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Antibiotic Source Apportionment in Stream Sediments Using PMF with Combined Antibiotic and Heavy Metal(loid) Profiles

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Abstract

Source attribution of antibiotics in stream sediments remains challenging because similar compounds can originate from multiple sectors and their environmental transformation may alter source-specific compositional patterns after release. This study evaluated whether heavy metal(loid)s could provide complementary information for PMF-based antibiotic source apportionment. Sediment samples from 12 sites in the Muhan Stream watershed were analyzed for 11 antibiotics and 10 heavy metal(loid)s, of which 9 antibiotics and all 10 heavy metal(loid)s were detected. A PMF model based on the 9 detected antibiotics was directly compared with an integrated model incorporating both antibiotics and heavy metal(loid)s. The antibiotic-based model produced a five-factor solution, but several site-specific factor contributions were not fully consistent with the spatial distribution of known pollution sources.

The integrated model retained comparable diagnostic performance and stability while providing additional multielement contrasts that reduced ambiguity among source-related factors. In particular, factors associated with the wastewater treatment plant, livestock manure treatment facility, and livestock farms showed more distinct site-specific contribution patterns and greater spatial coherence with known source distributions. These results indicate that heavy metal(loid)s can complement antibiotic profiles and support clearer PMF-based source interpretation. Factor-attributed screening analysis further indicated that manure-related factors accounted for relatively large shares of the estimated ecological risk. This approach may provide a practical basis for source-oriented pollution management in watersheds affected by multiple pollution sources.

Keywords: Emerging contaminants; Positive matrix factorization; Receptor modeling; Risk assessment; Environmental management

1. Introduction

The widespread use of antibiotics in human medicine and livestock production has raised concerns about their release and persistence in the environment (Aladekoyi et al., 2024). Antibiotics that are not fully metabolized in the body can enter the environment through domestic wastewater and livestock manure, eventually reaching aquatic ecosystems such as rivers and streams (Khalil et al., 2023). Although antibiotic residues are generally detected at lower concentrations in aquatic environments than in soils (Jia et al., 2023), continuous inputs can lead to long-term contamination and promote the emergence and spread of antibiotic-resistant bacteria and antibiotic resistance genes (Zhang et al., 2024b). Antibiotic pollution in

aquatic environments is an important concern for ecosystems and public health, and its effective management requires reliable identification of pollution sources.

After entering streams, antibiotics may remain dissolved in the water column or sorb to suspended particles and organic matter before accumulating in sediments (Dong et al., 2022). Dissolved antibiotics are sensitive to seasonal and hydrological variability, and their concentrations may vary substantially with streamflow and rainfall (Cao et al., 2024). In contrast, sediments can retain antibiotic residues over longer periods and are less affected by short-term fluctuations (Chen et al., 2022). Therefore, antibiotic concentrations and compositional patterns in sediments can reflect cumulative inputs within a watershed and provide useful information for contamination assessment and source tracing.

Positive matrix factorization (PMF) has been widely used for source apportionment of contaminants in aquatic environments (Liu et al., 2025a; Miao et al., 2025) and has increasingly been applied to antibiotic pollution profiles (Pang et al., 2025; Wang et al., 2023; Zhang et al., 2020). However, in antibiotic source apportionment, distinguishing individual sources from antibiotic profiles alone can be difficult. The same antibiotics can be used across different sectors, including human medicine and livestock production, while degradation, sorption, transformation, and transport can further alter their original compositional patterns after release (Gothwal and Shashidhar, 2015). These processes can obscure source-specific signatures and complicate the attribution of PMF-derived factors to specific pollution sources. Accordingly, antibiotic-based PMF source assignments may not always correspond closely to known anthropogenic activities within a watershed (Zhang et al., 2025).

To address such uncertainty in PMF-based source interpretation, complementary tracers and independent analytical approaches have increasingly been incorporated into source

apportionment studies. Stable isotope analysis has been combined with PMF to provide complementary evidence for heavy metal(loid) source attribution in sediments (Xi et al., 2026). Machine-learning approaches have also been integrated with PMF to refine source discrimination and interpretation (Zhang et al., 2024a; Zhou et al., 2025). Similarly, fluorescence characteristics of organic matter have been used together with PMF to provide complementary evidence for pollution-source identification (Wang et al., 2022). Such complementary approaches have also been applied to antibiotic source apportionment in watersheds affected by multiple pollution sources, providing additional support for PMF-based source interpretation (Xu et al., 2025). In many of these approaches, complementary information has been used outside the PMF input matrix to support the interpretation of factors after they have been resolved. Incorporating source-related variables directly into the input matrix would instead allow this information to contribute to factor resolution itself. In particular, variables from a different contaminant group may provide source-related contrasts that are not captured by antibiotic profiles alone, potentially providing additional contrasts for factor resolution and subsequent source interpretation.

Among the potential variables that may complement antibiotic profiles, heavy metal(loid)s are potentially useful because they can originate from many of the same anthropogenic sources, including livestock manure, manure-derived fertilizers, agricultural inputs, and wastewater (Liu et al., 2025b; Shen et al., 2023; Wang et al., 2024). Unlike antibiotics, which can undergo degradation and transformation after release, heavy metal(loid)s can persist in sediments, while different source types can exhibit distinct heavy metal(loid) composition profiles (Li et al., 2020). Together, these characteristics may provide source-related information that complements antibiotic profiles and supports source interpretation. Previous antibiotic studies have evaluated heavy metal(loid)s alongside PMF-based source apportionment, using

correlation or multivariate analyses to examine their associations with antibiotic occurrence and environmental factors (Mu et al., 2024; Zhang et al., 2020). However, heavy metal(loid)s have generally been evaluated separately from the antibiotic concentration matrix used for PMF rather than incorporated as PMF input variables. It therefore remains unclear whether jointly incorporating antibiotic and heavy metal(loid) concentration data into PMF can provide complementary information for antibiotic source apportionment.

This study evaluated whether incorporating heavy metal(loid) information into the existing PMF framework could provide complementary information for antibiotic source apportionment in stream sediments. The antibiotic-based and integrated models were compared in terms of model diagnostics, factor profiles, reproducibility, and the spatial correspondence between site-specific factor contributions and known potential pollution sources. Particular attention was given to whether the additional multielement information produced factor contribution patterns that were more consistent with the spatial distribution of known sources and reduced ambiguity among source-related factors.

2. Materials and methods

2.1. Chemicals and reagents

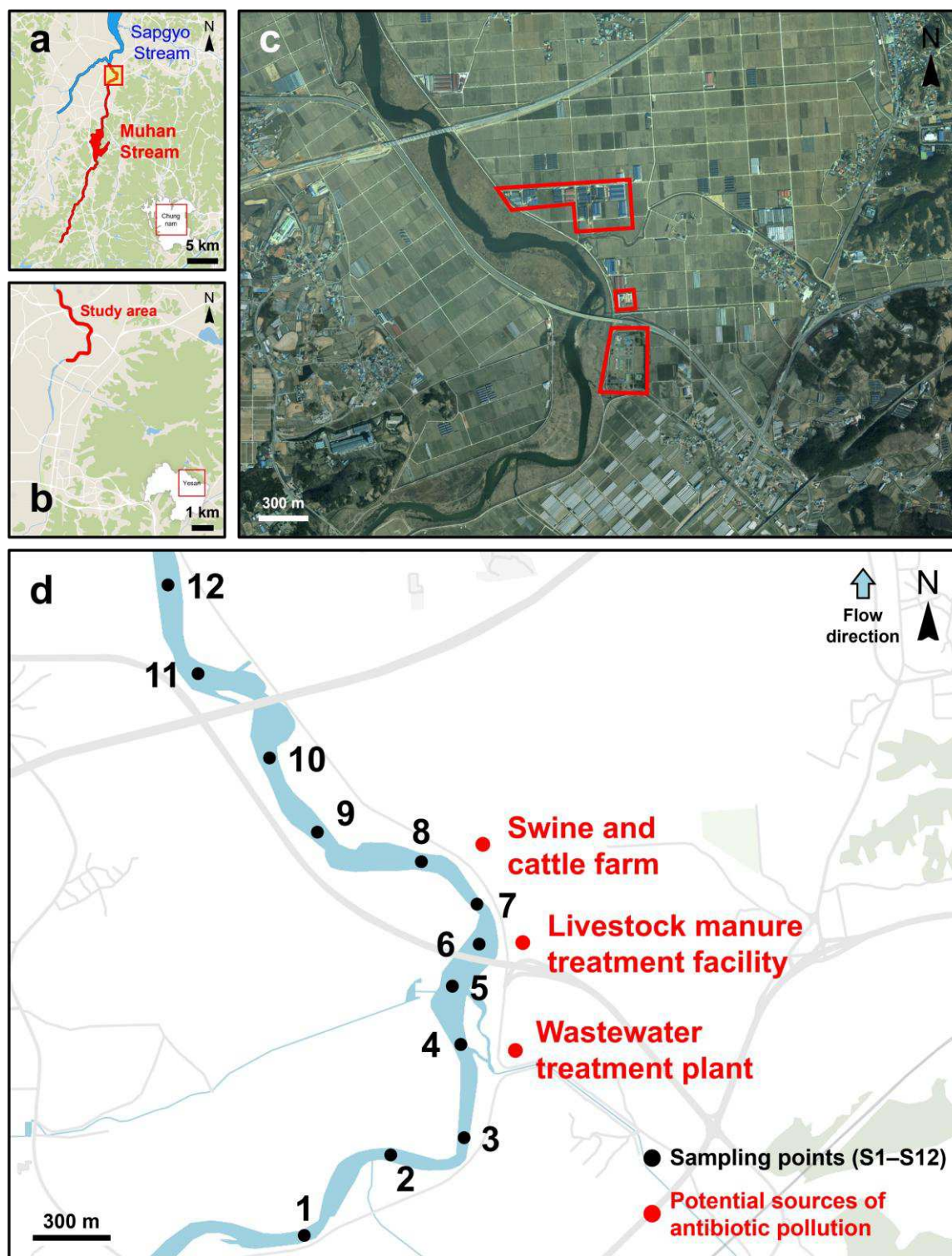
Antibiotics from four classes, namely penicillins (PNs), tetracyclines (TCs), macrolides (MLs), and sulfonamides (SAs), were selected for analysis in this study. These classes were chosen to include major antibiotic classes used in South Korea while encompassing compounds with different physicochemical properties and environmental fate and behavior. The target compounds were ampicillin (AMP) and penicillin G (PNG) for penicillins; chlortetracycline (CTC), oxytetracycline (OTC), and tetracycline (TC) for tetracyclines; spiramycin (SPM) and

tylosin (TYL) for macrolides; and sulfadiazine (SDZ), sulfamethazine (SMZ), sulfamethoxazole (SMX), and sulfathiazole (STZ) for sulfonamides. These compounds were selected based on their sales and use patterns in South Korea, together with their relevance to the major potential sources and input pathways in the study area, including livestock activities, agricultural runoff, and domestic wastewater. The physicochemical properties of the target antibiotics are summarized in Table S1, and their sales volumes in South Korea are provided in Table S2. Analytical standards of the target antibiotics were purchased from Sigma-Aldrich (St. Louis, MO, USA). All other reagents and solvents were purchased from Samchun Pure Chemical Co., Ltd. (Pyeongtaek, South Korea) and J. T. Baker (Phillipsburg, NJ, USA).

2.2. Sample collection

Muhan Stream was selected because multiple potential antibiotic sources, including agricultural drainage, wastewater discharges, and livestock activities, co-occur within the watershed. In this study, stream sediment samples were collected from 12 sites along the lower reach of Muhan Stream in May 2023 (Fig. 1). Geographic information and sediment chemical properties for each sampling point are provided in Table S3. The study area contains numerous agricultural drainage channels and sewer lines, and the public wastewater treatment plant (WWTP) and the public livestock manure treatment facility (LMTF) are located nearby, indicating a high potential for residual antibiotic inputs. In addition, cattle and swine farms are distributed throughout the watershed, suggesting possible influence from livestock-derived antibiotic pollution. To support the interpretation of the PMF results, environmental samples associated with major antibiotic pollution sources were collected near the study area, including livestock manure (cattle, swine, and poultry; n = 3 each), livestock manure-based compost (n

138 = 4) and liquid manure-based fertilizer (n = 5), arable soils (n = 6), and effluents from the
 139 WWTP (n = 1) and LMTF (n = 1).



140 **Fig. 1.** Map of the Muhan Stream study area and sampling points. (a) Location of Muhan

Stream in Chungcheongnam-do, South Korea. The study area, located in the lowermost reach of Muhan Stream near its confluence with Sapgyo Stream, is indicated by the red outline. (b) Enlarged map of the study area in Yesan-gun, with the study reach of Muhan Stream indicated by a red line. (c) Satellite image showing surrounding agricultural fields, urban areas, and Muhan Stream, with red outlines indicating the actual locations of point-source facilities. (d) Sampling points S1–S12 and nearby potential sources of antibiotic pollution, including livestock farms, a livestock manure treatment facility (LMTF), and a wastewater treatment plant (WWTP).

2.3. Sample pretreatment and instrumental analysis

The overall procedure used for the analysis of residual antibiotics in the collected samples is presented in Fig. S1. Briefly, 1.0 g of freeze-dried and sieved sediment was extracted twice with 10 mL of McIlvaine buffer (pH 4.0), 10 mL of methanol, and 250 μ L of 5% (w v⁻¹) Na₂-EDTA solution by shaking for 15 min, followed by centrifugation at 4,000 rpm for 15 min. The combined extract was diluted to a final volume of 120 mL with HPLC-grade water and vacuum-filtered. The filtrate was then purified using an Oasis hydrophilic-lipophilic balance (HLB) cartridge (3 cc, 60 mg), followed by elution of antibiotics with 5.0 mL of methanol. The resulting extract was concentrated to 0.05 mL, mixed with 0.07 mL of HPLC-grade water, and stored at –20°C until instrumental analysis. The 11 target antibiotics were analyzed using high-performance liquid chromatography (Agilent 1290 Infinity II, Agilent, Santa Clara, CA, USA) coupled with a triple quadrupole mass spectrometer (6500 QTRAP, SCIEX, Framingham, MA, USA) equipped with an electrospray ionization (ESI) source. All antibiotics were analyzed in positive ion mode using multiple reaction monitoring (MRM). Detailed pretreatment

procedures for the other samples are described in Text S1, whereas the HPLC and mass spectrometric conditions and method validation results are presented in Table S4, Table S5, and Table S6.

The target heavy metal(loid)s were selected to capture potential pollution source signals within the watershed. A total of ten heavy metal(loid)s, including As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V, and Zn, were analyzed. For the analysis of heavy metal(loid) concentrations, the collected samples were pretreated using pseudo-total digestion with aqua regia and then analyzed using inductively coupled plasma optical emission spectrometry (ICP-OES, iCAP PRO X Duo, Thermo Fisher Scientific, USA). Detailed analytical conditions and method validation results are summarized in Table S7. The enrichment factor (EF) and geo-accumulation index (I_{geo}) were calculated to evaluate the level of heavy metal(loid) contamination in the sediments (Text S2).

2.4. PMF analysis

The U.S. Environmental Protection Agency (EPA) PMF software (version 5.0) was applied to resolve source-related factors associated with antibiotic contamination in Muhan Stream sediments and estimate their relative contributions. Concentration data for the detected antibiotics and heavy metal(loid)s in sediment samples from the 12 sites in the Muhan Stream watershed were used as input data for PMF analysis. Modeling was performed under two conditions. The first was an antibiotic-based model based solely on antibiotic concentration data, and the second was an integrated model using both antibiotic and heavy metal(loid) concentration data. Concentration and uncertainty data for each variable were used as PMF input values, and values below the detection limit were treated according to the PMF input

criteria. Detailed procedures for PMF input preparation and uncertainty estimation are provided in Text S3. Solution stability and rotational ambiguity were further evaluated using Q values, DISP, bootstrap, and Fpeak analyses, while the reproducibility of factor profiles and site-specific factor contributions was assessed across repeated random-seed runs. Detailed diagnostic procedures are provided in Texts S5 and S6. Following model selection, factor-specific contaminant profiles, site-specific contributions, and known source locations were jointly considered to identify the most plausible source-related influence represented by each factor. Factor labels should therefore be interpreted as source-related influences rather than mutually exclusive source categories. Because source-location information contributed to factor interpretation, spatial correspondence was used to assess the coherence of the source-related interpretations rather than as an independent measure of source-apportionment accuracy.

2.5. Ecological risk assessment and PMF-based factor attribution

The potential ecological risks of antibiotics in sediments were evaluated using risk quotients (RQs), calculated as the ratio of the measured environmental concentration (MEC) to the predicted no-effect concentration ($PNEC$). Because chronic toxicity data for benthic organisms were limited, sediment $PNECs$ were estimated from water-based toxicity data using the equilibrium partitioning approach (Voznakova et al., 2025). Water-based $PNECs$ were derived using either species sensitivity distributions or deterministic assessment-factor methods, depending on the availability of chronic toxicity data. An $RQ \geq 1$ was considered to indicate potential ecological risk, whereas an $RQ < 1$ indicated low ecological concern in this screening-level assessment. Detailed procedures for toxicity-data selection, $PNEC$ derivation, assessment factors, and equilibrium partitioning are provided in Text S4 and Tables S8 and S9.

To evaluate the relative distribution of ecological risk among the source-related factors resolved by PMF, antibiotic-specific RQ s were allocated according to the site-specific factor contributions and factor-specific antibiotic composition. The resulting factor-attributed risk quotients (RQ_{source}) were aggregated across the 12 sampling sites and expressed as relative contributions among the five factors. These values were interpreted as factor-attributed screening indicators conditional on the PMF factor interpretation, rather than as independently validated estimates of source-specific ecological risk. Detailed calculations are provided in Text S4.

$$RQ_{source} = F \sum (RQ \times G) \quad (\text{Eq. 1})$$

2.6. Statistical analysis

Statistical analyses were performed using the Statistical Package for the Social Sciences (SPSS version 26.0, SPSS Inc., Chicago, IL, USA). All analytical measurements were conducted in triplicate, and the mean and standard deviation were calculated. Correlations between variables were evaluated using Spearman's correlation analysis, with statistical significance set at $p < .05$. Site-specific factor contributions estimated by the PMF model were visualized spatially using QGIS version 3.20. The spatial distribution of each factor contribution was interpolated using inverse distance weighting (IDW), and source-related contribution maps were generated based on the interpolation results.

3. Results and discussion

3.1. Occurrence and spatial distribution of antibiotics in sediments

Sediments from the 12 sites in the Muhan Stream watershed contained detectable residues of multiple antibiotic classes (Fig. 2, Table S10). PNs (AMP and PNG) were not detected at any site, whereas TCs, MLs, and SAs were detected at all sites, with concentrations ranging from 0.01 to 5.76 $\mu\text{g kg}^{-1}$. Among the detected classes, TCs showed the highest mean concentration across individual compounds and sampling sites ($1.12 \pm 1.43 \mu\text{g kg}^{-1}$), with CTC being the most abundant compound overall ($2.39 \pm 1.75 \mu\text{g kg}^{-1}$). This predominance may be attributed to the extensive use of CTC in livestock production and the strong sediment affinity of TCs mediated by clay minerals, organic matter, and multivalent cations such as Ca^{2+} and Mg^{2+} (Conde-Cid et al., 2019; Zhang et al., 2011). In contrast, MLs and SAs occurred at lower mean concentrations of $0.33 \pm 0.50 \mu\text{g kg}^{-1}$ and $0.45 \pm 0.56 \mu\text{g kg}^{-1}$, respectively. The comparatively low concentrations of SAs in the sediments may partly reflect their pH-dependent speciation, which can reduce sediment retention and enhance mobility in aquatic environments (Hu et al., 2019; Straub, 2016). The non-detection of PNs is also consistent with their limited environmental persistence due to the hydrolysis of the β -lactam ring (Ana et al., 2021; Braschi et al., 2013).

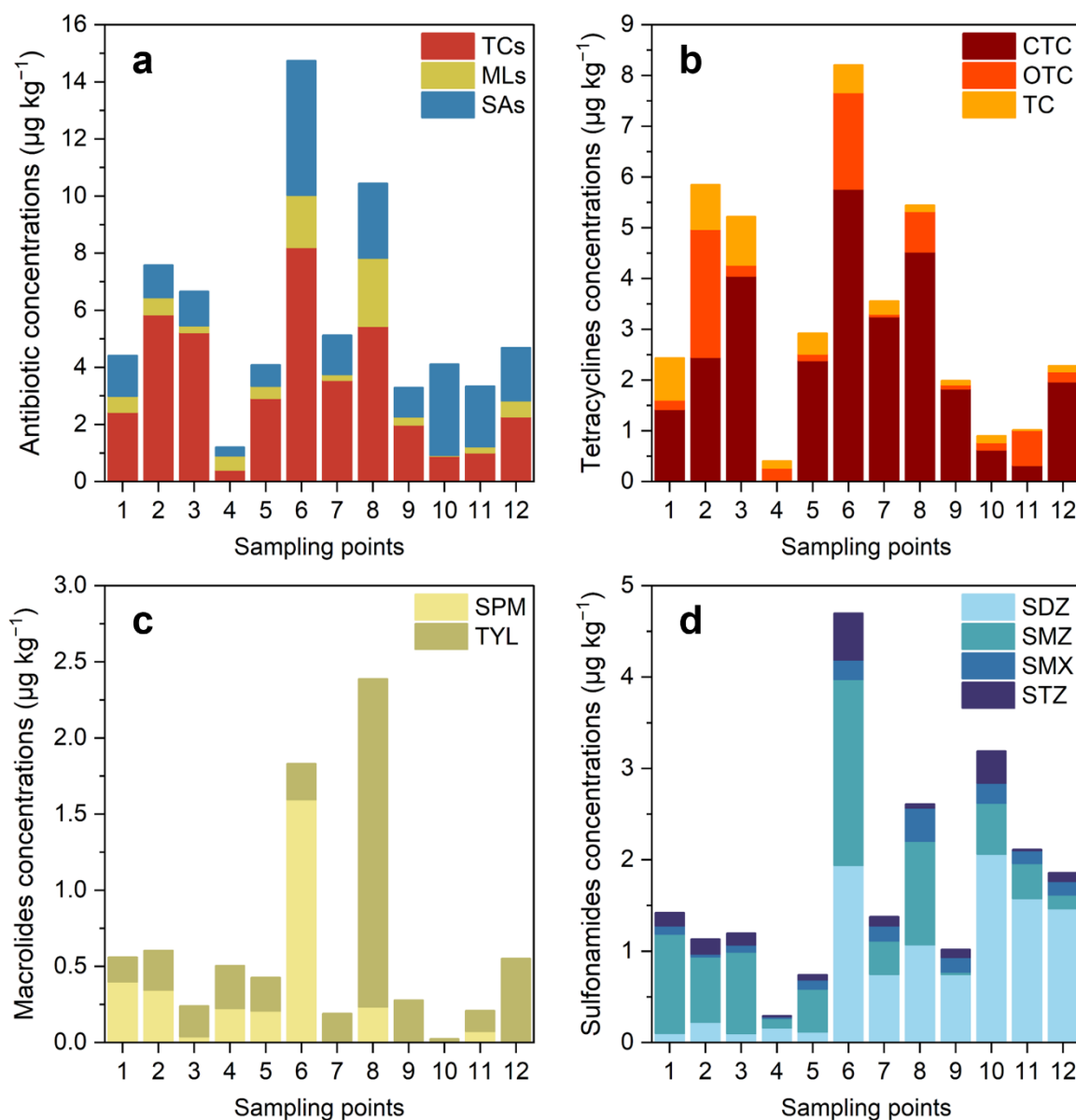


Fig. 2. Antibiotic concentrations in sediments from Muhan Stream. (a) total antibiotics, (b) tetracyclines, (c) macrolides, and (d) sulfonamides. Penicillins were excluded from the plots because they were not detected in the sediment samples.

The spatial distribution of antibiotics indicated localized source influences across the watershed. High coefficients of variation were observed for SPM (167.5%), TYL (147.2%), OTC (134.3%), and STZ (105.2%), suggesting strong site-specific inputs (Table S10). Relatively

high concentrations of SPM and TYL were observed at S6 near the LMTF and S8 near livestock farming areas, respectively, where total antibiotic concentrations were also highest. Livestock-related discharges are known to introduce antibiotics into aquatic environments (Kim and Carlson, 2007; Li et al., 2023; Li et al., 2016). These results suggest that the antibiotic hotspots at S6 and S8 were consistent with localized livestock-related inputs. By contrast, the lowest total antibiotic concentration was observed at S4 near the WWTP. CTC showed a relatively lower CV despite its high concentration, indicating a more widespread and relatively even spatial distribution across the sampled sites.

Antibiotic concentrations detected in Muhan Stream sediments were generally comparable to or lower than those reported in river sediments from China (Table S11), where antibiotic consumption has been reported to be high (Zhang et al., 2015). TCs in the Yangtze (Chen et al., 2022) and Huangpu rivers (Chen and Zhou, 2014) reached 18.18 and 21.7 $\mu\text{g kg}^{-1}$, respectively, while higher concentrations were reported in the Xiaoqing (Dong et al., 2022) and Chaobai rivers (Zhang et al., 2020), with CTC reaching 51.10 and 89.8 $\mu\text{g kg}^{-1}$, respectively. SAs were also detected at higher concentrations than those observed in Muhan Stream, with the highest level among the compared rivers reported in Xiong'an, where SMZ reached 5.77 $\mu\text{g kg}^{-1}$ (Fu et al., 2022). Higher SA concentrations were also reported in the Xiaoqing, Huangpu, and Chaobai rivers. MLs were reported only in Xiong'an and the Chaobai River, where they occurred at relatively low concentrations. Overall, these studies indicate that antibiotic contamination in sediments is influenced by livestock and anthropogenic activities, with the magnitude and composition of residues varying among antibiotic classes.

3.2. Antibiotic-based PMF source apportionment

PMF analysis was performed using antibiotic concentrations to identify potential sources and estimate their relative contributions in Muhan Stream sediments. The three- to five-factor solutions were evaluated based on model fit, DISP, bootstrap results, and source interpretability (Text S5 and Table S12). All three solutions showed generally stable diagnostic performance based on Q , DISP, and bootstrap analyses. However, in the three- and four-factor solutions, some PMF factors showed high estimated contributions across sites associated with different types of known pollution sources, making it difficult to attribute each factor to a single source. This ambiguity was reduced in the five-factor solution, allowing clearer source interpretation. Based on these comparisons, the five-factor solution was further evaluated across random-seed runs, which generally supported the reproducibility of factor profiles and site-specific contributions (Table S13). Therefore, the five-factor solution was selected as the final solution, and the corresponding factor profiles are shown in Fig. 3a.

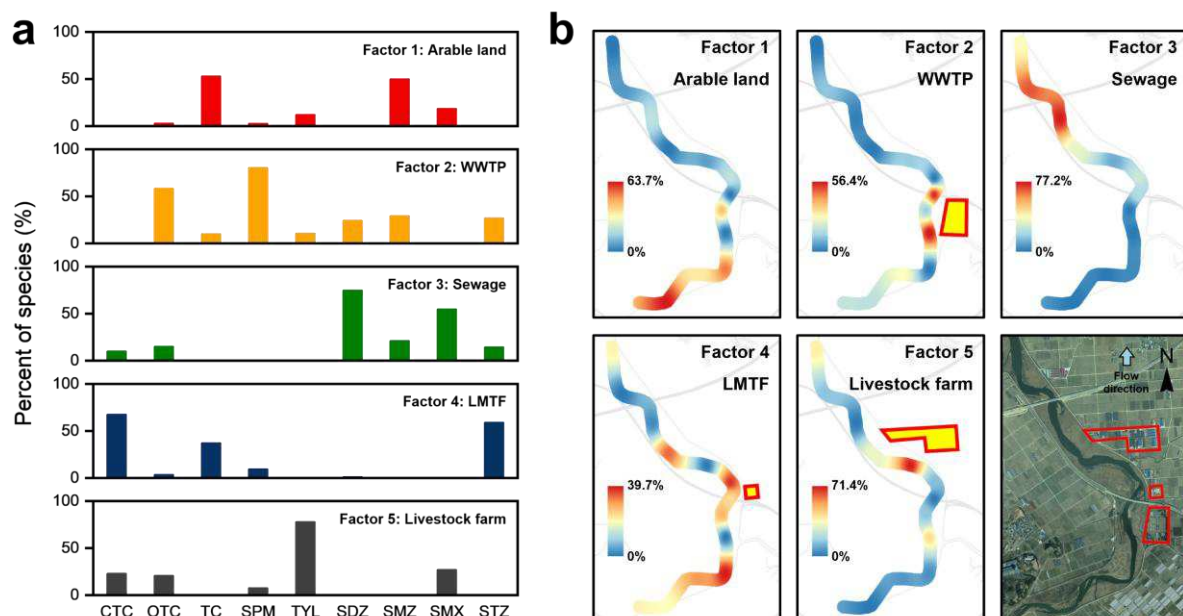


Fig. 3. Results of the antibiotic-based PMF model for sediments from Muhan Stream. (a) PMF

factor profiles for five factors interpreted in relation to arable land, wastewater treatment plant (WWTP), sewage, livestock manure treatment facility (LMTF), and livestock farm. (b) Maps generated by applying inverse distance weighting (IDW) interpolation to the site-specific factor contributions. The locations or areas of the corresponding potential pollution sources for Factors 2, 4, and 5 are marked with red-outlined boxes. The color scale for each factor map was independently scaled to the maximum site-specific contribution observed for that factor.

Factor 1 was characterized by high contributions of TC (53.7%) and SMZ (49.6%). SMZ introduced into agricultural soils through manure-derived amendments can be readily transported by surface runoff or drainage because of its relatively weak sorption (Pavlović et al., 2014; Wegst-Uhrich et al., 2014). In contrast, recent veterinary sales records in South Korea report no TC sales since 2009 (Park et al., 2025), suggesting that TC may originate from the environmental transformation of CTC and subsequent transport with soil particles (Chen et al., 2012a; Werner et al., 2009). Factor 1 was therefore interpreted as arable land runoff, representing antibiotics introduced into soils through compost, liquid fertilizer, or irrigation water and subsequently transported to the stream.

Factor 2 was dominated by SPM (80.4%) and OTC (58.5%). SPM has been frequently detected in WWTP effluents and shows limited removal during wastewater treatment (Calza et al., 2010; Lofrano et al., 2018). Similarly, OTC has been repeatedly detected in WWTP effluents and receiving streams in previous studies (Kim et al., 2016; Rodriguez-Mozaz et al., 2020). Although both compounds are also used for veterinary purposes, the relatively high contribution of Factor 2 near the WWTP supported its tentative interpretation as a WWTP-related source.

Factor 3 showed high contributions of SDZ (75.0%) and SMX (54.7%). Because SDZ and SMX are used in both human and veterinary medicine, these compounds alone could not clearly identify the source. However, Factor 3 was dominated by SAs, unlike the mixed antibiotic profile of Factor 2. Profiles dominated by SAs have been associated with untreated domestic wastewater (Zhi et al., 2023), which can enter streams through sewer leakage, direct discharge, or combined sewer overflows (Obradović et al., 2023; Ryu et al., 2014). Factor 3 was therefore interpreted as untreated domestic wastewater.

Factor 4 was characterized by high contributions of CTC (67.0%) and STZ (59.0%), both of which are strongly associated with veterinary use and livestock manure (Chen et al., 2012b; Huang et al., 2022). These antibiotics can be released from livestock manure treatment and storage systems (Kim et al., 2020), and their concentrations were relatively high near the LMTF in this study. Together, these findings and the high contribution of Factor 4 at this site indicated that Factor 4 represented an LMTF-related input.

Factor 5 was dominated by TYL (77.8%), an antibiotic used exclusively for veterinary purposes and particularly associated with swine production (Table S2). The highest TYL concentration occurred near livestock farms, consistent with the spatial distribution of Factor 5. As noted for Factor 4, the contribution of CTC also supported the association of Factor 5 with livestock sources. Factor 5 was therefore interpreted as runoff from livestock farming areas.

The PMF-derived profiles allowed relatively clear interpretation of the five factors, but their spatial contribution patterns were not always consistent with the known source locations (Fig. 3b and Table S14). Factor 2 was tentatively interpreted as a WWTP-related source based on the dominance of SPM and OTC and its relatively high contribution near the WWTP. However, Factor 2 showed similarly high site-specific contributions at S4 near the WWTP (56.4%) and

S6 near the LMTF (53.9%), making the WWTP-related interpretation difficult to distinguish spatially from the LMTF-related influence. Factors 4 and 5 were associated with the LMTF and livestock farming areas, respectively, although their contributions were also high at sites not clearly associated with these sources. These mismatches indicate a key limitation of relying only on antibiotic profiles for source apportionment in a watershed containing multiple distinct pollution source types. Many antibiotics are not source-specific because they can originate from both human and veterinary use and may also enter agricultural environments (Lim et al., 2017), while a single source can release multiple antibiotic classes simultaneously (Spielmeyer, 2018; Yang et al., 2021). In addition, differences in sorption, degradation, and transport behavior can alter antibiotic composition after release, causing the sediment profile to differ from the original source signal (Chabilan et al., 2023; Ding et al., 2021; Pastor-López et al., 2025). Therefore, the spatial inconsistency between PMF contributions and known source locations indicates the need for additional source indicators to improve factor interpretation.

3.3. Evaluation of heavy metal(loid)s as complementary source indicators

To evaluate whether heavy metal(loid)s could provide information complementary to antibiotic profiles, ten elements (As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V, and Zn) were analyzed in stream sediments and potential pollution source samples. The source samples comprised cattle, swine, and poultry manure, compost, liquid fertilizer, soil, WWTP effluent, and LMTF effluent, and both antibiotics and heavy metal(loid)s were detected across these source categories (Tables S16 and S17). Heavy metal(loid)s occurred broadly across the source samples, with relatively high concentrations of Cu, Zn, and Mn observed in livestock manure and compost. Mean concentrations reached 1387.6 mg kg⁻¹ for Mn in cattle manure, 1272.1 mg kg⁻¹ for Zn in

swine manure, 480.2 mg kg⁻¹ for Cu in poultry manure, and 835.6 mg kg⁻¹ for Zn in compost. Multiple heavy metal(loid)s were also detected in soil, liquid fertilizer, WWTP effluent, and LMTE effluent, indicating that their occurrence was not confined to livestock-derived materials. Contaminants originating from these potential sources can enter receiving waters through agricultural runoff and effluent discharge and subsequently accumulate in sediments (Ohore et al., 2020).

Heavy metal(loid)s in the stream sediments showed substantial spatial variability across the sampling sites (Fig. S2). As, Cu, and Zn showed the greatest spatial variability, with coefficients of variation of 69.0%, 62.1%, and 57.6%, respectively (Table S15). Cu and Zn also showed more pronounced anthropogenic enrichment than the other analyzed elements, with EF values >3 and *I_{geo}* values >1 at multiple sites (Fig. S3). The relatively high Cu and Zn concentrations in livestock-derived source samples, together with their elevated concentrations in sediments near the LMTE, suggest that livestock manure-derived inputs may have contributed to their enrichment at some sites. Similar accumulation of Cu and Zn associated with manure inputs has been reported in intensive livestock farming areas (Jiao et al., 2019). Spearman correlation analysis showed significant positive correlations among several heavy metal(loid)s (Fig. 4a). CTC was significantly and positively correlated with Cu and Mn, whereas significant correlations between antibiotics and other heavy metal(loid)s were limited (Fig. 4b). The limited correlations may partly reflect differences in the fate and transport of antibiotics and metal(loid)s, as well as spatial variability in geogenic contributions to sediment metal(loid) concentrations (Han et al., 2021; Xu et al., 2023). Such source-related associations can also weaken with increasing distance from common inputs as differences in fate and transport become more pronounced (Kerrigan et al., 2018). Nevertheless, relatively high

concentrations of antibiotics and some heavy metal(loid)s were observed at S6 and S8 (Fig. 4c), indicating their concurrent accumulation at sites influenced by local pollution sources.

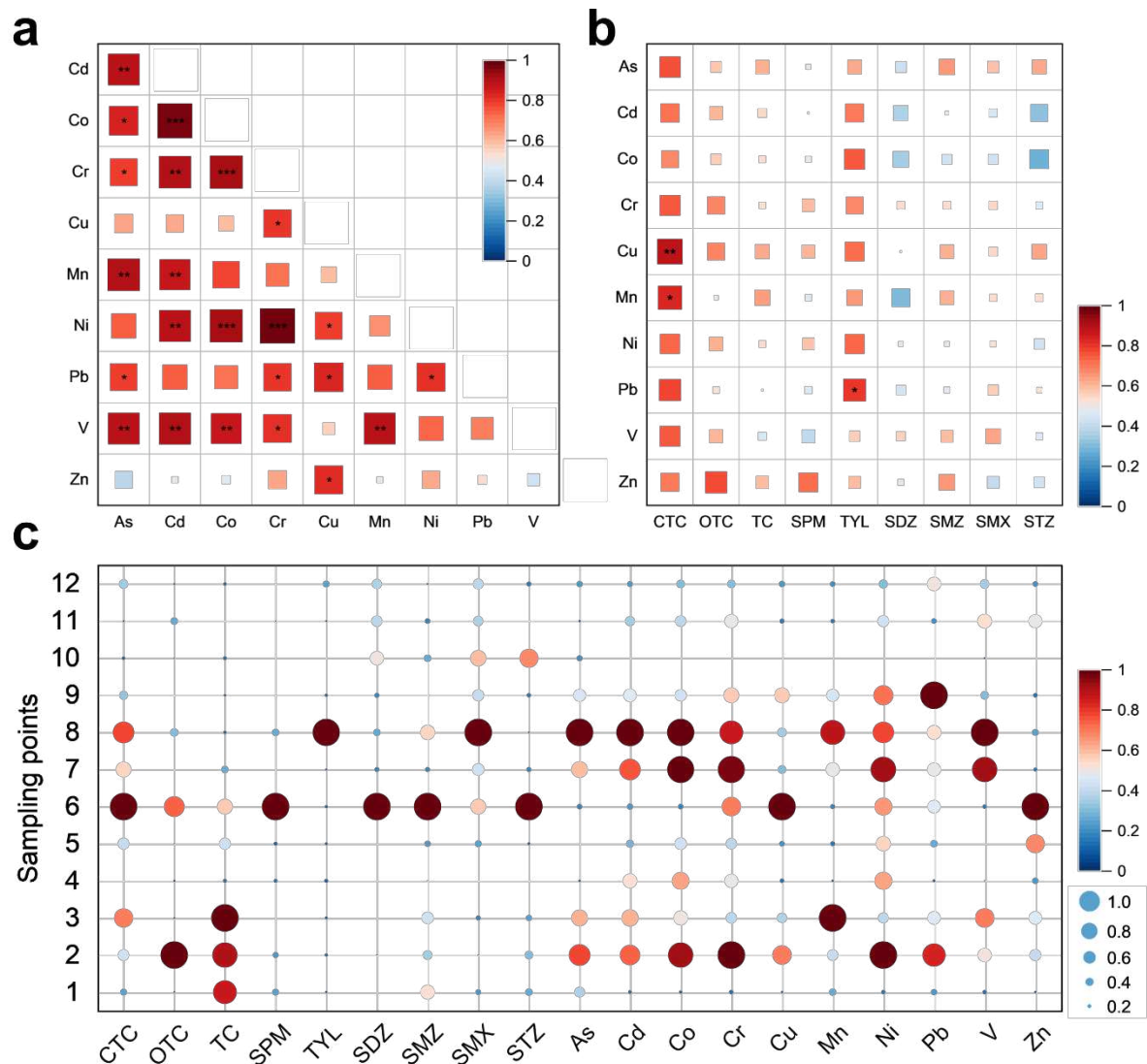


Fig. 4. Correlations and site-specific distribution patterns of antibiotics and heavy metal(loid)s in sediments from Muhan Stream. Correlation matrices showing the relationships (a) among heavy metal(loid)s and (b) between antibiotics and heavy metal(loid)s. Statistical significance levels are denoted by asterisks as follows: * $p < .05$, ** $p < .01$, and *** $p < .001$. (c) Site-specific patterns of min-max normalized concentrations of antibiotics and heavy metal(loid)s. For each

compound, concentrations measured across the sampling points were normalized to a 0–1 scale using min–max normalization. Bubble color and size indicate the normalized relative concentration of each compound at each sampling point.

The observed co-occurrence of antibiotics and heavy metal(loid)s, together with their presence in potential source samples, suggests that both contaminant groups may reflect common local pollution inputs. However, the limited correlations between them indicate that heavy metal(loid)s did not simply duplicate the information contained in antibiotic profiles. Their distinct multielement variation may therefore provide complementary contrasts for resolving source-related factors. Although Cu and Zn showed the clearest anthropogenic enrichment and were consistent with livestock manure-derived inputs, limiting the auxiliary indicators to these elements could overlook multielement differences among potential sources. Accordingly, all ten measured heavy metal(loid)s were retained in the integrated PMF analysis to evaluate whether their combined patterns could reduce ambiguity in the interpretation of the resolved source-related factors. Individual heavy metal(loid)s were not assigned to specific sources a priori; instead, their roles were interpreted after factor resolution using the resulting contaminant profiles, site-specific contribution patterns, and information from the source samples.

3.4. Source apportionment using an integrated PMF model with heavy metal(loid) indicators

An integrated PMF model was constructed by incorporating ten heavy metal(loid)s into the antibiotic dataset as complementary source indicators. The same modeling procedures and

diagnostic criteria used for the antibiotic-based model were applied to allow direct comparison between the two models (Text S6 and Table S18). Across the three- to five-factor solutions, the diagnostic results did not indicate major instability. Source interpretation, however, remained ambiguous in the three- and four-factor solutions and improved in the five-factor solution. The five-factor solution was therefore examined further using repeated random-seed runs. Reproducibility differed among factors, but the overall results supported its selection as the final model for source apportionment (Table S13). The resolved factors were then interpreted in relation to their potential pollution sources (Fig. 5a).

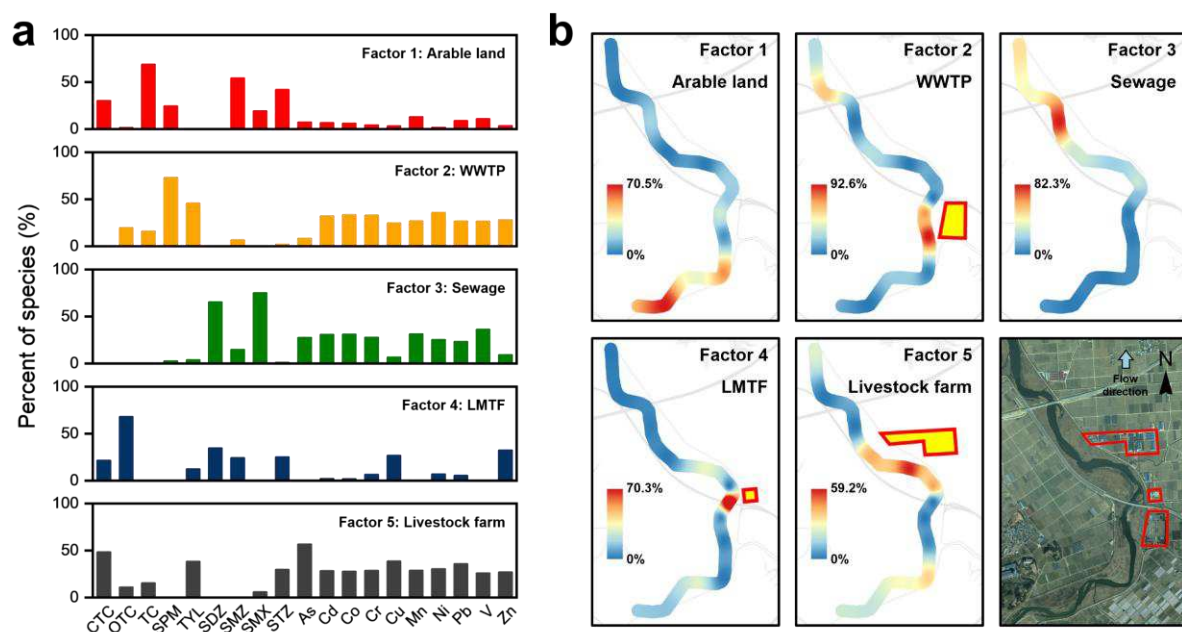


Fig. 5. Results of the integrated PMF model using antibiotics and heavy metal(loid)s as input variables in sediments from Muhan Stream. (a) PMF factor profiles for the five resolved factors, arranged in the same source-interpretation order as the antibiotic-based PMF model. (b) Maps generated by applying inverse distance weighting (IDW) interpolation to the site-specific factor contributions. The locations or areas of the corresponding potential pollution sources for

Factors 2, 4, and 5 are marked with red-outlined boxes. The color scale for each factor map was independently scaled to the maximum site-specific contribution observed for that factor.

Factor 1 retained the arable land-related antibiotic profile identified in the antibiotic-based PMF model, with high contributions of TC (76.8%), SMZ (57.2%), and STZ (44.2%). The heavy metal(loid) profile showed relatively even contributions, as indicated by Co (18.1%), Cr (15.5%), and Mn (21.8%), without a pronounced contribution from any single element. This pattern was consistent with diffuse inputs from agricultural soils (Conrad et al., 2020).

Factor 2 showed markedly lower contributions of OTC (13.5%) and SPM (19.2%) than in the antibiotic-based model, consistent with the low concentrations of these antibiotics near the WWTP. The heavy metal(loid) profile showed relatively even contributions, with a mean contribution of 23.5%, which was compatible with the mixed composition of municipal wastewater (Feng et al., 2018). The relatively high site-specific contribution of Factor 2 near the WWTP further supported its interpretation as WWTP effluent.

Factor 3 retained the SA-dominated profile observed in the antibiotic-based model, with SDZ (71.7%) and SMX (70.3%) remaining the dominant antibiotics. The heavy metal(loid) profile was broad, with a mean contribution of 21.0% and no clear dominance by any single element. This pattern was consistent with the occurrence of multiple metals reported in untreated domestic wastewater (Houhou et al., 2009; Van de Moortel et al., 2010). Although the broad heavy metal(loid) profile was not distinctive for this source, the persistent predominance of SAs supported the interpretation of Factor 3 as untreated domestic wastewater.

Factor 4 was characterized by high contributions of SPM (70.5%) and OTC (58.3%) in the integrated model. The predominance of SPM and OTC was consistent with the relatively high

concentrations of these antibiotics near the LMTF. Factor 4 also showed contributions of Cu (21.3%) and Zn (21.0%), which are commonly associated with livestock manure (Shelton et al., 2011; Yazdankhah et al., 2014). Taken together, the antibiotic profile, heavy metal(loid) profile, and spatial distribution supported the interpretation of Factor 4 as an LMTF-related input.

Factor 5 retained a clear livestock-related antibiotic profile, with high contributions of TYL (63.8%) and CTC (55.0%), both predominantly used in swine production in South Korea. The heavy metal(loid) profile was broader than that of Factor 4, with contributions from multiple elements. This multielement pattern was consistent with runoff carrying manure, wastewater, soil, and particulate matter from livestock farms (Du et al., 2014). Among the elements, As showed a particularly high contribution (62.5%), which may partly reflect livestock-related inputs associated with the historical use of organoarsenic feed additives in animal production (Eom et al., 2015; Makris et al., 2008). These characteristics supported the interpretation of Factor 5 as runoff from livestock farming areas.

The site-specific contribution patterns of the integrated model were more spatially coherent with known source locations than those of the antibiotic-based model (Fig. S4 and Table S14). In the antibiotic-based model, Factor 2 showed similar contributions at S4 near the WWTP (56.4%) and S6 near the LMTF (53.9%). In contrast, the integrated model showed a distinct spatial pattern, with Factor 2 contributing 92.6% at S4 and Factor 4 contributing 70.3% at S6, reducing ambiguity between the WWTP- and LMTF-related interpretations. Although Factor 2 was not restricted to the immediate vicinity of the WWTP, its overall spatial pattern was more consistent with the WWTP-related interpretation, while Factor 5 showed higher contributions near livestock farming areas. These changes suggest that the multielement information

provided by heavy metal(loid)s introduced complementary contrasts that were not captured by the antibiotic dataset alone. Such multielement profiles have been shown to provide useful source-related information in sediment source-apportionment studies (Chen et al., 2016; Li et al., 2020). Overall, the combined elemental patterns supported a clearer and more spatially coherent interpretation of the resolved factors.

3.5. Ecological risk distribution based on PMF-derived factor contributions

The RQs of the nine antibiotics detected across the 12 sampling sites ranged from 0.00 to 1.48 (Fig. 6a). Among the antibiotic classes, TCs had the highest mean RQ (0.36), followed by MLs (0.16) and SAs (0.04). Most antibiotics in the Muhan Stream watershed had RQ values below 1. However, CTC detected at S6 and TYL at S8 exceeded 1.00 with RQ values of 1.48 and 1.38, respectively. The total RQs were also highest at S6 and S8, reaching 3.71 and 3.23, respectively. CTC and TYL both showed high RQs, but the factors influencing their ecological risks in sediments differed between the two compounds. For CTC, the high RQ was associated with its relatively high sediment concentration, which may reflect its substantial use in livestock production and strong retention in sediments, as discussed in Section 3.1. TYL occurred at lower sediment concentrations than CTC but showed a similar maximum RQ, indicating that toxicological sensitivity also played an important role in its risk. Previous studies have reported effects of TYL on the growth and photosynthetic activity of algae and cyanobacteria at relatively low concentrations (Halling-Sørensen, 2000; Li et al., 2021). In contrast, SAs showed the lowest mean RQ (0.04), even though some SAs occurred at concentrations higher than TYL at several sites. Their low sediment RQs, however, do not necessarily indicate negligible environmental concern because SAs can persist in aquatic

environments and result in prolonged exposure (Duan et al., 2022). These results suggest that the ecological risks of individual antibiotics in sediments were influenced by environmental fate and toxicity as well as by their concentrations.

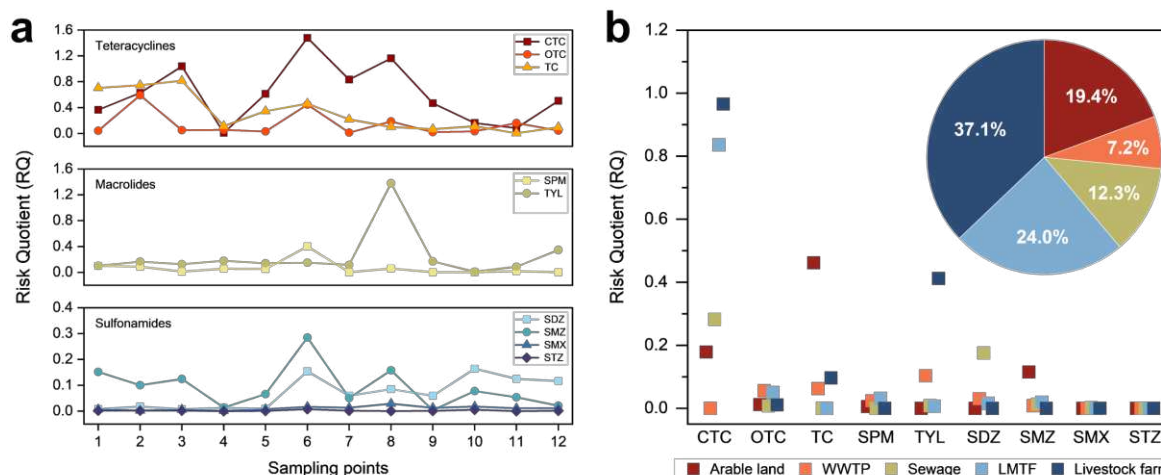


Fig. 6. Ecological risk assessment of antibiotics in Muhan Stream sediments. (a) Risk quotients (RQ_s) of the detected antibiotics across the sampling points. (b) Factor-attributed risk quotients (RQ_{source}) estimated using PMF-derived factor contributions and factor-specific antibiotic composition. The inset pie chart shows the relative contribution (%) of each factor to the total RQ_{source} across all antibiotics.

Factor-attributed RQ_s (RQ_{source}) were estimated by allocating the RQ of each antibiotic according to the PMF-derived factor contributions and factor-specific antibiotic composition (Fig. 6b). For CTC, RQ_{source} values attributed to the livestock farm runoff- and LMTF-related factors were 0.97 and 0.83, respectively. For TYL, the livestock farm runoff-related factor accounted for the largest RQ_{source} (0.41). When RQ_{source} values were summed across antibiotics, the livestock farm runoff-related factor accounted for the largest relative share (37.1%),

followed by the LMTF-related factor (24.0%) and the arable land-related factor (19.4%). Factors interpreted as domestic wastewater- and WWTP-related accounted for comparatively smaller shares, at 12.3% and 7.2%, respectively. These results indicate that factors interpreted as manure-related, encompassing livestock production, manure treatment, and agricultural land application, accounted for the largest relative shares of the factor-attributed screening RQ across the sampled sites. Accordingly, these manure-related source influences may warrant greater attention in future monitoring and source-oriented management within the watershed.

3.6. Study limitations and implications

The present study has several limitations that should be considered when interpreting the integrated PMF results. The relatively small receptor dataset and single-time-point sampling design may limit the generalizability of the resolved factor structure. The source samples were used mainly to support factor interpretation rather than as an independent quantitative validation dataset. Accordingly, the resolved factors and their contributions should be interpreted within the conditions represented by the sampled watershed. Nevertheless, the incorporation of heavy metal(loid)s provided complementary multielement information that reduced ambiguity among several source-related factors and supported a more spatially coherent interpretation. Future studies using larger receptor datasets, repeated sampling, and more extensive source characterization would help assess the robustness and transferability of the integrated PMF approach.

4. Conclusion

This study evaluated the complementary value of heavy metal(loid) information in PMF-based interpretation of antibiotic sources in stream sediments. The antibiotic-based model showed generally stable performance but retained ambiguity in distinguishing several source-related influences. Incorporating heavy metal(loid)s provided additional multielement contrasts while maintaining overall model stability and produced more distinct site-specific contribution patterns, particularly for the WWTP-, LMTF-, and livestock-related factors. These findings indicate that heavy metal(loid)s can complement antibiotic profiles by reducing ambiguity in the interpretation of source-related factors. Overall, the combined chemical and elemental information provided a clearer basis for source-related interpretation and subsequent screening-level risk prioritization.

Author's contribution

Jinwook Kim: Writing – original draft, Visualization, Investigation, Data curation, Conceptualization. Youngkyu Hong: Writing – review & editing, Investigation. Hojin Jung: Formal analysis, Data curation. Jeongmin Lee: Investigation, Data curation. Sungchul Kim: Writing – review & editing, Supervision, Conceptualization.

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Data availability

Data will be made available on request.

Declarations

Conflict of interest

The authors declare no competing financial or personal interests.

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