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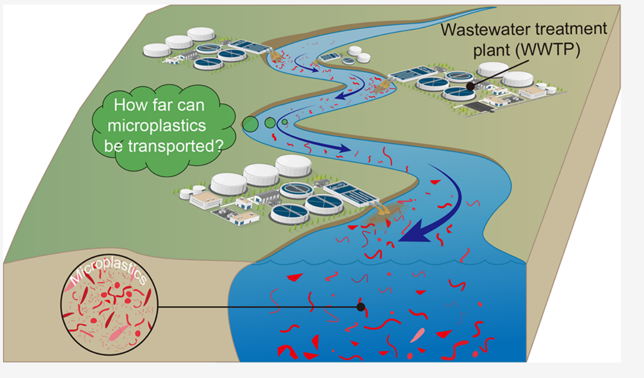
ABSTRACT: Wastewater discharge from wastewater treatment plants continuously pumps microplastics into rivers, yet their transport distances within these waterways remain unknown. Herein, we developed a conceptual framework by synthesizing the microplastic data from the Yangtze River Basin to evaluate its transport distances, quantifying a significant spatial dependence between large-scale wastewater discharge and riverine microplastics ($p < 0.05$). The presence of microplastics at a specific sampling site could be attributed to wastewater discharge within a large-scale range spanning >1000 km upstream, encompassing a substantial portion equivalent to one-third of the Yangtze River Basin. The dominance analysis indicated that the contribution of wastewater discharge in rivers with higher discharge (>100 m^3/s) to riverine microplastic pollution exceeded 65% within the Yangtze River Basin. The spatial dependence framework of riverine microplastics on wastewater discharge advances our prior understanding of the prevention and control of riverine microplastics by demonstrating that such pollution is not limited to nearby environmental factors.



KEYWORDS: *microplastics, wastewater discharge, upstream boundary, spatial dependence, basin scale, the Yangtze River Basin*

1. INTRODUCTION

Research on riverine microplastics (MPs) holds paramount significance for the fields of environmental conservation, ecology, public health, and policy formulation.[1−3] Rivers are regarded as dominant pathways or reservoirs for MP transmission and retention from freshwater to marine ecosystems.[4−6] As direct receivers of treated and untreated urban, industrial, and agricultural waste,[1,7] rivers with high MP pollution show a strong correlation with population density and wastewater treatment plant's (WWTP) effectiveness.[8−10] Although existing wastewater processes employed by WWTPs remove most MPs (98.7−99.99%),[11] wastewater discharge remains a suspected important point source of MPs in the environment.[5,12−16] A comprehensive understanding of the relationship between wastewater discharge and riverine MPs provides a scientific foundation for environmental conservation, human health, and sustainable water resource management.

Wastewater discharge, as a primary source, lacks certain traceability measures for assessing its role in riverine MP pollution. The relationship between wastewater discharge and riverine MPs is usually based on proximity to the distance from the point of wastewater discharge, with sampling sites strategically positioned both upstream and downstream to evaluate the potential impact of the discharge on MP pollution.[17−19] Due to downstream transportation of runoff with wastewater discharge, it may be necessary to trace back to more distant upstream pollution sources and consider broader MP input pathways, especially in large rivers.[20,21] Relatively little is known about how MPs move in rivers,[22,23] as their distribution in watersheds exhibits complex spatial patterns due to the location of aforementioned MP sources and transport characteristics that influence their dispersion.[5,20,24,25] MP transportation in river systems varies widely, encompassing mechanisms like bed load, settling suspended load, and wash load, all influenced significantly by the MP shape, density, and chemical composition.[26] To assess the MPs at a river sampling

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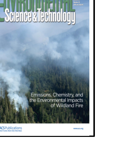

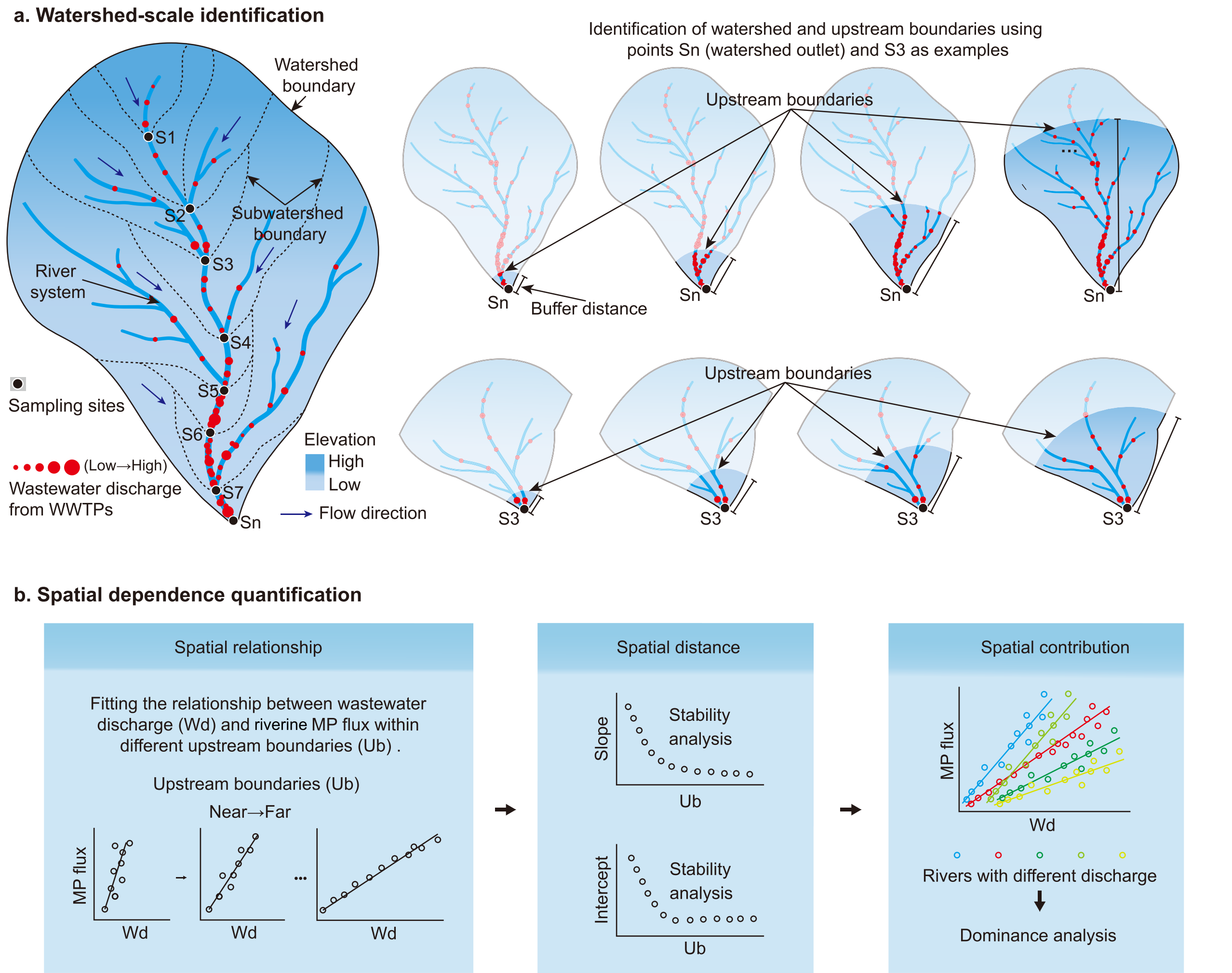

Figure 1. Conceptual framework illustrating the spatial dependence of riverine MPs driven by wastewater discharge within different upstream boundaries. (a) For each sampling site in a watershed, the total wastewater discharge is calculated within varying upstream boundaries set by different buffer distances. This ensures that upstream impacts are isolated from downstream discharges, even if they are near. Red dots represent wastewater discharge from WWTPs. Larger dots indicate higher discharge volumes, while smaller dots indicate lower discharge volumes. (S1, S2, ..., *Sn*: hypothetical sampling site locations). (b) Analysis on the spatial dependence of riverine MP flux on wastewater discharge within different upstream boundaries. This involves constructing linear relationships, determining the distance at which the relationship stabilizes through slope and intercept analysis, and assessing the contribution of wastewater discharge to riverine MPs based on rivers with different discharge.

site influenced by upstream WWTP discharges, several key factors must be considered. These include the location and discharge volume of the WWTPs, as well as the hydrological and hydraulic characteristics of the river (such as flow rate, volume, and channel morphology), and environmental conditions (like biochemical activity and the geological composition of the riverbed).[5,15,26−28] Additionally, the regional land use patterns and other potential sources of pollution should be taken into account.[28,29] These factors collectively determine the movement and distribution of MPs in water systems, making the modeling of MP transport in rivers highly complex.[27,28] Therefore, in the absence of large-scale MP flux surveys within the watershed, estimating the relationship between key variables, such as wastewater discharge from upstream WWTPs and riverine MP flux, is a feasible approach to assess the potential impact of wastewater discharge on a broader scale.

Analyzing the spatial dependence can improve our understanding of the relationship between wastewater discharge and riverine MPs. Herein, we conducted a spatial dependence framework for riverine MPs driven by wastewater discharge from upstream WWTPs, especially for a large river basin. The Yangtze River Basin in China was studied as a representative case. By systematically collecting MP data from 70 sampling sites and analyzing the MP flux in relation to wastewater discharge from upstream WWTPs, this framework offers comprehensive insights into the influence of wastewater discharge on MP pollution in major rivers. The use of regression parameters to delineate this relationship further enhances our ability to quantify and predict MP movement and abundance, providing a valuable tool for environmental researchers and policymakers. This approach not only deepens our understanding of MP pollution dynamics but also aids in developing targeted strategies for mitigating the impact of wastewater discharge on riverine ecosystems, thereby contributing to the broader goal of environmental conservation and sustainability.

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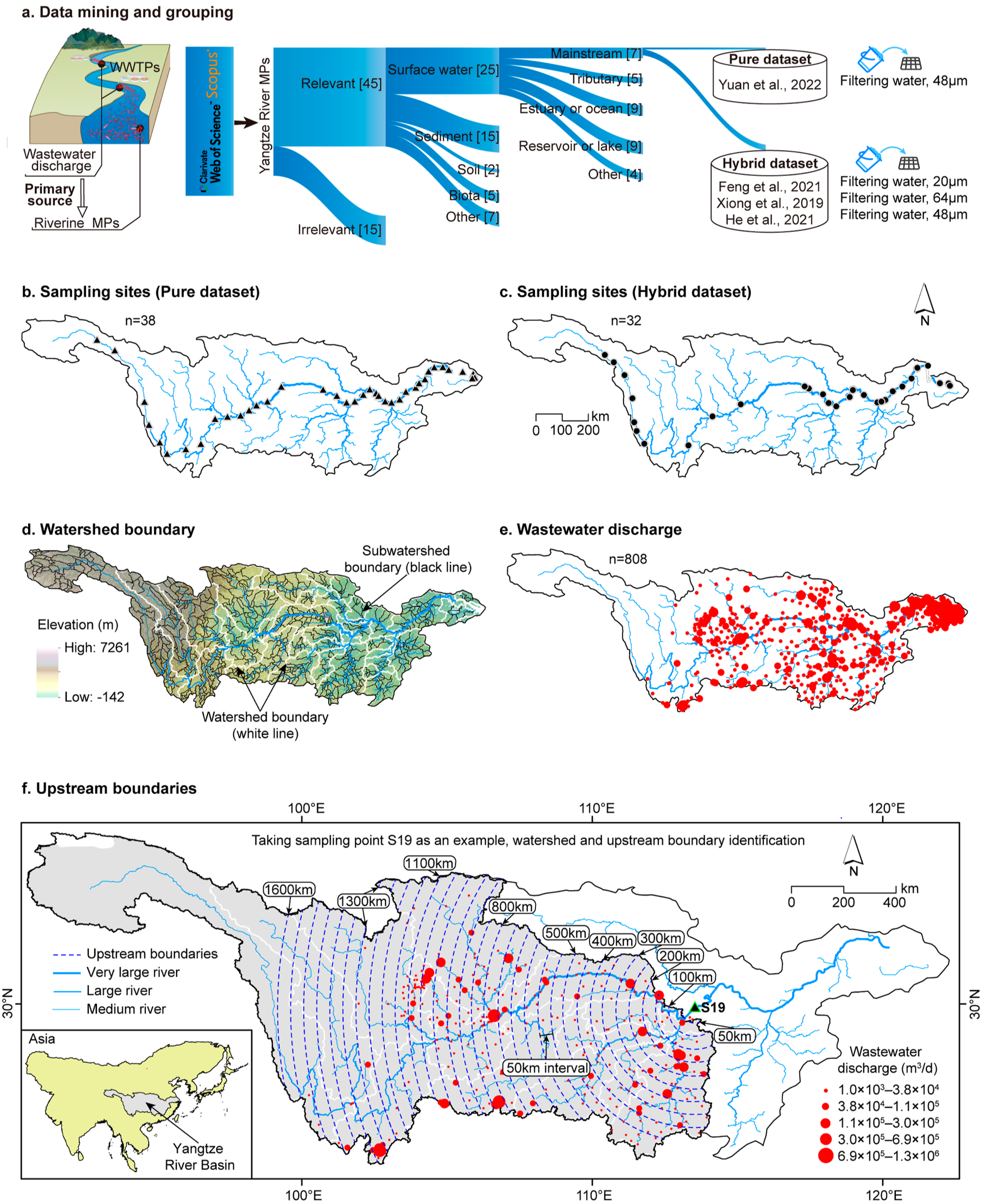

Figure 2. Spatial dependence of riverine MPs on wastewater discharge applied in the Yangtze River Basin. (a) Data mining and grouping on MPs in the Yangtze River Basin; (b) MP sampling sites in the pure data set; (c) MP sampling sites in the hybrid data set; (d) delineation of watershed and subwatershed boundaries to identify respective watershed boundaries for each sampling site; (e) wastewater discharge in the Yangtze River Basin; and (f) taking sampling site S19 as an example, the identification of watershed boundaries, along with 50 km interval buffer distance (i.e., upstream boundaries) to ascertain wastewater discharge quantities associated with different upstream boundaries. This schematic procedure was iteratively applied to all other sampling sites.

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2. MATERIALS AND METHODS

We proposed a framework for quantifying the spatial dependence of riverine MP flux on wastewater discharge from upstream WWTPs (Figure 1). This framework, divided into two main modules—watershed-scale identification and spatial dependence quantification—primarily focuses on wastewater discharge as a key correlating variable, considering its location, discharge volumes, and river discharge.

2.1. Data Mining and Grouping. *2.1.1. Literature Mining.* Starting from the perspective that wastewater discharge from WWTPs is the primary source of riverine MPs, we sought to explore the spatial extent and distribution of wastewater discharge and how it affects MP abundance in rivers. The Yangtze River, with a length of 6300 km, is the longest river in China and Asia and the third longest river in the world, with a highly mismanaged plastic waste generation.[4,30] We compiled research for the Yangtze River Basin (the most studied region on MPs in the world[31]) with high data integrity and availability by searching literature in the *Web of Science* and *Scopus* in June 2022. Our query included *"Yangtze"* and *"microplastic*"* and was limited to literature-type "*Article*", yielding 60 articles. After systematically reviewing each article and discerning its relevance, we identified 45 articles with data related to MPs in the Yangtze River. The relevant articles were further classified into five categories, including surface water (25 articles), sediment (14 articles), soil (2 articles), biota (5 articles), and others (7 articles). The subset focusing on surface water was further subdivided into five categories based on sampling site location, including mainstream (7 articles), tributary (5 articles), estuary or ocean (9 articles), reservoir or lake (9 articles), and others (4 articles). We utilized data reported for surface water from the mainstream of the Yangtze River (7 articles) in our primary analysis to contextualize the findings (Figure 2a). We exclusively selected these data for the following reasons: first, river surface water directly receives wastewater discharge; second, the scientific literature on surface water MPs in the Yangtze River was comparatively more extensive than that of other research subjects; and third, conducting MP analysis in the mainstream of the Yangtze River provides a more representative assessment than in other locations, as the overall impact on MPs can be effectively captured across the entire watershed, including the influence of wastewater discharge from tributaries.

2.1.2. Data Grouping. Data harmonization in MP research is challenging due to the use of nonstandardized methods across studies.[32] From the retrieved articles, we selected the study by Yuan et al. (2022)[21] as our benchmark (thereafter, named the "pure dataset") since it covered the entire basin, utilized a bulk sampling method for surface water, and collected data during the dry season. Sampling during the dry season helps to reduce the effects of water flow scouring on MPs, enhances sampling precision, and eliminates precipitation interference,[14,33] thus providing an accurate reflection of the relationship between wastewater discharge and riverine MPs. Other studies were integrated, owing to their investigations of the Yangtze River Basin being concentrated only on specific river sections and having relatively fewer sampling sites. This integration yielded a data set comprising multiple studies, thereby facilitating assessment of the framework's applicability. Due to the lack of uniformity in sampling methods, compiling and analyzing studies on riverine MPs can be challenging, especially since different methods and mesh sizes can significantly impact results.[34,35] To address this, we meticulously reviewed all sampling information from the seven selected articles, including the sampling method, water sample volume, sampling depth, sampling time, and mesh size (Table S1). To minimize variation caused by different sampling techniques, we focused on three studies that used water filtering as their sampling method: Feng et al. (2021), Xiong et al. (2019), and He et al. (2021), with mesh sizes of 20, 64, and 48 μm, respectively. We refer to the data from these studies as a "hybrid dataset" (Figure 2a).

2.2. Watershed-Scale Identification. *2.2.1. Hydrologic Data.* Identifying the sampling site locations, the extent of the watershed to which they corresponded, and the amount of wastewater discharge within the watershed was essential. Thus, we developed a watershed-scale process for identifying riverine MPs and the corresponding wastewater discharges within the watershed boundaries. First, we identified MP sampling site locations in the river. Second, based on the position of each sampling site, we used available hydrological connectivity data such as elevation, flow direction, and river network morphology[36] to identify the corresponding watershed area. Finally, we spatially overlaid the data of the wastewater discharge outlet points within the watershed boundaries.

We applied the above-mentioned process in the Yangtze River Basin, as shown in Figure 2. The sampling site locations of surface water MPs were vectorized using ArcGIS 10.5 software (Environmental Systems Research Institute, Inc., Redlands, CA, USA), utilizing data obtained through literature data mining. The vectorized data comprised the pure (Figure 2b and Table S2) and hybrid (Figure 2c and Table S3) data sets. The river system and watershed boundaries were used for determining the spatial extents of riverine MPs and wastewater discharge. The hydrologic features (river system and watershed boundaries) were derived from the HydroRIVERS and HydroBASINS databases (https://www.hydrosheds.org/, accessed on August 15, 2022). The river system was divided into very large (>10,000 m^3/s), large (1000−10,000 m^3/s), and medium (100−1000 m^3/s) rivers according to the amount of discharge.[37] Different watershed and subwatershed boundaries were selected according to the MP sampling site locations (Figure 2d). After identifying the riverine MPs at the sampling sites in the mainstream of the Yangtze River Basin and the corresponding watershed boundaries, we proceeded to calculate the wastewater discharge from WWTPs within the watershed area. The wastewater discharge data from WWTPs were derived from the HydroWASTE database, which is a spatially explicit global database of 58,502 WWTPs and their characteristics (https://www.hydrosheds.org/, accessed on September 3, 2022). To estimate wastewater discharge amounts, we employed the "WASTE_DIS" field, which refers to the amount of treated wastewater discharged by the WWTPs in m^3/day (m^3/d).[38] A total of 808 WWTP outfalls were reported in the Yangtze River Basin with a total discharge of 2.94×10^7 m^3/d (Figure 2e).

2.2.2. Upstream Boundaries. Upstream boundaries in geography refer to the upstream extent of a watershed, specifically the area along the river in the region through which it flows in the upstream direction. Herein, we utilized the buffer function in ArcGIS 10.5 software to establish the upstream boundaries corresponding to each sampling site at different buffer distances. The buffer distance was centered on the sampling site, extending a specified spatial distance

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outward to generate a new region,[39] enabling the spatial dependence of riverine MPs on wastewater discharge to be assessed within varying upstream boundaries, with a buffer distance interval at 50 km. Notably, some sampling sites lacked an upstream watershed beyond a certain distance, resulting in identical wastewater discharge characteristics for all analyses conducted beyond that point. The identification of upstream boundaries applied in the Yangtze River Basin is illustrated in Figure 2f.

2.3. Spatial Dependence Quantification. We constructed an approach to elucidate the spatial dependence of riverine MPs on wastewater discharge within various upstream boundaries. This spatial dependence was mainly characterized by spatial relationship, distance, and contribution. First, we established the spatial relationship between the MP flux and wastewater discharge within different upstream boundaries. Second, we calculated the spatial distance by considering the trends in the slope and intercept. Finally, we estimated the spatial contribution of wastewater discharge in rivers with different discharges to riverine MP pollution.

2.3.1. Spatial Relationship. We performed least-squares linear fitting to analyze the relationship between wastewater discharge and riverine MP flux within different upstream boundaries. Additionally, we conducted normality, heteroscedasticity, and autocorrelation tests on the residuals of the fitted linear models using the Shapiro−Wilk, Breusch−Pagan, and Durbin−Watson tests, respectively. In Python, these tests were performed using the "scipy.stats.shapiro" function for the Shapiro−Wilk test, "statsmodels.stats.diagnostic.het_breuschpagan" for the Breusch−Pagan test, and "statsmodels.stats.stattools.durbin_watson" for the Durbin−Watson test. These tests aimed to verify the appropriateness of the model assumptions, thereby ensuring accuracy and reliability of the model predictions. Lastly, we plotted the change in Pearson's correlation coefficient (r) between wastewater discharge and riverine MPs within different upstream boundaries to observe their variations. A significantly high and strong r value ($r > 0.6$, $p < 0.05$) suggested a significant and correlated impact of the independent variable (wastewater discharge) on the dependent variable (riverine MP flux). By contrast, a nonsignificant r value indicated that the MPs at the sampling site were strongly influenced by factors other than wastewater discharge, leading us to reject the hypothesis that wastewater discharge was the primary source of riverine MPs for that site.

To further discuss the differences between the pure and hybrid data sets, we examined the spatial autocorrelation degree of riverine MP abundance at sampling sites and their proximity to cities. Spatial autocorrelation, which refers to the correlation between observations at different locations in geographical space,[40] was analyzed because we speculated a cumulative effect on riverine MP abundance from upstream to downstream in a large basin.[41] We utilized GeoDa software (https://geodacenter.github.io/index-cn.html) to compute the spatial autocorrelation, employing Global and Local Moran's I indices as measurement metrics. The Global Moran's I index provides insights into the overall spatial autocorrelation across the entire geographical space, with a numerical value ranging from −1 to 1. Positive values indicate clustered spatial patterns, implying the aggregation of similar values; negative values suggest dispersed spatial patterns, indicative of dissimilar values, while values close to zero imply a lack of discernible spatial autocorrelation, indicating a relatively random spatial distribution. Furthermore, the Local Moran's I index facilitates finer-grained analysis than the Global Moran's I index, allowing for the identification of specific local patterns. High values spatially clustered together signify a high−high cluster, whereas low values exhibiting clustering indicate a low−low cluster. This nuanced approach provides a detailed understanding of localized spatial patterns, contributing to a comprehensive analysis of the spatial distribution of MP abundance. The distance calculation for the sampling site to the nearest city was implemented with ArcGIS 10.5 software. Meanwhile, urban levels were classified based on administrative hierarchy from high to low, including provincial capitals, prefecture-level cities, and county-level cities, generally corresponding to their respective population size.

2.3.2. Spatial Distance. The deposition of riverine MPs due to wastewater discharge is influenced by various factors, limiting their impact downstream. Thus, the MPs at a given river sampling site are likely closely related to wastewater discharge within a certain upstream boundary, and the relationship between wastewater discharge and riverine MPs becomes relatively stable. In this state, the increase in wastewater discharge no longer substantially influences the MP abundance, and the slope and intercept of the linear equation should exhibit minimal changes and remain at relatively small values.[42] Actually, if wastewater discharge within a certain range provides sufficient predictive information, additional wastewater discharge beyond a specific upstream boundary may be less important. For this study, we performed a stability analysis of the independent variable (i.e., wastewater discharge) and the dependent variable (i.e., riverine MP flux) based on the equation parameters (slope and intercept) of the linear fit. To determine stability, we plotted a line graph of slope and intercept trends for different upstream boundaries. We considered the data in the upstream boundary series as tending toward stationarity when the variance of ten consecutive data points after a specific point was <5% or 10%.[43] To further verify the stability of the series data, we conducted the augmented Dickey−Fuller (ADF) test to determine the presence of a unit root in both series around a specific point.[44] Based on the determination of stability and results of the ADF test, we identified the upstream boundary wherein the variance was minimal, and the series was stationary. This boundary represented the spatial distance at which riverine MP abundance depended on wastewater discharge.

2.3.3. Spatial Contribution. Riverine MP abundance was related to the amount of wastewater discharged from WWTPs and the type of river in which the wastewater was discharged. For example, wastewater discharged into a slow-flowing river may result in relatively fewer MPs being carried downstream and a great proportion of MPs being deposited.[14] Conversely, MPs are more likely to be transported downstream when wastewater is discharged into a fast-flowing river, potentially leading to MPs being carried over longer distances.[28] To address this phenomenon, we categorized the wastewater discharge in rivers into five groups based on the "RIVER_DIS" field: X1, very low discharge (0−10 m^3/s); X2, low discharge (10−100 m^3/s); X3, medium discharge (100−1000 m^3/s); X4, high discharge (1000−10,000 m^3/s); and X5, very high discharge (>10,000 m^3/s)[37] (Table S7). This classification allowed us to examine the contribution of wastewater discharge to riverine MPs under different river discharge conditions.

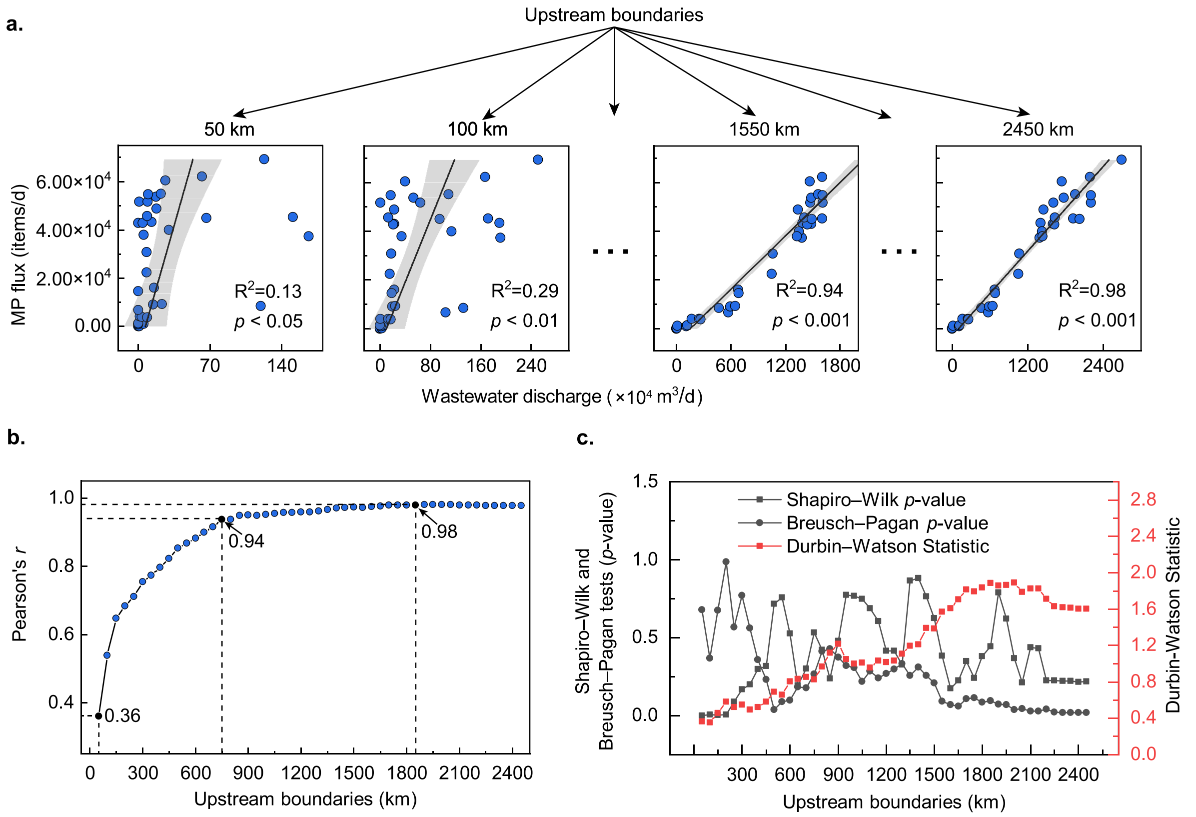

Figure 3. Spatial relationship between wastewater discharge and riverine MP flux within different upstream boundaries in the pure data set. The Shapiro–Wilk test, with p-values < 0.05 indicating non-normal residuals, the Breusch–Pagan test where significant values (p-values < 0.05) suggest heteroscedasticity, and the Durbin–Watson test where values near 2 imply no autocorrelation, values closer to 0 indicate positive autocorrelation, and values closer to 4 indicate negative autocorrelation.

For this purpose, the primary boundary range affecting MPs in the Yangtze River was identified based on spatial distance, and further assessments were conducted within this upstream boundary range to examine the correlation between wastewater discharge in rivers with different amounts of discharge and riverine MPs. Significant correlations provided preliminary evidence of the relationship. Subsequently, we performed dominance analysis (DA)[45] by constructing multiple linear regression equations to analyze the relative contributions of wastewater discharge in rivers with different discharge amounts to the MP flux. DA, a method for assessing the relative importance of independent variables in multiple regression, reveals the contribution of each variable by comparing the reduction in explanatory power when excluding individual variables, providing insights into their relative contributions to the overall model. The multiple linear regression equation could be expressed as shown in eq 1. The contributions of independent variables should be equivalent to their marginal utility in the goodness of fit (R^2). The marginal utility of the independent variable x_k to R^2 could be expressed as shown in eq 2. Herein, the analysis was mainly implemented using the "domin" command in Stata MP17 software (StataCorp LLC, College Station, TX, USA). Five independent variables were introduced for wastewater discharge in rivers with X1, X2, X3, X4, and X5. A multiple linear regression equation was obtained by substituting these variables into eq 1, which was then applied to eq 2 to obtain the contributions of the five independent variables to riverine MPs

$$y = a + b_{X1}x_{X2} + b_{X2}x_{X2} + b_{X3}x_{X3} + b_{X4}x_{X4} + b_{X5}x_{X5} + e \quad (1)$$

$$M_K = R^2[y = a + \sum_{j \in s} b_j x_j + b_k x_k + e] - R^2[y = a + \sum_{j \in s} b_j x_j + b_k x_k + e^*] \quad (2)$$

where s is another independent variable that does not contain the variable k. This equation was employed to essentially calculate the difference between the R^2 values of complete regression and the regression excluding the variable k. Since the coefficient of regression usually changes after removal of an independent variable, the coefficient of regression that did not include the variable k was expressed as *.

3. RESULTS

3.1. Riverine MP Flux Is Correlated with Upstream Wastewater Discharge.

Wastewater discharge from WWTPs and riverine MP flux was significantly correlated ($r > 0.6$, $p < 0.05$), especially within the pure data set. In this data set, the correlation between wastewater discharge and riverine MP flux showed a linear fit at upstream boundaries of 50 km ($R^2 = 0.13$; $p < 0.05$), 100 km ($R^2 = 0.29$; $p < 0.01$), and 1550 km ($R^2 = 0.94$; $p < 0.001$). As the upstream boundary increased at 50 km intervals, reaching the maximum upper boundary of 2450 km (i.e., the distance from the sampling site at the Yangtze River estuary to the farthest wastewater discharge outlet in the upper reaches), the R^2 value of the linear fit reached 0.98 ($p < 0.001$) (Figure 3a). Meanwhile, the

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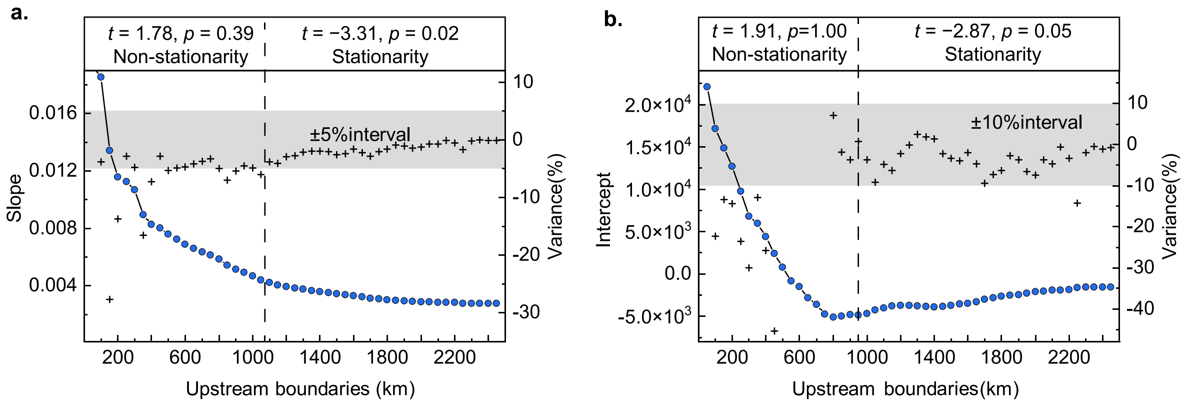

Figure 4. Trends in the slope and intercept used to determine the spatial distance between wastewater discharge and riverine MP flux in the pure data set. The *t*-value represents the test statistic of the ADF test. A *p*-value < 0.1 rejects the original hypothesis, indicating stationarity, whereas a *p*-value > 0.1 fails to reject the original hypothesis, suggesting nonstationarity. The gray areas depict ±5% or ±10% variation intervals.

Pearson's correlation coefficient (r) value gradually increased from 0.36 at 50 km to 0.94 at 750 km, followed by a less pronounced change in the trend, reaching its peak of 0.98 at 1900 km (Figure 3b and Table S2). In the hybrid data set, the correlation showed a linear fit at the upstream boundaries of 50 km (R^2 = 0.10, p = 0.08, not significant) and 100 km (R^2 = 0.22, $p < 0.01$). The R^2 value of the linear fit progressively increased in 50 km increments, reaching 0.48 ($p < 0.01$) at the maximum upper boundary of 2450 km (Figure S1a). Concurrently, the r value demonstrated a gradual increase from 0.31 at 50 km to a peak value of 0.75 at 1050 km, with notably elevated values observed within the range of 900−1200 km (Figure S1b and Table S3).

A linear regression model with a strong r and satisfied assumptions ensures reliable predictions and valid insights at long upstream boundary distances in the pure data set. Applying the Shapiro−Wilk normality tests in the pure data set, it was evident that MP flux significantly deviated from normality in the upstream boundaries between 50 and 200 km, with p-values all below 0.05 (Figure 3c and Table S4). However, from 500 km onward, the distributions approached normality, particularly between 1350 and 1450 km, where p-values neared 0.9, indicating a closer adherence to a normal distribution. The Breusch−Pagan test results generally showed p-values greater than 0.05 across most distances, indicating a lack of strong evidence of heteroscedasticity in the residuals of our linear models. However, notable exceptions included a significant result at 500 km (p = 0.037) and a trend toward lower p-values in distances beyond 2000 km, suggesting potential issues in uniform variance as distance increase. The Durbin−Watson statistic demonstrated a decrease in positive autocorrelation as upstream boundaries increased, with values steadily rising from 0.366 at 50 km to 1.891 at 2000 km. This pattern suggests that autocorrelation concerns were substantially reduced at greater distances, enhancing the reliability of the regression models applied. Additionally, the hybrid data set, comprising different upstream boundaries for wastewater discharge and MP flux, underwent linear regression fitting as well as tests for basic assumptions of linear models. The fit was inferior compared to the pure data set, and the results of the assumption tests were not satisfactory (Figure S1 and Table S5). This implied that the hybrid data set might have had significant heterogeneity or complexity, making it difficult for traditional linear regression models to effectively explain or predict the relationship between the wastewater discharge and riverine MP flux.

3.2. Riverine MPs Are Subject to Long-Distance Wastewater Discharge. Wastewater discharge within an upstream boundary of >1000 km significantly influenced the presence of riverine MPs in the Yangtze River ($p < 0.01$). The linearly fitted equations for wastewater discharge and riverine MP flux within different upstream boundaries indicated that the slope and intercept gradually reached a steady state after a certain buffer distance. This implied that wastewater discharge less influenced MPs beyond a specific upstream boundary. The slope in the pure data set continued to decrease from 0.019 at 50 km to 0.0044 at 1050 km, with a maximum variation of over −25.00% (Figure 4a and Table S6). The slope then decreased from 0.0042 at 1100 km to 0.0028 at 2450 km, but the variation was <5% at ten consecutive points, with a maximum variation of −4.06% (Figure 4a). The ADF test results indicated that the test statistic (t) for series with an upstream boundary <1050 km was 1.78 (p > 0.1), indicating non-stationarity. The test statistic (t) for series with an upstream boundary >1100 km was −3.31 (p < 0.05), suggesting stationarity (Figure 4a). The intercept in the pure data set decreased from 22, 075.45 at 50 km to −5109.47 at 800 km, with a maximum variation of over −40.00% (Figure 4b and Table S6). From 850 to 2450 km, the intercept fluctuated from −5016.24 at 850 km to −1535.57 at 2450 km. Although not all ten consecutive points had a variation of <10%, but 97.00% of points had variation of <10%, with a maximum variation of 14.24% (Figure 4b). The ADF test results also showed that the test statistic (t) for the series with an upstream boundary <900 km was 1.91 (p > 0.1), indicating nonstationarity. By contrast, the test statistic (t) was −2.87 (p < 0.05) for the series with an upstream boundary >1000 km, indicating stationarity.

3.3. Variable Contribution of Wastewater Discharge from Different Locations. The contribution between wastewater discharge and riverine MP flux within the Yangtze River Basin was more pronounced in rivers with a discharge exceeding 100 m^3/s compared to those with lower discharge levels. The contribution of wastewater discharge in these higher discharge rivers to MP pollution in the mainstream of the Yangtze River exceeded 65%. All wastewater discharges in rivers with different discharge amounts exhibited a significant correlation with riverine MP flux. The R^2 values for X1 (rivers with very low discharge), X2 (rivers with low discharge), X3 (rivers with medium discharge), X4 (rivers with high

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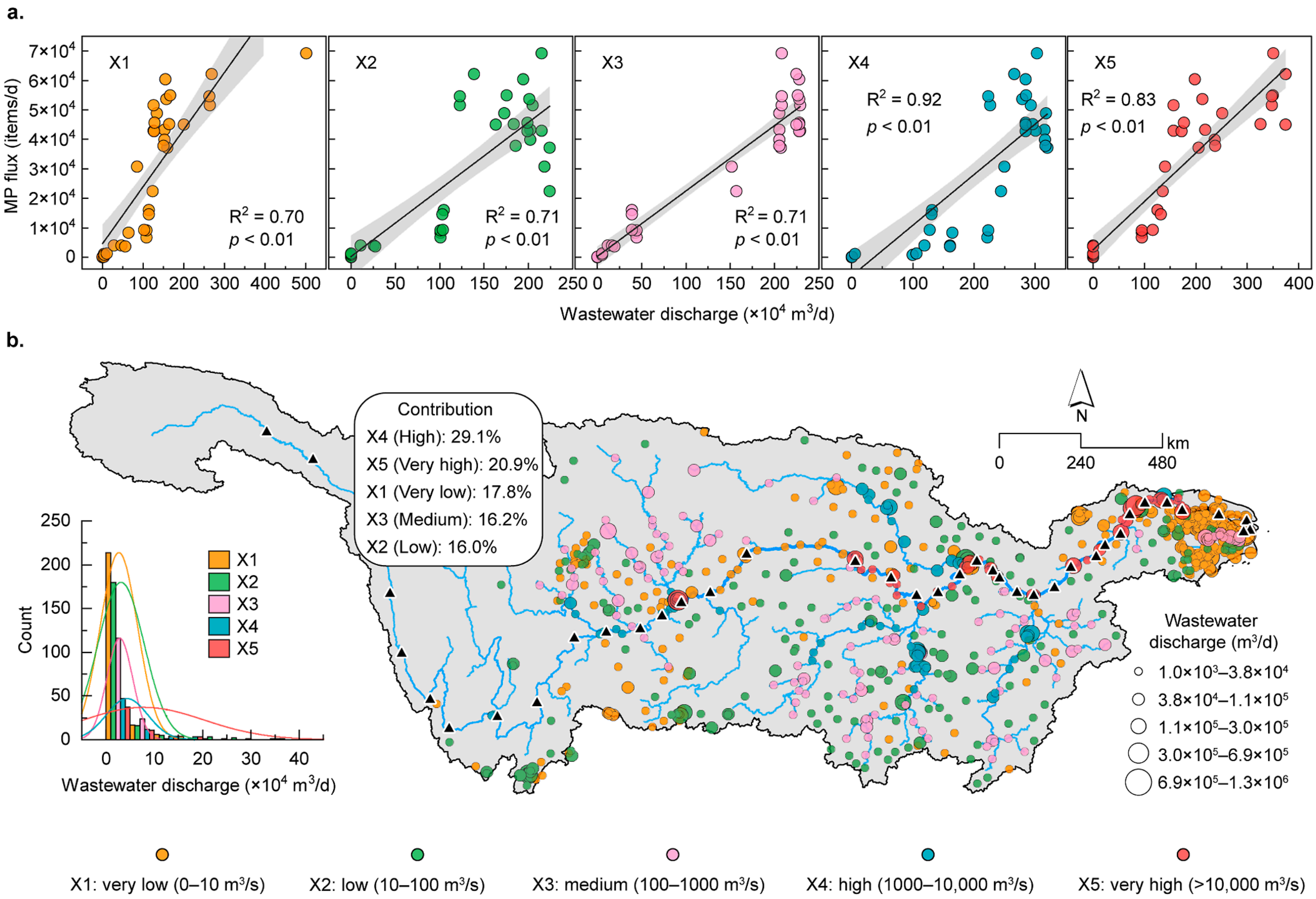

Figure 5. Spatial contribution of wastewater discharge to riverine MP flux under different river discharge conditions. X1: very low (0−10 m^3/s); X2: low (10−100 m^3/s); X3: medium (100−1000 m^3/s); X4: high (1000−10,000 m^3/s); and X5: very high (>10,000 m^3/s).

discharge), and X5 (rivers with very high discharge) were 0.70 ($p < 0.01$), 0.71 ($p < 0.01$), 0.71 ($p < 0.01$), 0.92 ($p < 0.01$), and 0.83 ($p < 0.01$), respectively (Figure 5a). The relative contributions of wastewater discharge to MP pollution indicated that the highest contribution occurred in rivers with high discharge (X4), reaching 29.1%, followed closely by those with very high discharge (X5), with a contribution of 20.9%, while rivers with low discharge contributed to 17.8% of MP pollution (with the highest amount of wastewater discharge under this condition). Rivers with medium discharge contributed to 16.2% of MP pollution, and those with low discharge had the lowest contribution at 16.0% (Figure 5b). The analysis revealed that wastewater discharge in rivers with low and very low discharge collectively contributed to >50% of total basin discharge, although their contribution to MP pollution in the mainstream of the Yangtze River was only 32%. The results indicated that MPs in the mainstream of the Yangtze River Basin primarily originated from wastewater discharge in rivers with a higher discharge. Nevertheless, the contribution from rivers with a low discharge could not be overlooked.

4. DISCUSSION

4.1. Data Set with Positive Spatial Autocorrelation Better Explains the Relationship with Basin-Scale Wastewater Discharge. The strong correlation between long-distance upstream wastewater discharge and riverine MP flux was confirmed through hypothesis testing, but the spatial dependence framework proved ineffective for the hybrid data set. Since the wastewater discharge at each sampling site was known and constant across different upstream boundaries, the variations in discharge were solely due to differing buffer distances. However, the abundance of MPs in the river was influenced not only by wastewater discharge but can also be affected by surrounding environmental factors.[14,15,18,20] Several reasons may account for why the pure data set fits the spatial dependence framework, while the hybrid data set did not. The Moran's I index of the pure data set was up to 0.90, indicating a strong spatial autocorrelation of riverine MPs and a cluster distribution (Figure 6a). By contrast, the Moran's I index of the hybrid data set was 0.53, indicating a weaker spatial autocorrelation than in the pure data set (Figure 6b). The MP abundance demonstrated a significant high−high cluster ($p < 0.05$) in the lower reaches of the basin, while the upper and middle reaches showed a significant low−low cluster ($p < 0.05$), with a good clustering effect. The high−high and low−low cluster distribution of riverine MPs became more evident as the value of Moran's I index increased, suggesting that neighboring sampling sites had similar values, and that MP abundance at downstream sites was significantly influenced by upstream sites with similar abundance values. Therefore, we could infer that the primary driver for the riverine MP data set with strong spatial autocorrelation was long-distance wastewater discharge, while the data set with weak spatial autocorrelation was influenced not only by wastewater discharge but also significantly by other factors.

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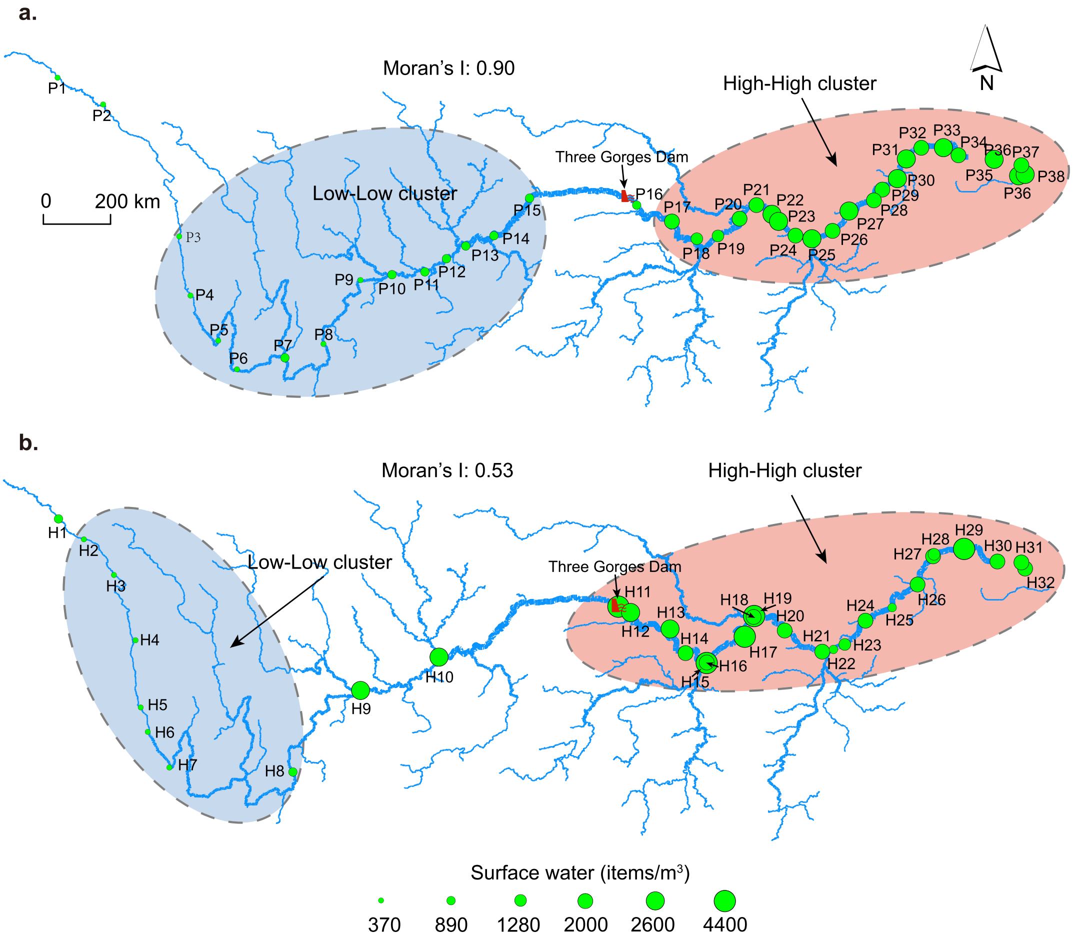

Figure 6. Spatial autocorrelation of MP abundance in the (a) pure and (b) hybrid data sets in the Yangtze River Basin. The Global Moran's I index measures spatial autocorrelation: positive values indicate clustering, negative values suggest dispersion, and values near zero imply randomness. The Local Moran's I highlights a specific pattern with a high−high cluster of high values and a low−low cluster of low values.

The pure data set, sourced from a single article,[21] exhibited a high Moran's index of 0.90, indicating extremely high spatial autocorrelation between sampling points (Figure S2a). Despite efforts to ensure the selection of studies utilizing a consistent sampling method, the Moran's index of the hybrid data set, derived from three different articles, was 0.53, indicative of a reduced spatial autocorrelation. Therefore, we formulated various hypotheses to elucidate the reasons underlying the spatial heterogeneity observed in the hybrid data set. Notably, in the hybrid data set, there were significant variations in MP abundance at two sampling sites near each of Yueyang, Wuhan, and Nanjing, with up to a fourfold difference across seasons (Figure S2b). The variability in MP abundance at the same location was a notable characteristic, primarily influenced by factors such as seasonal changes, human activities, climatic conditions, and urban emissions.[33,34] Furthermore, the obstructive effect of the Three Gorges Dam located in the upper reaches of the Yangtze River caused in the deposition of MP in the river, resulting in significantly higher MP abundance upstream of the dam.[46,47] In addition, the lack of standardized and completely consistent sampling methods may have contributed to differences in MP abundance, thus affecting the spatial autocorrelation between sampling sites.[34,48]

In the hybrid data set, the median distance of sampling sites from cities was 9.8 km, positioning them closer to urban areas compared to those in the pure data set, in which the median distance was 16.8 km (Figure S2c). Additionally, nearly 50% of cities proximate to the hybrid data set's sampling sites were populous prefecture-level cities, whereas 37% of adjacent cities were more populated prefecture-level cities in the pure data set, with the majority (47%) being less populated county-level cities (Figure S2d). This arrangement indicated that the selection of sampling sites in the hybrid data set was designed to focus on studying the spatial variability of MPs specifically in urban rivers. We hypothesize that the reduced spatial autocorrelation among sampling sites situated in urban rivers was attributable to the variability in MP abundance, influenced by discharges from nearby and upstream WWTPs. Additionally, the substantial influx of MP entering the river through road discharge and sewer sediment scouring, often exacerbated by inadequate municipal waste management practices, likely contributed to this variability.[49−51] In summary, the spatial dependence framework of riverine MPs on wastewater discharge is suitable for the pure data set. However, this correlation may not be applicable to sampling sites near urban areas, which may provide a data set with spatial variations that are strongly influenced by the surrounding environment. These data sets exhibited nonspatial gradient characteristics from upstream to downstream.

4.2. Basin-Wide Dependence of Riverine MP Flux on Wastewater Discharge. We proposed a new strategy for quantifying the source of MPs and describing their spatial pattern. The size of the river channel and amount of discharge affects the flux and transport distance of pollutants in

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rivers.[27,52,53] Herein, we highlighted that MPs in the Yangtze River could be traced back from wastewater discharge over a distance of almost 1000 km. With the increase in the upstream boundary, the dependence of riverine MPs on wastewater discharge tended to be stable, indicating that the relationship between MP flux and wastewater discharge seemingly reached a state of equilibrium and no longer exhibited a significant increase, as initially occurred with the increasing amount of wastewater discharge. This conclusion was supported by riverine MP transport[27] in which MP loss per kilometer from the source (river discharge: 0.01 m^3/s) to the mainstream (river discharge: 70 m^3/s) was reportedly 5%. Hence, it was estimated that MPs discharged at a certain point could be transported up to 50−100 km. Based on the study findings, we can further infer that the transport of MPs in the Yangtze River can reach >1000 km.

The primary contribution to MP pollution in the Yangtze River originated from wastewater discharge near the main channel. On the one hand, this was because low-discharge rivers were often located in the watershed's tributaries,[37] leading to long distances between sampling sites for MPs in the mainstream. On the other hand, low-discharge rivers have limited capacity for MP transport, which may result in deposition before reaching the mainstream.[27] The results of the DA indicated that, despite very small and small rivers contributing only to 32% of the wastewater discharge in the mainstream of the Yangtze River, the wastewater discharge into such water bodies exceeded 50% of the total basin discharge. Therefore, in studying pollutants at a large basin scale, we should not only pay attention to mainstream wastewater discharge but also recognize the contribution of tributary wastewater discharge.

4.3. Outlook. Our study established a strong linear correlation between wastewater discharge (key correlating variable) and riverine MP flux within long upstream boundaries, which passed the hypothesis tests. The approach proposed in our study effectively simplified the complex transport processes of riverine MPs by focusing on calculating the correlation between wastewater discharge and river MP flux, efficiently utilizing existing data, and avoiding the challenges of mechanistic modeling.[27,28] By establishing a strong linear relationship between these variables over long distances, this study inferred the influence of long-distance wastewater discharge on the river MP flux. However, our spatial dependence framework primarily considered the riverine MP flux, wastewater discharge from WWTPs, spatial location of the wastewater discharge, and impact of different runoff volumes entering the river. These were the main external factors affecting the transport distance of riverine MPs. Our model did not directly explore the differences between MPs with different characteristics, such as density, shape, and chemical composition, nor did it include the impact of biofilms produced by wastewater discharge on the transport of MPs in rivers.[15,16,28] Despite these limitations, we believe that it is feasible to use our spatial dependence framework to establish the spatial dependence relationship between MPs with different characteristics and the amount of wastewater discharge.

A comprehensive mass balance approach to MPs is needed to quantify the spatial dependence of riverine MP abundance on wastewater discharge from WWTPs in aquatic ecosystems. While it is feasible to address how much the upstream boundary range influences the significance of the correlation between wastewater discharge and MP abundance, we recommend employing a mass balance approach to obtain a thorough understanding.[30,54,55] In future mass balance calculations of riverine MPs, the aquatic system boundaries of interest need to be specified according to numerical simulation or emerging technologies[56] in order to determine which boundaries are relevant to samples and to monitor or assess the MP flux at each boundary. For a river MP boundary, at a minimum, this should include the MP flux in upstream water, the effluent from WWTPs,[16,54] and when exiting the river. This further validation will help to confirm the accuracy of our framework in identifying the spatial dependence of riverine MPs on wastewater discharge. Certainly, other boundaries that could potentially serve as sources or sinks should also be taken into consideration, including riverbeds and riverbanks, other surface water media, and atmospheric media.[5]

■ ASSOCIATED CONTENT

Data Availability Statement

The data sets generated for and/or analyzed during the current study are available from the author on reasonable request. Further information about the data and the conditions for access can be provided upon request.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.4c02255.

Detailed information on the spatial relationship between wastewater discharge and riverine MP flux within different upstream boundaries in the hybrid data set and the distance of the sampling site to the nearest city (PDF)

Detailed information on the sampling methods for MPs in surface water of the Yangtze River mainstream from literature search, statistics of flux corresponding to pure and hybrid data sets, wastewater discharge data within different upstream boundaries, statistical trends in the slope and intercept of linear equations, and statistical data of wastewater discharge volumes under different river discharge conditions (XLSX)

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Notes

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■ REFERENCES

(1) Rochman, C. M. Microplastics research—from sink to source. *Science* **2018**, *360* (6384), 28−29.

(2) Thompson, R. C.; Olsen, Y.; Mitchell, R. P.; Davis, A.; Rowland, S. J.; John, A. W. G.; McGonigle, D.; Russell, A. E. Lost at sea: Where is all the plastic? *Science* **2004**, *304* (5672), 838.

(3) Wagner, M.; Lambert, S. M. *Freshwater Microplastics: Emerging Environmental Contaminants?*; Springer: Cham, Switzerland, 2018.

(4) Lebreton, L. C. M.; van der Zwet, J.; Damsteeg, J. W.; Slat, B.; Andrady, A.; Reisser, J. River plastic emissions to the world's oceans. *Nat. Commun.* **2017**, *8*, 15611.

(5) van Emmerik, T. H. M.; Mellink, Y.; Hauk, R.; Waldschläger, K.; Schreyers, L. Rivers as plastic reservoirs. *Front. Water* **2022**, *3*, 786936.

(6) Wang, C.; Zhao, J.; Xing, B. Environmental source, fate, and toxicity of microplastics. *J. Hazard. Mater.* **2021**, *407*, 124357.

(7) Klein, S.; Worch, E.; Knepper, T. P. Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main area in Germany. *Environ. Sci. Technol.* **2015**, *49* (10), 6070−6076.

(8) Best, J. Anthropogenic stresses on the world's big rivers. *Nat. Geosci.* **2019**, *12*, 7−21.

(9) Cowger, W.; Gray, A. B.; Schultz, R. C. Anthropogenic litter cleanups in Iowa riparian areas reveal the importance of near-stream and watershed scale land use. *Environ. Pollut.* **2019**, *250*, 981−989.

(10) Talbot, R.; Chang, H. Microplastics in freshwater: A global review of factors affecting spatial and temporal variations. *Environ. Pollut.* **2022**, *292*, 118393.

(11) Park, H. J.; Oh, M. J.; Kim, P. G.; Kim, G.; Jeong, D. H.; Ju, B. K.; Lee, W. S.; Chung, H. M.; Kang, H. J.; Kwon, J. H. National reconnaissance survey of microplastics in municipal wastewater treatment plants in Korea. *Environ. Sci. Technol.* **2020**, *54* (3), 1503−1512.

(12) Murphy, F.; Ewins, C.; Carbonnier, F.; Quinn, B. Wastewater treatment works (WwTW) as a source of microplastics in the aquatic environment. *Environ. Sci. Technol.* **2016**, *50* (11), 5800−5808.

(13) Rochman, C. M.; Munno, K.; Box, C.; Cummins, A.; Zhu, X.; Sutton, R. Think global, act local: Local knowledge is critical to inform positive change when it comes to microplastics. *Environ. Sci. Technol.* **2021**, *55* (1), 4−6.

(14) Woodward, J.; Li, J.; Rothwell, J.; Hurley, R. Acute riverine microplastic contamination due to avoidable releases of untreated wastewater. *Nat. Sustain.* **2021**, *4* (9), 793−802.

(15) Wu, X. W.; Zhao, X. L.; Chen, R. Z.; Liu, P.; Liang, W. G.; Wang, J. Y.; Teng, M. M.; Wang, X.; Gao, S. X. Wastewater treatment plants act as essential sources of microplastic formation in aquatic environments: A critical review. *Water Res.* **2022**, *221*, 118825.

(16) Ziajahromi, S.; Neale, P. A.; Rintoul, L.; Leusch, F. D. L. Wastewater treatment plants as a pathway for microplastics: Development of a new approach to sample wastewater-based microplastics. *Water Res.* **2017**, *112*, 93−99.

(17) Baldwin, A. K.; Corsi, S. R.; Mason, S. A. Plastic debris in 29 Great Lakes tributaries: Relations to watershed attributes and hydrology. *Environ. Sci. Technol.* **2016**, *50* (19), 10377−10385.

(18) Cowger, W.; et al. Evaluating wastewater effluent as a source of microplastics in environmental samples. In *Microplastics in Water and Wastewater*; Karapanagioti, H. K., Kalavrouziotis, I. K., Eds.; IWA Publishing: London, 2019; pp 109−131.

(19) He, Y.; Lu, J.; Li, C.; Wang, X.; Jiang, C.; Zhu, L.; Bu, X.; Jabeen, K.; Vo, T. T.; Li, D. From pollution to solutions: Insights into the sources, transport and management of plastic debris in pristine and urban rivers. *Environ. Res.* **2024**, *245*, 118024.

(20) Rico, A.; Redondo-Hasselerharm, P. E.; Vighi, M.; Waichman, A. V.; Nunes, G. S. d.S.; de Oliveira, R.; Singdahl-Larsen, C.; Hurley, R.; Nizzetto, L.; Schell, T. Large-scale monitoring and risk assessment of microplastics in the Amazon River. *Water Res.* **2023**, *232*, 119707.

(21) Yuan, W. K.; Christie-Oleza, J. A.; Xu, E. G.; Li, J. W.; Zhang, H. B.; Wang, W. F.; Lin, L.; Zhang, W. H.; Yang, Y. Y. Environmental fate of microplastics in the world's third-largest river: Basin-wide investigation and microplastic community analysis. *Water Res.* **2022**, *210*, 118002.

(22) Newbould, R. Understanding river plastic transport with tracers and GPS. *Nat. Rev. Earth Environ.* **2021**, *2* (9), 591.

(23) van Emmerik, T. H. M.; Schwarz, A. Plastic debris in rivers. *WIREs Water* **2019**, *7*, No. e1398.

(24) Barrows, A. P. W.; Christiansen, K. S.; Bode, E. T.; Hoellein, T. J. A watershed-scale, citizen science approach to quantifying microplastic concentration in a mixed land-use river. *Water Res.* **2018**, *147*, 382−392.

(25) Mintenig, S. M.; Kooi, M.; Erich, M. W.; Primpke, S.; Redondo-Hasselerharm, P. E.; Dekker, S. C.; Koelmans, A. A.; van Wezel, A. P. A systems approach to understand microplastic occurrence and variability in Dutch riverine surface waters. *Water Res.* **2020**, *176*, 115723.

(26) Cowger, W.; Gray, A. B.; Guilinger, J. J.; Fong, B.; Waldschlager, K. Concentration depth profiles of microplastic particles in river flow and implications for surface sampling. *Environ. Sci. Technol.* **2021**, *55* (9), 6032−6041.

(27) Drummond, J. D.; Schneidewind, U.; Li, A.; Hoellein, T. J.; Krause, S.; Packman, A. I. Microplastic accumulation in riverbed sediment via hyporheic exchange from headwaters to mainstems. *Sci. Adv.* **2022**, *8* (2), No. eabi9305.

(28) Mennekes, D.; Nowack, B. Predicting microplastic masses in river networks with high spatial resolution at country level. *Nat. Water* **2023**, *1* (6), 523−533.

(29) van Emmerik, T. H. M.; Frings, R. M.; Schreyers, L. J.; Hauk, R.; de Lange, S. I.; Mellink, Y. A. M. River plastic transport and deposition amplified by extreme flood. *Nat. Water* **2023**, *1* (6), 514−522.

(30) Meijer, L. J. J.; van Emmerik, T. H. M.; van der Ent, R.; Schmidt, C.; Lebreton, L. More than 1000 rivers account for 80% of global riverine plastic emissions into the ocean. *Sci. Adv.* **2021**, *7* (18), No. eaaz5803.

(31) Huang, Q. E.; Liu, M. G.; Cao, X. W.; Liu, Z. Occurrence of microplastics pollution in the Yangtze River: Distinct characteristics of spatial distribution and basin-wide ecological risk assessment. *Water Res.* **2023**, *229*, 119431.

(32) Cowger, W.; Booth, A. M.; Hamilton, B. M.; Thaysen, C.; Primpke, S.; Munno, K.; Lusher, A. L.; Dehaut, A.; Vaz, V. P.; Liboiron, M.; Devriese, L. I.; Hermabessiere, L.; Rochman, C.; Athey, S. N.; Lynch, J. M.; De Frond, H.; Gray, A.; Jones, O. A. H.; Brander, S.; Steele, C.; Moore, S.; Sanchez, A.; Nel, H. Reporting guidelines to increase the reproducibility and comparability of research on microplastics. *Appl. Spectrosc.* **2020**, *74* (9), 1066−1077.

(33) Hurley, R.; Woodward, J.; Rothwell, J. J. Microplastic contamination of river beds significantly reduced by catchment-wide flooding. *Nat. Geosci.* **2018**, *11* (4), 251−257.

(34) Bai, M.; Lin, Y.; Hurley, R. R.; Zhu, L.; Li, D. Controlling factors of microplastic riverine flux and implications for reliable monitoring strategy. *Environ. Sci. Technol.* **2022**, *56* (1), 48−61.

(35) Eo, S.; Hong, S. H.; Song, Y. K.; Han, G. M.; Shim, W. J. Spatiotemporal distribution and annual load of microplastics in the Nakdong River, South Korea. *Water Res.* **2019**, *160*, 228−237.

(36) Lehner, B.; Grill, G. Global river hydrography and network routing: baseline data and new approaches to study the world's large river systems. *Hydrol. Process.* **2013**, *27* (15), 2171−2186.

(37) Ouellet Dallaire, C.; Lehner, B.; Sayre, R.; Thieme, M. A multidisciplinary framework to derive global river reach classifications at high spatial resolution. *Environ. Res. Lett.* **2019**, *14* (2), 024003.

(38) Ehalt Macedo, H.; Lehner, B.; Nicell, J.; Grill, G.; Li, J.; Limtong, A.; Shakya, R. Distribution and characteristics of wastewater treatment plants within the global river network. *Earth Syst. Sci. Data* **2022**, *14* (2), 559−577.

(39) Kim, J. H.; Lee, D. H.; Kang, J. H. Associating the spatial properties of a watershed with downstream Chl-a concentration using spatial analysis and generalized additive models. *Water Res.* **2019**, *154*, 387−401.

(40) Haining, R. P. Spatial Autocorrelation. In *International Encyclopedia of the Social & Behavioral Sciences*; Smelser, N. J., Baltes, P. B., Eds.; Pergamon: Oxford, 2001; pp 14763−14768.

(41) Su, Z.; Wang, K.; Yang, F.; Zhuang, T. Antibiotic pollution of the Yellow River in China and its relationship with dissolved organic matter: Distribution and source identification. *Water Res.* **2023**, *235*, 119867.

(42) Davis, R. B.; Thompson, J. E.; Pardue, H. L. Characteristics of statistical parameters used to interpret least-squares results. *Clin. Chem.* **1978**, *24* (4), 611−620.

(43) Qiu, Q.; Liang, Z.; Xu, Y.; Matsuzaki, S. i. S.; Komatsu, K.; Wagner, T. A statistical framework to track temporal dependence of chlorophyll−nutrient relationships with implications for lake eutrophication management. *J. Hydrol.* **2021**, *603*, 127134.

(44) Rineau, V.; Smyčka, J.; Storch, D. Diversity dependence is a ubiquitous phenomenon across Phanerozoic oceans. *Sci. Adv.* **2022**, *8* (43), No. eadd9620.

(45) Azen, R.; Budescu, D. V. The dominance analysis approach for comparing predictors in multiple regression. *Psychol. Methods* **2003**, *8* (2), 129−148.

(46) He, D.; Chen, X.; Zhao, W.; Zhu, Z.; Qi, X.; Zhou, L.; Chen, W.; Wan, C.; Li, D.; Zou, X.; Wu, N. Microplastics contamination in the surface water of the Yangtze River from upstream to estuary based on different sampling methods. *Environ. Int.* **2021**, *196*, 110908.

(47) Xu, D.; Gao, B.; Wan, X.; Peng, W.; Zhang, B. Influence of catastrophic flood on microplastics organization in surface water of the Three Gorges Reservoir, China. *Water Res.* **2022**, *211*, 118018.

(48) Nava, V.; Chandra, S.; Aherne, J.; Alfonso, M. B.; Antão-Geraldes, A. M.; Attermeyer, K.; Bao, R.; Bartrons, M.; Berger, S. A.; Biernaczyk, M.; Bissen, R.; Brookes, J. D.; Brown, D.; Cañedo-Argüelles, M.; Canle, M.; Capelli, C.; Carballeira, R.; Cereijo, J. L.; Chawchai, S.; Christensen, S. T.; Christoffersen, K. S.; de Eyto, E.; Delgado, J.; Dornan, T. N.; Doubek, J. P.; Dusaucy, J.; Erina, O.; Ersoy, Z.; Feuchtmayr, H.; Frezzotti, M. L.; Galafassi, S.; Gateuille, D.; Gonçalves, V.; Grossart, H.-P.; Hamilton, D. P.; Harris, T. D.; Kangur, K.; Kankılıç, G. B.; Kessler, R.; Kiel, C.; Krynak, E. M.; Leiva-Presa, A.-.; Lepori, F.; Matias, M. G.; Matsuzaki, S.-i.S.; McElarney, Y.; Messyasz, B.; Mitchell, M.; Mlambo, M. C.; Motitsoe, S. N.; Nandini, S.; Orlandi, V.; Owens, C.; Özkundakci, D.; Pinnow, S.; Pociecha, A.; Raposeiro, P. M.; Rõõm, E. I.; Rotta, F.; Salmaso, N.; Sarma, S. S. S.; Sartirana, D.; Scordo, F.; Sibomana, C.; Siewert, D.; Stepanowska, K.; Tavşanoğlu, Ü. N.; Tereshina, M.; Thompson, J.; Tolotti, M.; Valois, A.; Verburg, P.; Welsh, B.; Wesolek, B.; Weyhenmeyer, G. A.; Wu, N.; Zawisza, E.; Zink, L.; Leoni, B. Plastic debris in lakes and reservoirs. *Nature* **2023**, *619* (7969), 317−322.

(49) Li, R.; Zhu, L.; Wang, Y.; Zhu, Y. G. Metagenomic insights into environmental risk of field microplastics in an urban river. *Water Res.* **2022**, *223*, 119018.

(50) Wagner, S.; Klöckner, P.; Stier, B.; Römer, M.; Seiwert, B.; Reemtsma, T.; Schmidt, C. Relationship between discharge and river plastic concentrations in a rural and an urban catchment. *Environ. Sci. Technol.* **2019**, *53* (17), 10082−10091.

(51) Zhou, Y.; Li, Y.; Yan, Z.; Wang, H.; Chen, H.; Zhao, S.; Zhong, N.; Cheng, Y.; Acharya, K. Microplastics discharged from urban drainage system: Prominent contribution of sewer overflow pollution. *Water Res.* **2023**, *236*, 119976.

(52) Alexander, R. B.; Smith, R. A.; Schwarz, G. E. Effect of stream channel size on the delivery of nitrogen to the Gulf of Mexico. *Nature* **2000**, *403* (6771), 758−761.

(53) Tong, Y.; Qi, M.; Sun, P.; Qin, W.; Zhu, Y.; Wang, X.; Xu, Y.; Zhang, W.; Yang, J. Estimation of unintended treated wastewater contributions to streams in the Yangtze River Basin and the potential human health and ecological risk analysis. *Environ. Sci. Technol.* **2022**, *56* (9), 5590−5601.

(54) Chen, H.; Jia, Q.; Sun, X.; Zhou, X.; Zhu, Y.; Guo, Y.; Ye, J. Quantifying microplastic stocks and flows in the urban agglomeration based on the mass balance model and source-pathway-receptor framework: Revealing the role of pollution sources, weather patterns, and environmental management practices. *Water Res.* **2022**, *224*, 119045.

(55) Frehland, S.; Kaegi, R.; Hufenus, R.; Mitrano, D. M. Long-term assessment of nanoplastic particle and microplastic fiber flux through a pilot wastewater treatment plant using metal-doped plastics. *Water Res.* **2020**, *182*, 115860.

(56) Cook, S.; Chan, H. L.; Abolfathi, S.; Bending, G. D.; Schäfer, H.; Pearson, J. M. Longitudinal dispersion of microplastics in aquatic flows using fluorometric techniques. *Water Res.* **2020**, *170*, 115337.