out by LC–fluorescence detection (García-Prieto et al., 2008). Mobile phase was composed of acetonitrile and water (Zhou et al., 2011). The BPA standard was obtained from Sigma-Aldrich (USA). Moreover, all employed solvents and diluents were of HPLC grade and filtered through 0.45 μm membranes. The sample extracted was separated by a PerkinElmer Brownlee™ HRes Biphenyl 1.9 μm, 50 × 2.1 mm column with isocratic elution program of water: acetonitrile (6:4) at 0.5 mL min^{-1} for 4 min. The target analyte was measured using a fluorescence (FL) detector at excitation and emission wavelengths of 275 nm and 313 nm, respectively (García-Prieto et al., 2008). BPA levels were quantified based on measured peak areas (García-Prieto et al., 2008).

2.3 Oxidative potential determination with DTT assay

Four 0.526 cm^2 punches per sample from different sources were individually dissolved in 5 mL methanol (HPLC grade, Fisher Chemical) in an amber centrifuge tube and ultrasonically extracted for 2 h. The PM extract was used for the subsequent analysis.

The Dithiothreitol (DTT) consumption in this study was quantified following the methodology established by Luo et al. (2024c). 4 mL of sample extract was combined with 1 mL of 1 mM DTT solution (⩾98%, Meryer; pH 7.4 buffer), yielding a final concentration of 200 μM. At each time point (0, 5, 15, 30, 45, and 60 min), 0.5 mL of the DTT reaction mixture was added to the amber centrifuge tube preloaded with 0.5 mL of trichloroacetic acid (1%, w/v) to terminate the reaction. Subsequently, 25 μL of 10 mM 5,5'-dithiobis-(2-nitrobenzoic acid) (DTNB, ⩾98%, Meryer) and 1 mL of 1 M Tris-HCl buffer were added to each tube. The solutions (200 μL) were transferred to 96-well plates, and absorbance at 412 nm using a microplate reader (Flex Station 3 Multi-Mode, Molecular Devices). The volume-normalized DTT consumption rates for each sample were calculated from absorbance measurements taken at predetermined time points (nmol min^{-1} m^{-3}).

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All procedures were performed under dark conditions (Luo et al., 2024c).

2.4 Risk assessment model

To evaluate the potential ecological risks, the hazard indices of various MPs were estimated using the Formula 1 (Xu et al., 2018; Wang et al., 2021a). The risk index (H) was calculated by multiplying the proportion (P_n) of each polymer identified in MPs by its respective hazard score (S_n) (Lithner et al., 2011).

$$H = \sum Pn \times Sn \quad (1)$$

The average daily exposure dose (ADD) via respiratory inhalation was calculated by Formula 2, defined by the U.S. Environmental Protection Agency (U.S.EPA, 1989; Liu et al., 2023). The non-carcinogenic and carcinogenic health risks of MPs and plasticizers were quantified using the hazard quotient (HQ) (Formula 3) and incremental lifetime cancer risk (ILCR) (Formula 4), respectively.

$$ADD = \frac{C \times ET \times IR \times EF \times ED}{AT \times BW} \quad (2)$$

$$HQ = \frac{ADD}{RfD} \quad (3)$$

$$ILCR = ADD \times SF \quad (4)$$

where C represents the measured mass concentration of MPs and plasticizers from five sources. Exposure parameters included: ET (exposure time, 0.5 h d^{-1} for combustion sources (PB and FB), 1.5 h d^{-1} for others), IR (inhalation rate, 20 m^3 d^{-1}), EF (exposure frequency, 120 d y^{-1} for PB and FB, 350 d y^{-1} for others), ED (exposure duration, 30 years), AT (average exposure time, ED×365 d y^{-1}×24 h), and BW (adult body weight, 70 kg). Reference dose (RfD) and slope factor (SF) were obtained from the Integrated Risk Information System of U.S. EPA (https://www.epa.gov/iris) and Ma et al. (2020) as detailed in Table S6.

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2.5 Data analysis

Data entry and organization were conducted using Excel 2016 (Microsoft Corporation, Redmond, WA, USA), while one-way analysis of variance (ANOVA) was performed with SPSS 26.0 (IBM, Armonk, NY, USA). Spearman correlation analysis was used to assess the relationships of MPs and plasticizers with ROS, respectively. Additionally, all data are presented as mean ± standard deviation, with significant differences denoted by $P<0.05$.

The Source-Pathway-Receptor (SPR) model serves as a key tool for illustrating how environmental pollutants travel from their origins, navigate various pathways, and ultimately reach potential receptors (Waldschläger et al., 2020).

3. Results and discussion

3.1 Concentrations of microplastic and plasticizer

For the convenience of comparison, we subtracted the concentrations of MPs and plasticizers in $PM_{2.5}$ from PM_{10} in this study to obtain their concentrations in coarser particulate matter (PM_{coarse}). The total concentrations of MPs and plasticizers in $PM_{2.5}$ and PM_{coarse} from five different sources are presented in Figure 1. MPs were more enriched in PM_{coarse} in FB (59% of PM_{10}), while higher in $PM_{2.5}$ for the remaining four sources (PB, RT, AF and LB). The fruit bags are coated with wax layer for enhancing the waterproofing and durability of the material. The presence of this wax layer may affect particle formation during combustion, contributing to the creation of larger agglomerates and thus a higher proportion of coarse particles. Notably, MPs in PB and LB constituted a comparable proportion in both $PM_{2.5}$ and PM_{coarse}, both close to 50%. The variable order of MPs concentrations in the five sources in PM_{coarse} was roughly consistent with that of $PM_{2.5}$, showing PB>FB>LB>AF>RT. The average concentration of MPs was ranging from 77.7 ± 25.3 (RT) to 1906

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± 587 (PB) ng m^{-3} in fine fraction and from 41.5 ± 11.7 (RT) to 1634 ± 20.3 (PB) ng m^{-3} in coarse fraction of PMs, as summarized in Table S2. The highest MPs concentrations in fine and coarse PMs in PB source were both ~5 times higher than the averages of that in other sources. One possible explanation for this is that plastic waste can be crushed into MPs during the process of combustion (Yang et al., 2021; Luo et al., 2024a). Another important pathway for elevated MPs from PB is the resuspension of bottom ash (Yang et al., 2021).

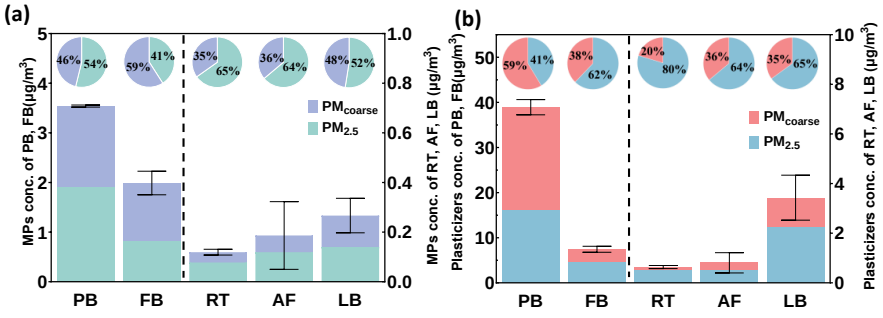

Figure 1 Average concentrations of MPs (a) and plasticizers (b) in $PM_{2.5}$ and PM_{coarse} from five sources (PB: Plastic burning, FB: Fruit bag burning, RT: Road traffic, AF: Agricultural film, LB: Livestock breeding. The error bars represent standard deviation of PM_{10}).

The total concentrations of the plasticizers in the samples were one order of magnitude higher than those of MPs (Table S2). The mass concentrations of plasticizers were higher in $PM_{2.5}$ than in PM_{coarse} for FB, RT, AF and LB sources, especially with the value of 80% in fine particles from RT. Both MP and plasticizers in RT were more abundant in $PM_{2.5}$, which enhances the potential for long-range transport and respiratory penetration. Therefore, even though the emission concentrations from RT were lower, the potential environmental and health risks posed by RT cannot be overlooked. Conversely, plasticizers in PB were abundant in PM_{coarse} (59%). The highest concentration values of plasticizer in this study were also observed in PB (15.6 ± 5.61 μg m^{-3} in

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245 $PM_{2.5}$, 22.3 ± 1.68 μg m^{-3} in PM_{coarse}), followed by FB (4.53 ± 0.39 μg m^{-3} in $PM_{2.5}$, 2.75 ± 0.65 μg
246 m^{-3} in PM_{coarse}). This is because that plastic products contain many additives to enhance their
247 performance (Do et al., 2022). Many additives are not covalently bound to the polymer matrix,
248 resulting in the liberation of plastic additives during the crushing and combustion (Do et al., 2022;
249 Billings et al., 2023). Furthermore, LB exhibited a higher emission for plasticizers in non-
250 combustion sources (RT and AF; $P<0.05$ in $PM_{2.5}$), with the values of 2.17 ± 1.05 and 1.16 ± 0.88
251 μg m^{-3} respectively for $PM_{2.5}$ and PM_{coarse}. The lack of an effective plastic recycling and disposal
252 system under the traditional retail farming may exacerbate the release of plasticizers.

253 3.2 Chemical composition of microplastics

254 The proportions of MPs identified in $PM_{2.5}$ and PM_{coarse} for the five sources are presented in
255 Figure 2. The composition of MPs from five sources varied greatly, but no significant size
256 distribution difference in the same source. RT exhibits distinctive features from other four rural
257 sources with the high proportions of both BR+SBR and NR in $PM_{2.5}$ (46.2% and 33.3% of MPs,
258 respectively) and PM_{coarse} (50.7% and 18.6% of MPs, respectively), which are the basic material of
259 tire treads. In previous studies, BR+SBR is observed to be the predominate MPs in light-duty
260 vehicle tires in the tunnel $PM_{2.5}$, and conversely, NR is extensively used in tire treads for trucks (Liu
261 et al., 2023). The RT sample collection in this work was done in the downtown flyover in urban
262 Xi'an, where light-duty cars are the dominant vehicle type, explaining the high proportion of
263 BR+SBR than NR both in $PM_{2.5}$ and PM_{coarse}.

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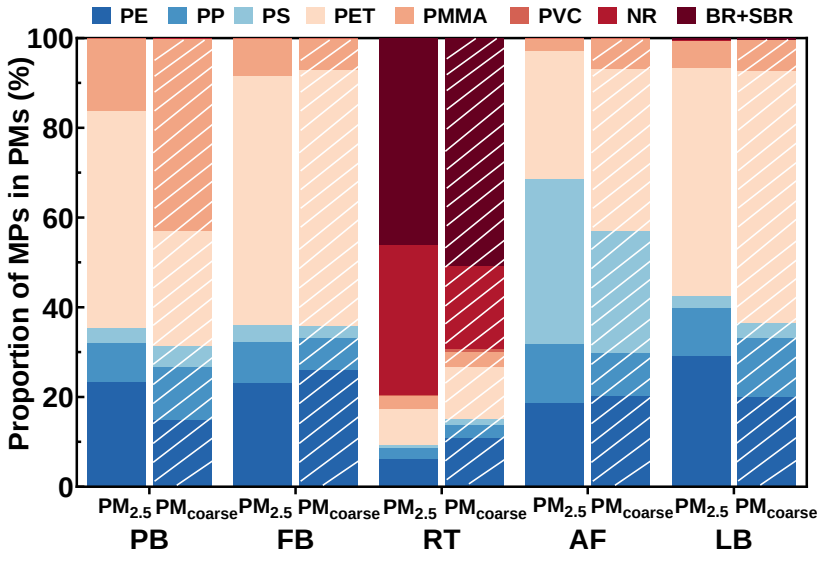

Figure 2 Chemical composition of microplastics in PM_{coarse} and $PM_{2.5}$ from the five sources.

The MP compositions of PB, FB, AF and LB were relatively similar. PET was the most common polymer type in MPs (Figure 2), which is widely used in the production of textiles. PB source inevitably included a certain amount of waste textiles, inducing the release of PET (Yang et al., 2021). Moreover, PET is widely applied in packaging and agriculture due to its advantageous properties, such as good strength, durability, elasticity, clarity, etc. (Liu et al., 2019; Lu et al., 2024). These materials may break into MPs due to wear and tear, subsequently discharging into the agricultural and breeding environment. Moreover, the highest proportion of PS was found in AF, attributed to its major ingredient in raw material of lamp-chimneys, electrical devices, packaging, etc., which are generally present in greenhouses (Qi et al., 2023).

3.3 Chemical composition of plasticizers

PAEs were the most prevalent (> 90%) among the three plasticizers in the five sources in this study. PAEs have been the most widely used plasticizer and the global production is expected to

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reach 500 million tons by 2050 (Huang et al., 2021; Billings et al., 2024). The levels of the total PAEs ranged from 468 ± 175 ng m^{-3} (AF)-15640 ± 5609 ng m^{-3} (PB) in $PM_{2.5}$ and 115 ± 54.4 ng m^{-3} (RT)-22274 ± 1680 ng m^{-3} (PB) in PM_{coarse} (Table S2). The percentages of BTs and BPA of the three detected plasticizers were typically below 2%. PB also exhibited the highest concentrations for other two kinds of plasticizers. The concentrations of BPA in PB and FB were an order of magnitude higher than RT and AF (Table S2). The result indicated that plastic incineration is the primary emission source of atmospheric plasticizers, in agreement with prior researches (Zhen et al., 2019; Chandra and Chakraborty, 2023).

Relative to other sources, RT demonstrated a higher concentration of BTs (34.8 ± 13.0 ng m^{-3} for $PM_{2.5}$, $P<0.05$; 12.9 ± 7.28 ng m^{-3} for PM_{coarse}) (Table S2). This may be related to the widespread use of BTs in tyre manufacturing and these additives are released into the air during friction between tyre and road surface (Liu et al., 2023). At the same time, some tire rubber substances were also involved in the plastic combustion source of this study. LB exhibited the highest emission of BPA among non-combustion sources, with the values of 50.9 ± 27.1 and 38.6 ± 22.2 ng m^{-3} respectively for $PM_{2.5}$ and PM_{coarse}, higher than RT (4.43 ± 1.45 and 7.8 ± 0.9 ng m^{-3}, $P<0.05$) and AF (1.4 ± 0.71 and 4.29 ± 6.68 ng m^{-3}, $P<0.05$), partly due to the migration of BPA from animal feed plastic packaging into the air (Wang et al., 2021b; Wang et al., 2021c). Furthermore, BTs, PAEs, and BPA from sources except for PB were prevalent in $PM_{2.5}$ relative to PM_{coarse}, contrary to the results reported by Nunez et al. (2020).

Compositions and distributions of PAEs: DnOP was the most abundant PAE specie across PB, FB, RT, and AF. For FB, DnOP was significantly more prevalent in PM_{coarse}, accounting for 51% of the total PAEs, compared to 36% in $PM_{2.5}$. Conversely, DnOP was more abundant in fine (59%)

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fraction of PMs than coarse (44%) in RT. As a common plasticizer, DnOP possesses a high molecular weight and low volatility, increasing its persistence in the environment. In addition to DnOP, DEHP and BBP were also identified as the major components in five sources. DEHP was a second abundant PAE component in RT (23% and 30% of PAEs in $PM_{2.5}$ and PM_{coarse}), as it has a high consumption in plasticizers market, especially in automobile industry (Zhen et al., 2019; Lu et al., 2023). While the lowest percentage of DEHP in AF in both $PM_{2.5}$ and PM_{coarse} (13%, 12%) among five sources is the significant characteristic for AF. BBP was the most abundant PAE in PM_{coarse} in LB, and the proportion was higher in coarse (40%) than fine (28%) PMs. Moreover, as shown in Figure 3, the proportions of sum of DMP, DEP and DBP were below 30% in both $PM_{2.5}$ and PM_{coarse}, and were even below 15% in FB and RT. The proportion of DEP (12% and 15% in $PM_{2.5}$ and PM_{coarse}, respectively) was the highest in PB compared to other sources, which could be used as the source marker (Figure 3a).

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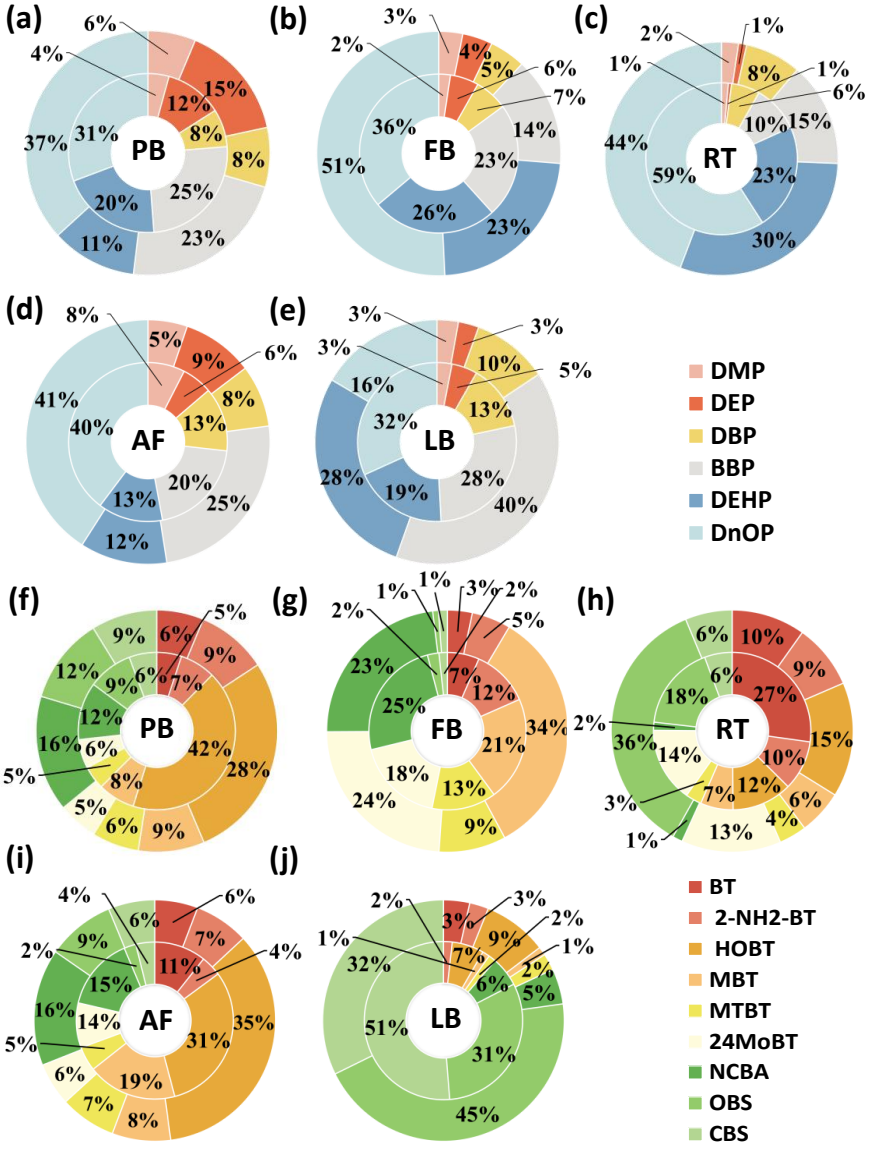

Figure 3 Mass proportions of PAEs (a, b, c, d, e) and BTs (f, g, h, i, j) in PM_{coarse} (outer ring) and $PM_{2.5}$ (inner ring) in the five typical sources.

Compositions and distributions of BTs: The distribution patterns of BTs in the five typical MP sources in $PM_{2.5}$ and PM_{coarse} were more different than PAEs. The compositions of PB and AF were quite similar, may proving that rural households use discarded agricultural film for heating or

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cooking during indoor fuel combustion. HOBT was the most abundant compound in PB and AF, with values of 42%, 31%, respectively for $PM_{2.5}$ and 28%, 35% for PM_{coarse} (Figure 3f, 3i). Furthermore, MBT was more prominent than other species for FB, with the values of more than 20%. The abundances of OBS in PM_{coarse} (36%) were higher than that in $PM_{2.5}$ (18%) for RT. Some previous studies have implied the main use of OBS in tire manufacture (Liao et al., 2018; Liu et al., 2023). BT in RT was more predominant in $PM_{2.5}$ (27%) compared to PM_{coarse} (10%), aligning with the prevalence of MPs and plasticizers in $PM_{2.5}$ in RT. A high concentration of BT in tire debris was reported from Sweden demonstrating that tire wear is the main cause of RT pollution (Avagyan et al., 2014). OBS+CBS accounted for more than 70% of BTs only in LB, which were significantly higher than those in other sources and could be used as the source markers.

3.4 Source profiles of MPs and plasticizers

The source profiles of MPs, BTs, PAEs, and BPA in PM_{10} and $PM_{2.5}$ emitted from the five emission sources are shown in Figure 4. The distribution patterns of each chemical species exhibit insignificant differences between $PM_{2.5}$ and PM_{10}. DnOP emerged as the predominant contributor across all sources, with PB being the most significant, representing 1.4% and 2.9% of $PM_{2.5}$ and PM_{10} mass concentrations, respectively. The profiles of the combustion sources (PB and FB) were more similar. However, PMMA exhibited a higher proportion in PB (0.085% and 0.23% in $PM_{2.5}$ and PM_{10}, respectively) compared to FB (0.023% and 0.041%). In addition, HOBT, the most abundant BT derivative in the current study, accounted for 0.024% in $PM_{2.5}$ and 0.037% in PM_{10} in PB, but less than 0.001% in FB. For non-combustion sources, RT were significantly influenced by tire wear particles, which characterized by high abundances of tire-related material, such as NR (0.047%) and BR+SBR (0.072%). Moreover, 24MoBT constituted the highest percentage of BTs in

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RT source, which can also be used as the indicator of vehicle emission plasticizer. PS is widely used in greenhouse agricultural films (Liu et al., 2019), which constituted a higher proportion in PMs than PET in AF, with 0.18%, 0.14% respectively to $PM_{2.5}$ and 0.16%, 0.15% to PM_{10}. OBS and CBS were the prevalent BT compounds in LB, and BPA played a more significant contribution to PM_{10} than $PM_{2.5}$ in LB source.

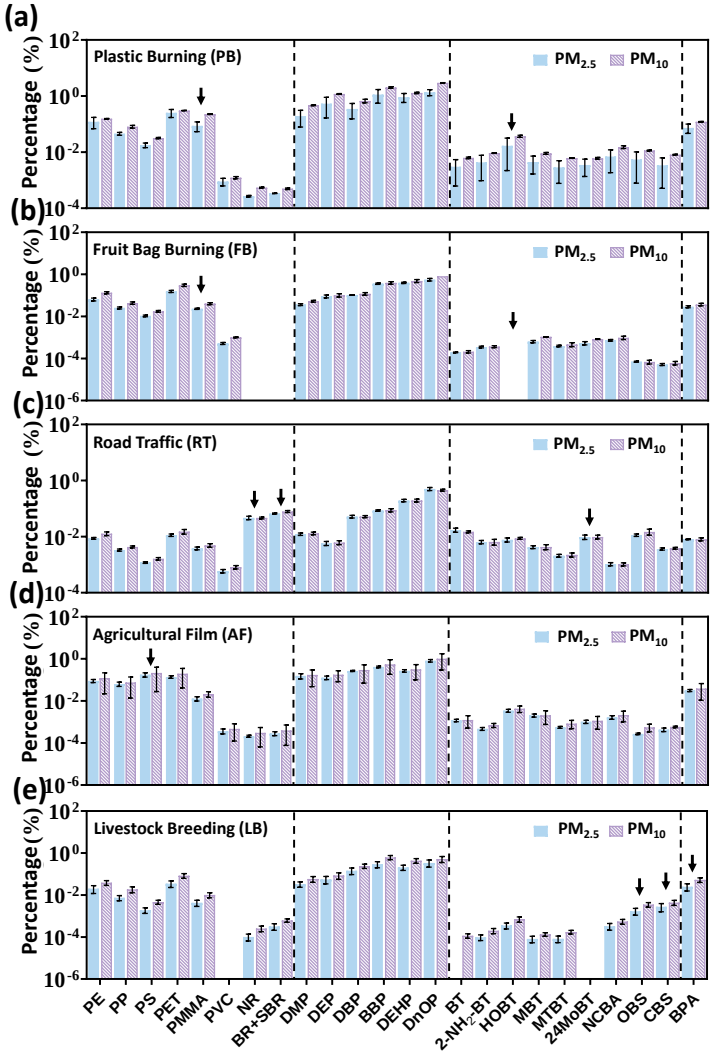

Figure 4 Source profiles of microplastics and plasticizers in $PM_{2.5}$ and PM_{10} (The black arrows

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indicate the source markers).

3.5 Eco-health significance

In this section, a comprehensive eco-health risk evaluation system was established to provide scientific support for estimating the hazards of MP and plasticizers from different sources. 1) The transport pathways of MPs and plasticizers from PB, RT, and AF sources were analyzed to clarify the exposure routes from "source" to "receptor" (Figure 5). 2) The ecological and health risks of MPs and plasticizers were assessed through different evaluation metrics (H, HI, ILCR and oxidation potential (OP)).

3.5.1 Transport pathways of MPs and plasticizers

As shown in Figure 5, plastic combustion emits MPs and attached plasticizers into the ambient air (Velis and Cook, 2021); the residual in the bottom ash can break into MPs via wind abrasion, then re-suspending into the air or depositing onto surrounding soil or into water with a risk of entering the food chain (Yang et al., 2021; Velis and Cook, 2021; Pathak et al., 2024). Small microplastics (micro-rubber) from RT emitted as airborne fine particles or trapped in the road surface, which can enter the water by surface runoff, migrating and transforming in different environmental media (Kole et al., 2017). Under ultraviolet degradation and wind erosion, agricultural films can release MPs and plasticizers into the air directly, while larger particles are deposited in farmland (Song et al., 2017). Disturbed by agricultural activities and wind, MPs created by residual films in the soil may resuspend into the air (Brahney et al., 2021; Jin et al., 2022). These pathways are all possible ways for MPs to be exposed to the human body, and controlling these pathways can reduce exposure levels.

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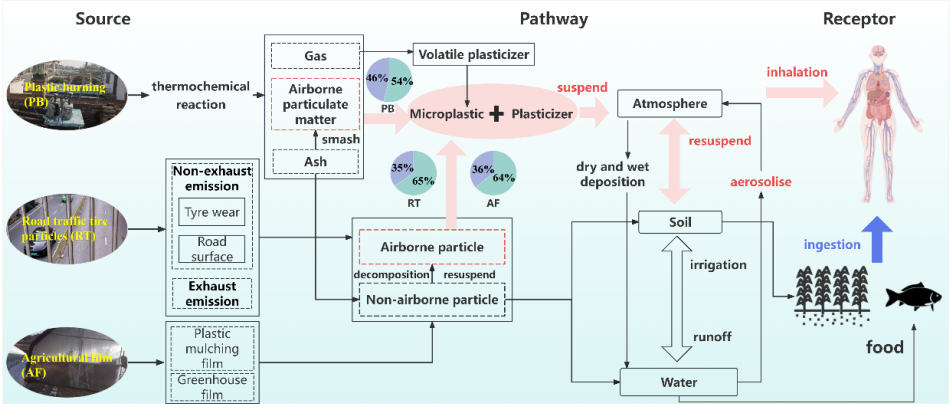

Figure 5 Source-Pathway-Receptor model associated with three different MP and plasticizer sources (The sections marked in red represent atmospheric transport pathways of MPs and plasticizers; the pie charts represent MP proportions in $PM_{2.5}$ (green) and PM_{coarse} (purple) from PB, RT, and AF).

3.5.2 Risk assessment of MPs

Based on the ecological risks of MPs for different sources (Table S5), PB and FB were categorized as Level III (high risk). This may be attributed to the fact that PMMA, a compound with high hazard score, accounted for a higher proportion of MPs emitted from combustion sources. In contrast, RT, AF, and LB sources, with lower hazard scores, were categorized as Level II (lower risk).

In this study, the health risks of MPs and plasticizers in $PM_{2.5}$ and PM_{10} from five sources were analyzed as well. The total non-carcinogenic risk (HI) ranged from 1.36×10^{-4} (AF) to 5.20×10^{-4} (LB) in $PM_{2.5}$ and 2.01×10^{-4} (RT) to 8.96×10^{-4} (LB) in PM_{10}, inconsistent with the mass concentration ranking of MPs and plasticizers in various sources. All HI values of each source were significantly lower than the international safety threshold (HI=1). The highest HI was observed in

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LB, followed by PB, with values of 4.49×10^{-4} and 8.73×10^{-4} for $PM_{2.5}$ and PM_{10}, respectively. Figure S2 illustrated the contributions of different compounds to HI. PAEs contributed most significantly to HI, accounting for more than 60% in most sources, especially in LB (93.6% and 92.9% for $PM_{2.5}$ and PM_{10}, respectively). Among all compounds, one of PAEs, DEHP displayed the highest non-carcinogenic risk (Figure S2). In RT source, BT and MBT exhibited the higher HI than other sources, with BT accounting for 20.3% ($PM_{2.5}$) and 18.1% (PM_{10}), followed by MBT (6.5% and 6.7%, respectively). Moreover, PS in AF exhibited the prominent HI values, with the proportion of 27.6% and 26.3% for $PM_{2.5}$ and PM_{10}, respectively, 10-77 times higher than other sources. These findings emphasize the need to focus on PAE in LB, BT and MBT in RT and PS in AF as a priority for MPs pollution control, aiming to minimize associated human non-carcinogenic risks.

ILCR for the three carcinogenic compounds (BT, BBP, and DEHP) were calculated in this study. The ILCR values for each compound varied between 7.03×10^{-16} and 1.77×10^{-7}, which are all below the safety threshold (10^{-6}). But this cannot be taken lightly, as there are many types of environmental pollutants, and their carcinogenic risks are additive and cumulative. Compared to other sources, LB had the highest total ILCR values (∑ILCR) (1.01×10^{-7} in $PM_{2.5}$ and 1.8×10^{-7} PM_{10}), although the mass concentration of MPs and plasticizers in this source is not the highest. Combined with the HI results, we can see that LB emit the higher concentrations of toxic MPs and plasticizers, increasing the human health risk. Comparison of the carcinogenic risks of different compounds showed that DEHP accounted for more than 97% of ∑ILCR in each source, which is the species that needs to be controlled the most in this study.

3.5.3 Effect of MPs and plasticizers on ROS generation

Figure S3 demonstrates the ROS generation capacity of $PM_{2.5}$ and PM_{10} from five sources.

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Overall, $PM_{2.5}$ exhibits a generally higher level of OP than PM_{10}, suggesting a greater contribution of fine particles to ROS generation. The larger specific surface area of $PM_{2.5}$ can enhance its reactivity with DTT and facilitate ROS production (Boogaard et al., 2012; Feng et al., 2016; Chirizzi et al., 2017). Moreover, the presence of certain components in PM_{coarse} may actually weaken the ability of $PM_{2.5}$ components to induce ROS production (Boogaard et al., 2012; Chirizzi et al., 2017). This may suggest a completely different mechanism for the generation of ROS between coarse and fine particles. Therefore, the results show that PM_{10} has lower ROS than $PM_{2.5}$ for mostly sources in this study (PB, FB, AF, and LB), which requires further research in the future.

$PM_{2.5}$ from FB exhibited the highest ROS activity with a value of 77.0 nmol min^{-1} m^{-3}, while its PM_{10} DTT value was only 18.32 nmol min^{-1} m^{-3}, indicating that OP of FB was mainly driven by $PM_{2.5}$. For PB, the DTT values of $PM_{2.5}$ and PM_{10} were 58.6 and 28.0 nmol min^{-1} m^{-3}, respectively, both at relatively high levels. In contrast, $PM_{2.5}$ from road sources showed a low OP (0.75 nmol min^{-1} m^{-3}), and RT was the only source with a higher ROS production potential for PM_{10} than $PM_{2.5}$. This is likely attributed to the unique characteristics of road dust, which is rich in coarse particles (Boogaard et al., 2012; Pant et al., 2015; Shirmohammadi et al., 2017). Road dust contains a high concentration of metal compounds, catalyzing the formation of ROS (Shirmohammadi et al., 2017). Future research should focus on the size dependency of OP for different sources, as it has significant implications for health impact.

To investigate the impact of MPs and plasticizers on OP, spearman correlation analysis was employed to assess the relationships between these compounds and DTT. As shown in Figure 6, PMMA ($R=0.77$, $p<0.01$), PET ($R=0.72$, $p<0.05$), and PE ($R=0.72$, $p<0.05$) showed a positive correlation with DTT, indicating that these components enhance ROS generation obviously.

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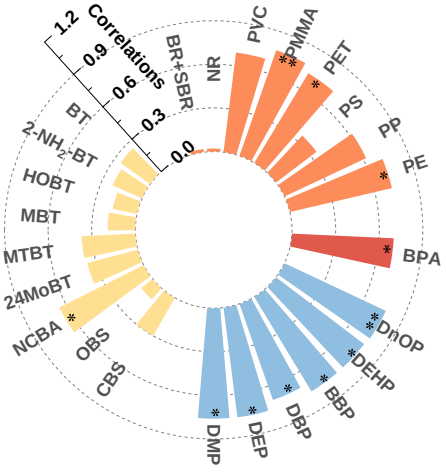
428 Additionally, all PAE species exhibited significant positive correlations with DTT (R=0.70-0.77,
429 p<0.05), especially DnOP (R=0.77, p<0.01) and DEHP (R=0.76, p<0.05). The significant
430 association of BPA with DTT (R=0.70, p<0.05) further corroborated its established impact on
431 oxidative damage (Zhang et al., 2022). However, among BTs, only NCBA showed a weak
432 correlation with DTT (R=0.65, p<0.05). These findings suggest that BTs may contribute minimally
433 to oxidative stress, on the contrary, PMMA, PET, PE, BPA and PAEs are the main drivers of ROS
434 generation.



435

436 Figure 6 Correlations between DTT, MPs, and plasticizers (*P < 0.05; **P < 0.01)

437 4. Conclusion

438 In this study, the five typical plastic emission sources in the Guanzhong Plain, China were
439 selected to investigate the characteristics of MPs and plasticizers in $PM_{2.5}$ and PM_{coarse}. The
440 concentration levels of MPs and plasticizers in combustion sources (PB and FB) were higher than
441 non-combustion sources (RT, AF, and LB), highlighting the necessity of tightening plastic
442 combustion regulations to address atmospheric MPs pollution. Most detected MP and plasticizer

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were more abundant in $PM_{2.5}$ than PM_{coarse} for most sources. PB is recognized by high loadings of HOBT, PMMA and DEP. FB exhibits the high abundances of DnOP and higher in PM_{coarse} than $PM_{2.5}$. Since tire wear particle is one of the main sources of road traffic MPs, rubber compositions (NR, BR+SBR) accounted for the highest proportions. AF is mainly characterized by high abundance of PS. The high proportions of OBS and CBS can distinguish LB from the other sources and there are still many unknown aspects of LB sources that require future research attention. This study develops a complete eco-health risk assessment system, identifying combustion sources (PB and FB) as the high ecological risk emitters, LB as the high health risk contributor, and DEHP as a key health damage pollutant due to its combined non-carcinogenic risk, carcinogenic risk, and oxidative stress generation effects. Our results could contribute to provide a scientific foundation for accurately identifying the sources and risks of atmospheric MPs, and developing efficient management strategies.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Supplementary data

Supplementary data in this manuscript can be found in Supporting Information.

Author contribution

Liyan Liu: Data Curation, Formal analysis, Writing-Original draft preparation; Hongmei Xu: Supervision, Project administration, Funding acquisition, Writing-Original draft preparation;

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Mengyun Yang: Data Curation, Investigation; Abdullah Akhtar: Investigation, Data Curation; Jian Sun: Investigation; Zhenxing Shen: Review & Edit, Supervision.

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