

目次

疫情期间人为源减排对城市大气氧化性的影响	朱剑蓝, 秦墨梅, 朱嫣红, 胡建林 (617)
不同天气形势对南京地区双高污染的输送及潜在源区分析	秦阳, 胡建林, 孔海江 (626)
不同方法判定南京臭氧生成敏感区的差异	陈柑羽, 李勋, 李琳, 秦墨梅, 谢鸣捷, 王鸣, 李婧伟, 胡建林 (635)
2006~2021 年夏半年上海臭氧浓度特征及其大气环流背景分析	郑庆峰, 梁萍, 段玉森, 林燕芬, 张宋嘉, 徐卫忠 (645)
基于大气成分观测网的山西省近地面 O ₃ 体积分数分布特征	李莹, 王淑敏, 裴坤宁, 闫世明, 孙鸿博, 张逢生, 高兴艾 (655)
伊宁市夏季大气臭氧生成机制及减排策略	王文婷, 谷超, 李丽明, 李新琪, 郑镇森, 耿春梅, 王晓丽, 杨文 (668)
运城市四季 VOCs 特征、来源及臭氧形成敏感物种	阴世杰, 刘新翌, 刘亚非, 李晨露, 张晨, 张欢, 王正, 程强 (678)
郑州市秋冬季污染过程中大气 VOCs 污染特征、来源解析及活性分析	赖梦洁, 张栋, 于世杰, 宋鑫帅, 李晚, 张瑞芹 (689)
郑州市 PM _{2.5} 中有机酸的污染特征、来源解析及二次生成	李子涵, 董喆, 尚瑞琪, 孔梓涵, 李晚, 张瑞芹 (700)
中国三大城市群 PM _{2.5} 浓度非线性变化分析	吴舒祺, 顾杨旸, 张天岳, 赵文吉 (709)
基于 LEAP 模型的临港新片区中长期碳排放预测及减排潜力分析	吴琰, 马昊, 任洪波, 郭明星, 陈鹏, 李琦芬 (721)
碳交易背景下中国华北地区碳代谢格局变化	郑宏媚, 沈方, 许光耀, 关欣 (732)
考虑区域特点和车型差异的氢燃料电池汽车全生命周期减碳预测分析	马菁, 蔡旭, 张春梅, 兰利波, 陈轶嵩, 付佩 (744)
我国主要河流水系硝态氮污染特征及定量源解析	韦英怀, 胡敏鹏, 陈丁江 (755)
不同时空尺度下土地利用结构与空间格局对苏州水质的影响	谭娟, 熊丽君, 王卿, 任志文, 朱丹丹, 王敏 (768)
深圳市 2015~2021 年雨源型河流水质时空变化及其对降雨的响应	韦必颖, 成建梅, 苏晓煜, 程天舜 (780)
河南黄河改道区浅层地下水化学特征与主控污染源解析	王帅, 任宇, 郭红, 曹文庚, 李祥志, 肖舜禹 (792)
北京西山岩溶地下水化学特征及成因分析	郭高轩, 代垠东, 许亮, 朱琳, 欧志亮, 戚琦, 辛宝东 (802)
店埠河流域地表水-地下水水化学特征及其成因分析	郑涛, 秦先燕, 吴剑雄 (813)
张家口地区枯水期地下水水化学特征及其成因机制分析	金爱芳, 殷秀兰, 李长青, 李文娟, 庞菊梅, 金晓媚 (826)
黄河中下游典型抗性细菌及抗性基因污染分布	闵威, 高明昌, 孙绍芳, 宋茜茜, 邱立平 (837)
制药废水中抗生素抗性的污染特征、检测手段和控制方法	彭安萍, 高虎, 张新波 (844)
水体组分对聚苯乙烯纳米颗粒聚沉行为的影响	汤端阳, 郑文丽, 陈关潼一, 陈思莉, 陈尧, 赵晓丽, 汪浩 (854)
富磷废弃钙基生物炭对水体中铅的去除	刘天, 吕思璐, 杜兴国, 程敏, 谢燕华 (862)
壳聚糖改性生物炭的制备及其对水溶液中 Cd ²⁺ 的吸附机制	姜凌, 安靖玥, 岳小琼, 李亚雄, 夏秋乐, 祝婷文佳, 柴丽红 (873)
硼掺杂介孔炭吸附四环素的效能与机制	邹震, 许路, 乔伟, 唐茂森, 金鹏康 (885)
磁性含磷油茶壳生物炭对水中磺胺甲噁唑的吸附特性	韩帅鹏, 唐李文, 刘勤, 林家亮, 李晚慢, 程建华, 胡勇有 (898)
广东省高分辨率温室气体排放清单及特征	卢清, 唐明双, 廖彤, 黄志炯, 钟庄敏, 宋佩珊, 沈劲, 张智胜, 梁小明, 孙家仁, 陈来国 (909)
辽河口“退塘还湿”修复区生态系统 CO ₂ 交换及其环境调控	刘思琪, 陈虹, 邢庆会, 程浩, 韩建波, 徐雪梅 (920)
生物炭施用两年后对热带地区稻菜轮作土壤 N ₂ O 和 CH ₄ 排放的影响	胡煜杰, 唐瑞杰, 胡天怡, 陈琦琦, 汤水荣, 阮延正, 孟磊 (929)
生物炭改良盐碱地研究与应用进展	魏盈, 焦乐, 张鹏, 刘福德, 肖辉, 董昱辰, 代红文 (940)
免耕对农田土壤团聚体的影响研究: Meta 分析	徐艺萍, 饶越悦, 孟艳, 温媛, 孟维伟, 王旭清, 李宗新, 刘开昌, 代红翠 (952)
黔中喀斯特地区典型县域碳储量时空演变及多情景模拟预测: 以普定县为例	李月, 罗红芬 (961)
不同改良剂对酸性紫色土团聚体和有机碳的影响	李越, 徐曼, 谢永红, 王颖, 黄容, 谢军, 王子芳, 高明 (974)
Ca 改性生物炭对土壤磷赋存形态影响及稳定化机制	张超, 翟付杰, 单保庆 (983)
秦岭中段不同恢复阶段弃耕农田植物多样性变化及其驱动因素	闫成龙, 薛悦, 王艺菲, 康海斌, 王得祥 (992)
我国典型制药厂污染场地中抗生素的污染特征及生态风险	杨炳彬, 黄争, 赵建亮, 何良英, 刘有胜, 胡立新, 石义静, 应光国 (1004)
广州市土壤多环芳烃污染特征及风险评估	邹子航, 陈莲, 张培珍, 王雨茜, 王振江, 林森, 唐翠明, 罗国庆, 钟建武, 李智毅, 王圆 (1015)
基于源导向的土壤重金属风险评价及管控因子分析	潘泳兴, 陈盟, 王樯樯 (1026)
基于 Monte-Carlo 模拟的湖南省典型工厂周边农田土壤重金属区域潜在生态风险特征及来源解析	罗豪杰, 潘俊, 陈小霞, 张敏, 沈良辰, 李歆, 丁平, 蔡丹, 蔡立梅, 胡国成 (1038)
基于参数优化和蒙特卡罗模拟的砷污染地块健康风险评估	袁贝, 刘虎鹏, 杜平, 陈娟, 张云慧, 张昊 (1049)
基于 APCS-MLR 和 PMF 模型的赤泥堆场周边耕地土壤重金属污染源解析	沈智杰, 李杰芹, 李彩霞, 廖泽源, 梅楠, 罗程钟, 王定勇, 张成 (1058)
PE-Cd 复合污染土壤中 Cd 释放迁移特征及机制	王迪, 徐绍辉, 邵明艳, 林青 (1069)
氯代乙烯的厌氧微生物还原脱氯特性	李伟, 刘贵平, 刘峻, 吕良华, 乔文静, 余欣, 张晚晓, 蒋建东 (1080)
昭通市农田土壤和蔬菜重金属污染评价及相关性分析	张好, 董春雨, 杨海婵, 孙思静, 韩宇, 黄祖志, 张乃明, 包立 (1090)
钝化剂对轻中度镉污染农田的安全利用效果	王晓晶, 张东明, 曹阳, 吕家琰, 代允超 (1098)
氧化石墨烯负载铁锰复合材料对镉污染土壤的钝化修复	袁婧, 吴骥子, 连斌, 袁峰, 孙洪, 田欣, 赵科理 (1107)
关键生育期施加外源灌溉水对水稻镉吸收转运的影响	周霞, 胡雨丹, 周航, 陈琼, 谭文韬, 曾鹏, 辜娇峰, 廖柏寒 (1118)
外源锌对镉胁迫下玉米幼苗生长及根系构型分级的影响	张辉红, 魏畅, 柳海涛, 张静静, 刘芳, 赵颖, 张雪海, 李鸽子, 姜瑛 (1128)
稀土元素铈对镉胁迫下小麦幼苗生长的缓解效应	张静静, 徐正阳, 焦秋娟, 范丽娜, 刘芳, 赵颖, 宋佳, 化党领, 李鸽子, 柳海涛 (1141)
根施伯克氏菌对小麦镉吸收转运的两段式阻控作用	郭佳佳, 王常荣, 刘仲齐, 黄青青, 张长波, 黄永春, 薛卫杰, 孙约兵 (1150)
高密度聚乙烯微塑料与氯噻磺隆对大豆生长和根际细菌群落的复合胁迫效应	胡晓玥, 滑紫微, 姚伦广, 杜丽, 牛秋红, 李玉英, 闫路, 陈兆进, 张浩 (1161)
微塑料的人体富集及毒性机制研究进展	包亚博, 王成尘, 彭吾光, 依代倩, 向萍 (1173)
机器学习在微塑料识别与环境风险评估中的应用研究进展	白润昊, 范瑞琪, 刘琪, 刘勤, 严昌荣, 崔吉晓, 何文清 (1185)
微塑料与农田土壤中典型污染物的复合污染研究进展	侯宇晴, 李冰, 王金花, 宋文慧, 王兰君, 王军, 朱鲁生 (1196)
水中微/纳塑料电化学检测及去除的研究进展	郑伟康, 刘振中, 项晓方 (1210)
基于分布式认知理论的农户面源污染治理支付意愿影响因素	郭晨浩, 李林霏, 夏显力 (1222)

我国主要河流水系硝态氮污染特征及定量源解析

韦英怀^{1,2}, 胡敏鹏^{1,2}, 陈丁江^{1,2,3*}

(1. 浙江大学环境与资源学院, 杭州 310058; 2. 浙江大学环境修复与生态健康教育部重点实验室, 杭州 310058; 3. 浙江大学浙江省农业资源与环境重点实验室, 杭州 310058)

摘要: 准确定量污染来源组成是有效控制水体硝态氮污染的关键科学基础。采用荟萃分析的方法, 收集了2000~2022年我国167条主要水系河流的硝态氮浓度和硝态氮的氮氧同位素等数据, 分析了七大主要河流水系硝态氮污染的时空变异规律及其转化特征, 定量识别了河流硝态氮的污染来源组成。结果表明, 我国主要河流水系 $\rho(\text{NO}_3\text{-N})$ 平均值为 $(4.54 \pm 3.99) \text{ mg} \cdot \text{L}^{-1}$, 其中9.6%的河流硝态氮浓度超过我国地表水环境质量标准(GB 3838-2002)规定的限值 $(10.0 \text{ mg} \cdot \text{L}^{-1})$, 海河水系的硝态氮污染最为严重。东部地区河流水系的硝态氮浓度总体高于西部, 各大河流水系支流的硝态氮浓度高于干流。除黄河水系以外, 其他水系枯水期的硝态氮浓度总体高于丰水期。珠江水系、黄河水系中下游地区、辽河水系中游地区、松花江水系, 以及海河水系河流水体存在显著的硝化作用, 而长江水系、淮河水系和珠江水系下游地区存在显著的反硝化作用。污水/粪肥是长江水系、海河水系、辽河水系, 以及东南诸河水系硝态氮的主要来源($>50\%$), 土壤氮是松花江水系硝态氮的主要来源(56.4%), 化肥氮、土壤氮和污水/粪肥对珠江水系、淮河水系和黄河水系硝态氮的污染贡献为20%~40%。污水/粪肥对水系支流硝态氮贡献率总体大于干流的, 土壤氮对干流硝态氮的贡献总体大于支流的。土壤氮、化肥氮和大气沉降氮对丰水期河流硝态氮的贡献率高于枯水期, 而污水/粪肥对枯水期河流硝态氮的污染贡献率高于丰水期。因此, 海河水系、长江水系、辽河水系、黄河水系支流与下游干流地区和珠江水系下游地区应重点控制生活和生产的污水排放等点源污染, 而淮河水系、松花江水系、黄河水系中游干流地区和珠江水系中上游地区要重点控制化肥和土壤氮等流失造成的非点源污染。研究结果可为有效控制我国各河流水系硝态氮的污染提供科学依据。

关键词: 硝态氮; 源解析; 河流; 氮氧同位素; 非点源污染; 点源污染

中图分类号: X522 文献标识码: A 文章编号: 0250-3301(2024)02-0755-13 DOI: 10.13227/j.hjxx.202304069

Nitrate Pollution Characteristics and Its Quantitative Source Identification of Major River Systems in China

WEI Ying-huai^{1,2}, HU Min-peng^{1,2}, CHEN Ding-jiang^{1,2,3*}

(1. College of Environmental & Resource Sciences, Zhejiang University, Hangzhou 310058, China; 2. Ministry of Education Key Laboratory of Environment Remediation and Ecological Health, Zhejiang University, Hangzhou 310058, China; 3. Zhejiang Provincial Key Laboratory of Subtropical Soil and Plant Nutrition, Zhejiang University, Hangzhou 310058, China)

Abstract: Accurate source identification/apportionment is essential for optimizing water $\text{NO}_3\text{-N}$ pollution control strategies. This study conducted a meta-analysis based on data from 167 rivers across China from 2000 to 2022 to analyze the spatial and temporal variation patterns of nitrate pollution in seven major river systems and to quantitatively identify the source composition of riverine nitrate. The average $\rho(\text{NO}_3\text{-N})$ in the seven major river systems was $(4.54 \pm 3.99) \text{ mg} \cdot \text{L}^{-1}$, with 9.6% of river $\rho(\text{NO}_3\text{-N})$ exceeding $10 \text{ mg} \cdot \text{L}^{-1}$. The riverine $\rho(\text{NO}_3\text{-N})$ in eastern China were higher than that in western China, and the highest concentration was observed in the Haihe River system. Additionally, tributaries experienced more serious $\text{NO}_3\text{-N}$ pollution than that in the main stream. The $\rho(\text{NO}_3\text{-N})$ in most river systems in the dry season was higher than that in the wet season, except in the Yellow River system. There was significant nitrification in the Pearl River system, the middle and lower reaches of the Yellow River system, the middle reaches of the Liaohe River system, the Songhua River system, and the Haihe River system, whereas there was significant denitrification in the Yangtze River system, the Huaihe River system, and the lower reaches of the Pearl River system. Based on the dual stable isotopes-based MixSIAR model, the major $\text{NO}_3\text{-N}$ source was sewage/manure ($>50\%$) in the Yangtze River system, Haihe River system, Liaohe River system, and Southeast River system. Soil nitrogen was the main $\text{NO}_3\text{-N}$ source in the Songhua River system (56.4%), and the contribution of fertilizer nitrogen, soil nitrogen, and sewage/manure to $\text{NO}_3\text{-N}$ pollution in the Pearl River system, Huai River system, and Yellow River system was 20%-40%. The contribution rate of sewage/manure to $\text{NO}_3\text{-N}$ in the tributaries was higher than that in the main stream, whereas the contribution rate of soil nitrogen to $\text{NO}_3\text{-N}$ in the main stream was higher than that in the tributaries. The contribution rate of soil nitrogen, fertilizer nitrogen, and atmospheric deposition nitrogen to nitrate nitrogen in the wet season was higher than that in the dry season, whereas the contribution rate of sewage/manure to $\text{NO}_3\text{-N}$ pollution in the dry season was higher than that in the wet season. Therefore, point source pollution such as domestic and production sewage discharge should be controlled in the Haihe River system, the Yangtze River system, the Liaohe River system, the tributaries and the downstream main stream areas of Yellow River system, and the downstream area of the Pearl River system, whereas non-point source pollution caused by the loss of fertilizer and soil nitrogen should be controlled in the Huaihe River system, the Songhua River system, the middle reaches of the main stream area of the Yellow River system, and the middle and upper reaches of the Pearl River system. The results can provide a scientific basis for the effective control of nitrate pollution in the river systems in China.

Key words: nitrate; source identification; river; nitrogen and oxygen isotopes; non-point source pollution; point source pollution

收稿日期: 2023-04-09; 修订日期: 2023-05-01

基金项目: 国家自然科学基金项目(42107393, 42177352); 国家重点研发计划项目(2021YFD1700802)

作者简介: 韦英怀(1997~), 男, 硕士研究生, 主要研究方向为流域非点源污染控制, E-mail: 22014131@zju.edu.cn

* 通信作者, E-mail: chendj@zju.edu.cn

随着社会经济和农业活动的迅速发展,近几十年来全球水环境污染问题日益严重.许多河流的硝态氮浓度迅速上升,超过了世界卫生组织以及我国地表水环境质量标准(GB 3838-2002)规定的安全界限(10.0 mg L^{-1})^[1~4].硝态氮污染不仅会对河流水生生态系统造成严重影响,引起水体富营养化和有害藻华的暴发^[5],而且会直接或者间接地威胁人体健康^[6,7].由于水体中硝态氮来源于多个污染源(如化肥和生活污水等),因此,定量其来源组成是实现高效防治的关键科学基础.

目前,定量解析河流等水体氮污染来源组成的方法主要包括模拟模型法(包括集总式模型和分布式模型)和基于氮氧同位素的源解析法^[8~10].由于基础数据(包括水文气象要素、水化学指标、土地利用分布和地形等)不足和空间异质性考虑不足等原因,应用模拟模型法解析大区域尺度河流水系氮污染源组成可能存在较大误差^[9,11~13],如Hu等^[11]的研究发现,应用不同模型对长江氮污染进行源解析得到的非点源污染贡献为35%~99%,结果存在较大的不确定性.近几十年来,随着同位素技术的发展,基于氮氧同位素的源解析研究日益增多^[4].相较于使用复杂的流域水文水质模型法,基于氮氧稳定性同位素的硝态氮源识别方法具有操作简便,源识别结果较可靠等优点,已被广泛应用于评估不同水体尤其是大区域尺度河流水系氮转化过程和来源组成^[15~18].例如,张金兰等^[9]利用氮氧同位素技术识别了我国河流水系的水体硝态氮来源组成,结果表明,城镇用地为主的河流点源污染问题(污水源)突出,而农业用地为主的河流则受土壤氮、化肥和污水/粪肥的共同影响.当然,在基于氮氧同位素的源识别方法应用过程中也存在一定局限性.例如,由于水体中硝态氮的潜在来源较多,其 $\delta^{15}\text{N}-\text{NO}_3^-$ 和 $\delta^{18}\text{O}-\text{NO}_3^-$ 特征值范围较广,使得在数据量偏少的情况下的各来源贡献识别结果存在较大的不确定性^[4,18].目前,我国针对水体氮氧同位素溯源的研究主要集中在对单个流域或某条河流的硝态氮污染的溯源.近几年,有少量研究应用荟萃分析方法,开展了基于氮氧同位素的全国尺度河流水系硝态氮污染源识别研究^[19,20].然而,已有研究收集的数据不够全面,且未考虑时间变异性以及干流与支流的差异性^[16],对我国各河流水系的硝态氮污染特征及来源组成尚缺乏较为系统认识.

本研究收集了2000~2022年我国七大河流水系硝态氮浓度和氮氧同位素等文献报道数据,结合各河流水系流域土地利用类型和人口密度等数据,分析了我国主要河流水系的硝态氮污染特征和来源组成,比较了不同区域、不同水文期以及干流与支流之

间的差异性,从而较为系统地揭示了我国主要河流水系硝态氮来源组成的时空分布特征,以期有效控制我国河流水系的硝态氮污染提供科学依据.

1 材料与方法

1.1 数据来源

本研究基于Web of Science和中国知网数据库,检索了2000~2022年间包含“nitrogen and oxygen stable isotopes”、“nitrate sources”、“source identification”、“river stream”、“硝酸盐”、“硝态氮”、“氮氧同位素”和“源解析”关键词的文献,共获得了406篇文献.对所得文献进行整理时,排除了研究区域为国外河流水系的文献、综述性文献和基于水文水质模型等进行数据模拟的文献,仅保留了提供我国流域内各采样点真实数据的178篇文献.数据收集过程中,对文献中有提供数据的就直接收集整合,而对文献中仅提供氮氧同位素特征值散点图的,则采用Solvusoft公司开发的GetData Graph Digitizer v2.24数据提取软件对散点图进行数据提取.最终得到的数据集包含了我国主要水系的167条河流水体的6124个硝态氮浓度数据和5689对硝态氮的氮氧同位素数据,以及268对硝态氮潜在污染源氮氧同位素特征值数据.

本研究对收集到的某条河流多年同位素数据进行取平均值的处理,处理的原则如下:①当进行水系内部整体分析时,干流数据保持各文献中收集的从上游到下游的数据点位不变,若同一个点位有多年数据的对其取平均值,若不同文献中存在相近点位(以乡镇为单位,点位处于同一乡镇范围内为相近点位)的亦对其进行取平均值的处理;支流数据按其支流等级进行处理,将涉及到的各支流依据一级支流、二级支流、三级支流及以下进行划分,对于一级支流,将其划分为上、中、下游3个部分,分别对上、中、下游的数据进行取平均值的处理,而二级支流分别对其上游与下游的数据进行取平均值的处理,三级支流及以下则整体取平均值,最终保持干流与所有支流的相对数据量不变;②当进行水系干流与支流的对比分析时,则保持各文献中支流数据点位不变,只对其多年数据取平均值,干流数据处理方式不变;③当进行不同水文期(只因降雨因子的改变形成的丰水期与枯水期)的对比分析时,先将数据分为丰水期与枯水期数据,不同水文期的干支流数据处理方式与水系内部整体分析时的处理方式相同.

表1中各水系流域面积、年均径流量和耕地面积来源于国家统计局(<http://www.stats.gov.cn/tjsj/>)和

中国年鉴网络出版总库(<http://acad.cnki.net/kns55/brief/result.aspx?dbPrefix=CYFD>),人口密度采用第六次与第七次全国人口普查数据并使用 ArcGIS 10.8 计算各水系人口密度,土地利用方式和土壤侵蚀强度数据来源于中国科学院资源环境科学与数据中心(<http://www.resdc.cn/>),废水排放量和肥料施用量来源于国家统计局与各省市的统计年鉴,采用 2008~

2020 年多年平均值,并使用 ArcGIS 10.8 计算排放强度与施用强度,畜禽养殖数量(2008~2020 年)来源于国家统计局与中国经济社会大数据研究平台(<http://data.cnki.net/YearData/Analysis>). 将全国污染普查《排放源统计调查产排污核算方法和系数手册》提出的清单法与 ArcGIS 10.8 结合,估算畜禽养殖氮排放强度(表 1).

表 1 我国主要河流水系的基本属性与人类活动情况
Table 1 Basic properties and human activities of major river systems in China

水系	流域面积 /万 km ²	年均径流量 /亿 m ³	人口密度 /人·km ⁻²	耕地面积 /×10 ³ hm ²	废水排放强度 /万 t·km ⁻²	肥料施用强度 /t·km ⁻²	畜禽养殖氮排放 强度/t·km ⁻²
珠江	45.3	4 685	181	4 667	2.70	13.3	0.81
长江	180	9 513	233	23 467	1.93	10.1	0.55
淮河	18.6	741	611	12 333	2.36	33.9	0.93
黄河	75.2	661	122	12 133	0.60	9.80	0.31
海河	26.9	288	417	11 333	1.95	16.7	0.84
辽河	16.4	487	148	4 400	1.16	9.95	0.65
松花江	55.7	733	92	10 467	0.43	8.58	0.36

1.2 基于氮氧同位素的河流硝态氮定量源解析

MixSIAR 混合模型是一种用于基于氮氧同位素定量识别硝态氮来源的方法,综合了传统的 SIAR 模型和 IsoSource 模型的优点,能够计算多种来源对硝态氮的贡献率,并将相似的来源进行分类.相比于传统的 SIAR 模型和 IsoSource 模型,MixSIAR 模型操作界面可视化程度高,模型各参数设置方便快捷. MixSIAR 混合模型丰富了来源数据的输入形式,增加了固定效应、随机效应、不同的误差结构选择和指定先验信息等参数,从而提升了模型的精确度.在模型运行结束后,增加了模型诊断功能,只有模型运行无误且结果符合要求才能进行分析^[21,22],进一步保证了源识别结果的可靠性. MixSIAR 模型的表达式如下^[23,24]:

$$\delta^{15}\text{N}_i = \sum_{k=1}^n P_k (S_k + C_k) + \varepsilon_i \tag{1}$$

$$\delta^{18}\text{O}_i = \sum_{k=1}^n P_k (S_k + C_k) + \varepsilon_i \tag{2}$$

$$S_k \sim N(\mu_k, \omega_k^2) \tag{3}$$

$$C_k \sim N(\lambda_k, \tau_k^2) \tag{4}$$

$$\varepsilon_k \sim N(0, \sigma^2) \tag{5}$$

式中, $\delta^{15}\text{N}_i$ 和 $\delta^{18}\text{O}_i$ 表示样本*i*的 $\delta^{15}\text{N}\text{-NO}_3^-$ 和 $\delta^{18}\text{O}\text{-NO}_3^-$ 值, $i=1,2,3,\cdots,n$; k 表示潜在的硝态氮来源, $k=1,2,3,4$,分别表示大气沉降、化肥、土壤氮以及污水/粪肥; P_k 表示源*k*的比例,需要通过贝叶斯混合模型来估计; S_k 表示源*k*的同位素组成,服从平均值为 μ_k 和标准偏差(SD)为 ω_k^2 的正态分布; C_k 表示源*k*的分馏因子,服从平均值为 λ_k 和标准偏差(SD)为 τ_k^2 的正态分布; ε_k 表示残余误差,是单个混合物之间的额外未量

化变化,并且服从均值为 0 和标准偏差(SD)为 σ^2 的正态分布^[25].

本研究利用由 R 软件包(pacman)创建的贝叶斯混合模型(MixSIAR 版本 4.0.4)量化 4 种氮源(表 2)对我国七大水系硝态氮的污染贡献.根据已有数据整理汇编,得到我国七大水系 4 种氮源的氮氧同位素特征值范围如表 2 所示.

1.3 统计分析

本研究采用 ANONA 单因素方差分析及多重比较方法(Turkey 检验, $P<0.05$ 表示有显著性)分析河流水体硝态氮浓度的时空差异显著性.对同位素特征值散点图进行数据提取时,采用 Solvusoft 公司开发的 GetData Graph Digitizer v2.24 数据提取软件.各大河流水系硝态氮浓度的空间分布采用美国 Esri 公司开发的 ArcGIS 10.8 处理生成,对于文献中未提供河流水质监测点位经纬度的,依据河流名称进一步网上查找,确定河流经纬度范围,从而确定七大水系的河流水质监测点位分布位置,以表示各支流及干流河段硝态氮浓度平均值.数据统计与分析采用 Excel 和 OriginLab 公司开发的 Origin 2020 软件,图表制作则采用了 R 语言(Version 4.04)、Excel 和 Origin 2020 软件.

2 结果与讨论

2.1 我国主要河流水系的硝态氮污染特征分析

2000~2022 年,我国主要河流水系 $\rho(\text{NO}_3^- \text{-N})$ 平均值(以 N 计,下同)为 $(4.54\pm3.99)\text{mg}\cdot\text{L}^{-1}$,超标率为 9.6%(表 3).从不同河流水系的硝态氮浓度平均值差异看,海河水系>长江水系>辽河水系>黄河水

表 2 我国主要河流水系硝态氮潜在来源的氮氧同位素特征值/‰

Table 2 Characteristic values of nitrogen and oxygen isotopes of potential sources of nitrate nitrogen in major river systems in China/‰

水系名称	硝态氮源	$\delta^{15}\text{N}$ 平均值	$\delta^{15}\text{N}$ 标准差	$\delta^{18}\text{O}$ 平均值	$\delta^{18}\text{O}$ 标准差	文献
珠江	大气沉降	3.10	1.50	56.7	17.8	[14,16,26,27]
	化肥氮源	4.20	0.20	-5.70	1.70	
	污水/粪肥	11.5	3.20	-5.70	1.70	
	土壤氮源	5.70	2.00	-6.20	0.40	
长江	大气沉降	-1.50	1.80	58.2	14.2	[28 ~ 30]
	化肥氮源	0.90	2.00	3.00	1.70	
	污水/粪肥	13.3	0.80	0.60	1.10	
	土壤氮源	2.20	2.60	0.60	2.00	
淮河	大气沉降	-2.70	2.10	61.7	13.7	[14,31]
	化肥氮源	0.04	1.90	2.90	0.60	
	污水/粪肥	15.7	4.70	2.90	0.60	
	土壤氮源	4.50	2.70	2.90	0.60	
黄河	大气沉降	3.40	2.40	21.3	8.00	[32,33]
	化肥氮源	0.20	2.20	1.40	0.30	
	污水/粪肥	12.5	2.50	2.50	2.20	
	土壤氮源	6.70	1.30	1.40	0.30	
海河	大气沉降	4.20	4.30	54.0	13.2	[34,35]
	化肥氮源	0.40	0.30	2.90	1.70	
	污水/粪肥	14.3	1.90	6.70	2.50	
	土壤氮源	1.80	1.60	1.70	0.50	
辽河	大气沉降	2.70	2.90	73.7	8.30	[14,36]
	化肥氮源	0.01	0.80	-3.30	1.40	
	污水/粪肥	16.7	8.50	-3.40	1.40	
	土壤氮源	6.40	0.60	-6.20	0.40	
松花江	大气沉降	0.50	2.90	60.0	8.30	[37 ~ 39]
	化肥氮源	0.50	0.80	10.0	1.40	
	污水/粪肥	15.3	8.50	-2.50	1.40	
	土壤氮源	5.50	0.60	0.00	0.40	
东南诸河	大气沉降	-1.50	1.60	58.2	14.2	[40,41]
	化肥氮源	-0.53	0.20	2.90	1.70	
	污水/粪肥	10.5	4.50	3.50	2.60	
	土壤氮源	2.20	2.60	0.60	2.10	

系 > 淮河水系 > 珠江水系 > 松花江水系 > 东南诸河水系,海河水系和长江水系河流水体的硝态氮浓度显著高于其余水系 ($P<0.01$,表 3,图 1)。从不同河流水系硝态氮浓度的超标率看,海河水系 > 淮河水系 > 黄河水系 > 松花江水系 > 辽河水系 > 长江

水系 > 珠江水系和东南诸河水系,海河水系和淮河水系河流水体的硝态氮浓度超标率高于其余水系,珠江水系和东南诸河水系未发现超标现象(表 3)。因此,海河水系是我国主要河流水系中硝态氮污染最为严重的。

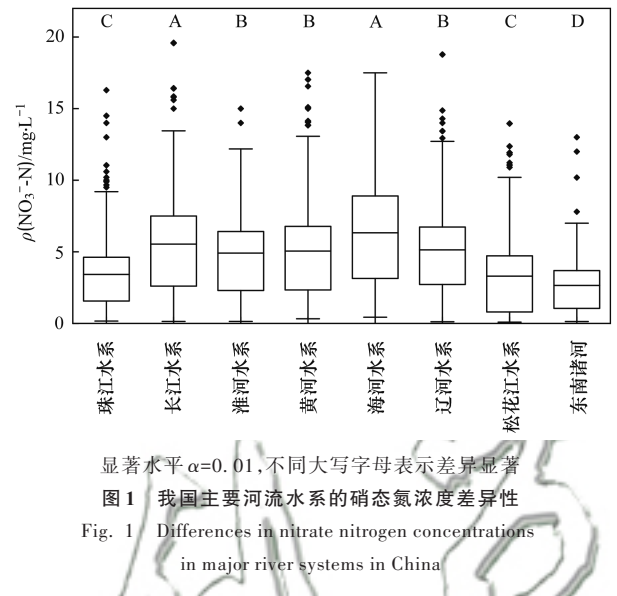
表 3 我国主要河流水系硝态氮浓度平均值及超标率

Table 3 Average values and exceeding percentages of nitrate nitrogen concentrations in major river systems in China

水系	数据量	$\rho(\text{NO}_3\text{-N})$ (平均值 $\pm$ 标准差)/ $\text{mg}\cdot\text{L}^{-1}$	河流数/条	超标河流数/条	超标率/%
珠江	812	3.37 $\pm$ 2.92	16	0	0.0
长江	1 564	5.53 $\pm$ 4.15	31	3	9.7
淮河	264	4.92 $\pm$ 2.33	6	1	16.7
黄河	1 276	5.08 $\pm$ 4.05	29	4	13.8
海河	884	6.38 $\pm$ 4.36	26	5	19.2
辽河	546	5.14 $\pm$ 3.87	20	2	10.0
松花江	431	3.25 $\pm$ 3.29	27	3	11.1
东南诸河	347	2.65 $\pm$ 2.75	12	0	0.0
全国	6 124	4.54 $\pm$ 3.99	167	16	9.6

珠江水系、松花江水系和东南诸河水系河流硝态氮浓度较低,大部分河流 $\rho(\text{NO}_3^--\text{N})$ 平均值介于0~4.99 $\text{mg}\cdot\text{L}^{-1}$ 之间,总体水质较好(图1和图2),但仍然存在恶化趋势,因此也需要加强保护.长江水系、淮河水系、黄河水系、海河水系和辽河水系河流硝态氮浓度较高,其中下游地区大部分河流 $\rho(\text{NO}_3^--\text{N})$ 平均值在10.0 $\text{mg}\cdot\text{L}^{-1}$ 左右,水质状况相对较差,特别是海河水系下游地区河流受生活污水以及畜禽粪便的影响严重^[34,42,43],硝态氮浓度超标率高,水体污染严重,因此需要重点治理.长江、淮河和海河流域的人口密度、耕地面积、废水排放强度、肥料施用强度和畜禽养殖业氮排放强度均较大(表1),尤其是中下游河流水体受人类活动影响较大^[44],导致河流硝态氮浓度较高.黄河流域耕地面积较大,土壤中含氮量较高,且水土流失较重,大量的土壤氮以及肥料和畜禽粪便容易流失进入到水体中^[33,45],导致水体硝态氮浓度偏高.辽河水系主要受工业活动的影响^[20],大量工业废水排放可能是河流硝态氮污染的重要原因.虽然珠江流域的废水排放强度与畜禽养殖业氮排放强度较高,但流域森林覆盖率高

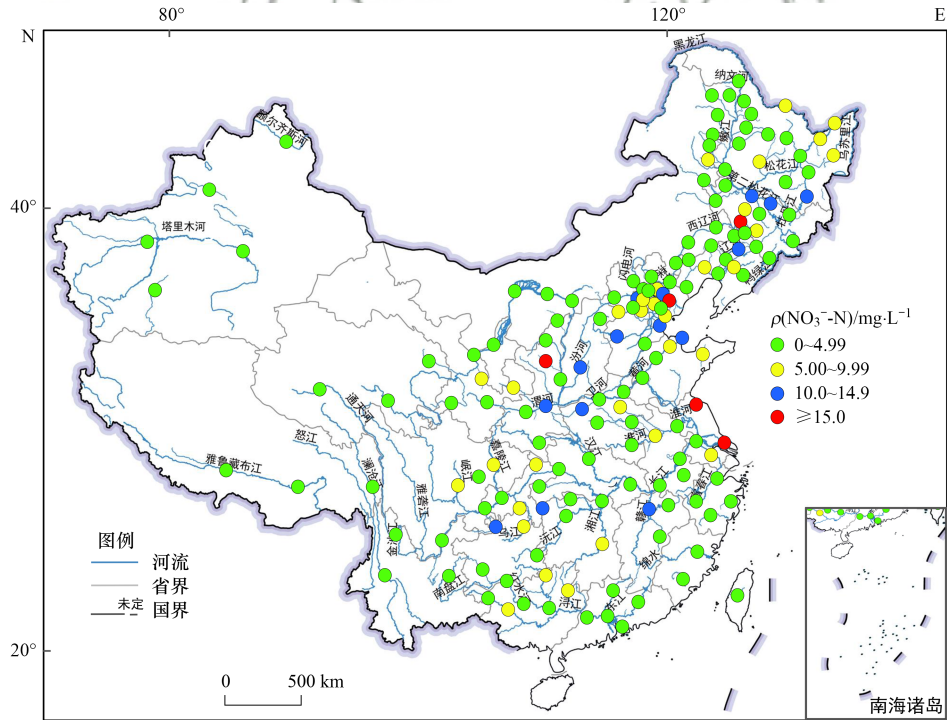
(51.4%),地表植被覆盖率高,流域对氮的同化能力较强^[46,47].同时,珠江水系河流流量较大,对氮的稀释作用较强,使得整个水系硝态氮浓度偏低.松花江流域的人类活动强度总体较弱(表1),导致其硝态氮的浓度较低^[48].



显著水平 $\alpha=0.01$, 不同大写字母表示差异显著

图1 我国主要河流水系的硝态氮浓度差异性

Fig. 1 Differences in nitrate nitrogen concentrations in major river systems in China



数据点位为各支流与干流河段代表性位置,表示该支流与干流河段硝态氮浓度的平均值

图2 我国主要河流水系硝态氮浓度的空间分布

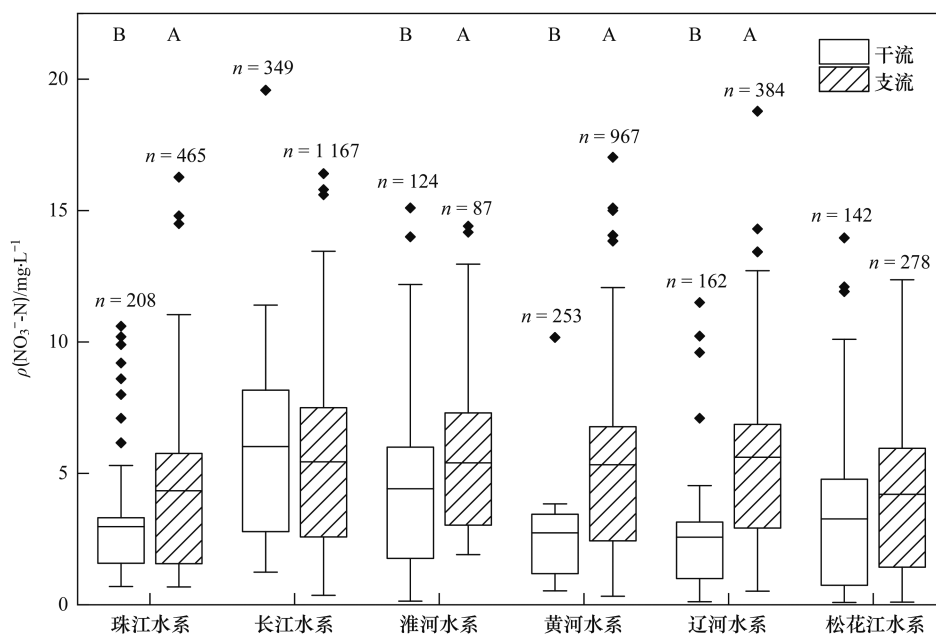
Fig. 2 Spatial distribution of nitrate nitrogen concentration in major river systems in China

我国各河流水系的支流硝态氮浓度整体上高于干流的(图3).相比干流,大多数支流需要流经人口聚集地,水体更容易受到人口密集地区生活污水和工农业废水等污染源的影响^[16,49,50].同时,由于支流的流量较小,稀释作用较弱,并且水体流速缓慢,水体交换的

整体效率较低,自我净化能力不佳^[50,51],使得水体硝态氮浓度较高,因此,需要加强对支流硝态氮污染的治理.例如,珠江水系中上游支流漓江和柳江受密集的城镇人口和旅游业以及大量工业区含氮废水排放的影响^[18,52],河流 $\rho(\text{NO}_3^--\text{N})$ 平均值分别为5.46 $\text{mg}\cdot\text{L}^{-1}$ 和

6.35 mg·L⁻¹,均高于干流硝态氮浓度平均值. 黄河水系的支流汾河,渭河和伊洛河周边人口密集,工农业生产活动频繁,河流水体受人类活动影响程度大,尤

其是受生活污水的影响^[53],使得河流硝态氮浓度超标率高. 辽河水系中游支流昭苏台河由于受工业废水直排的影响,水体 $\rho(\text{NO}_3^--\text{N})$ 高达18.7 mg·L⁻¹.



显著水平 $\alpha=0.01$, 仅表示同一水系内部干流与支流之间的差异性比较, 不同大写字母表示差异显著, n 为 $\rho(\text{NO}_3^--\text{N})$ 数据量

图3 我国主要河流水系干流与支流硝态氮浓度的差异性

Fig. 3 Differences in nitrate nitrogen concentrations between main stream and tributaries in major river systems in China

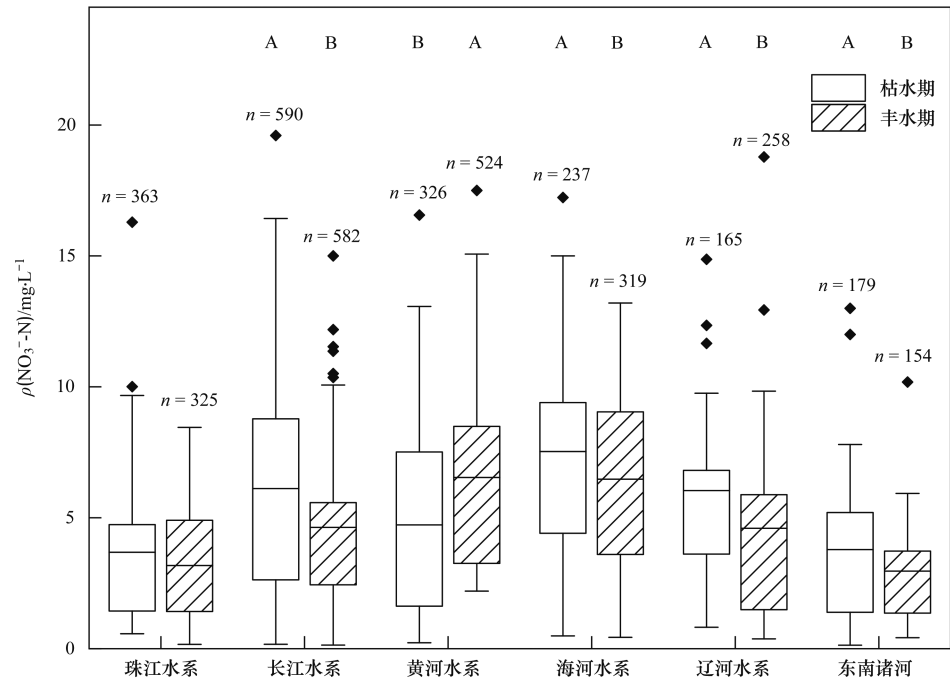
从时间变异性上看,除了黄河水系以外,其他河流水系枯水期的硝态氮浓度总体高于丰水期(图4),这主要可能是水文气候条件影响造成的. 由于我国大部分河流水系处于季风气候区,雨热同期,因此,丰水期的河流流量大导致稀释能力强,且因温度较高(20~40℃)导致反硝化作用强^[16,54],从而降低了河流硝态氮浓度. 旱季河流流量小且温度低,植被覆盖较少,降低了流域对氮的同化能力^[47],导致河流氮污染加重. 黄河流域的农用地面积较大,在雨季时农业活动频繁,水土流失严重,使得农业面源污染成为雨季时河流硝态氮污染的主要原因^[55],导致了雨季硝态氮浓度要高于旱季.

2.2 我国主要河流水系的氮转化特征分析

总体上,珠江水系、黄河水系中下游地区、辽河水系中游、松花江水系和海河水系的河流 $\delta^{18}\text{O}-\text{NO}_3^-$ 值处于硝化作用 $\delta^{18}\text{O}-\text{NO}_3^-$ 理论值范围之内(表4),表明这些河流水系可能存在显著的硝化作用. 一般地,当水体中的 NO_3^- 的 $\delta^{18}\text{O}/\delta^{15}\text{N}$ 值在0.43~1的范围内时,推断水体中发生了反硝化作用^[14,40,56]. 长江水系、淮河水系和珠江水系下游河流的 $\delta^{15}\text{N}-\text{NO}_3^-$ 与 $\delta^{18}\text{O}-\text{NO}_3^-$ 之间存在明显的线性关系(图5),且其斜率落在0.43~1范围之间,意味着存在反硝化作用. 珠江水系中游和上游地区河流水体中硝化作用较为明显,这可能是由于珠江水系中上游地区河流落差较大,整

体流速较快,水体溶解氧高(均值:8.96 mg·L⁻¹,范围:6.58~13.6 mg·L⁻¹),水温(20~35℃)也较为适合硝化细菌生长活动^[16,57,58],使得硝化作用强烈,导致珠江水系上中游地区河流(如右江、柳江和漓江等)的硝态氮浓度较高(图2). 但是,珠江水系下游的反硝化作用较为强烈^[18,59],使得硝态氮浓度较低;黄河水系、海河水系、辽河水系和松花江水系流域的土壤整体偏碱性,较高的pH可能会抑制反硝化微生物对营养物质的吸收及其酶的活性,而弱碱性环境则更适合硝化细菌的生长^[28,60],使得硝化作用较强,这也可能是黄河水系、海河水系和辽河水系硝态氮浓度较高的原因之一. 长江水系和淮河水系的有机物质含量高^[18,28,31]且溶解氧浓度较低(长江均值:5.98 mg·L⁻¹,范围:1.04~11.6 mg·L⁻¹;淮河均值:6.10 mg·L⁻¹,范围:0.34~13.2 mg·L⁻¹),从而一定程度上抑制硝化作用而促进反硝化作用^[31,61].

从不同水文期的氮转化过程看,我国主要河流水系的大部分河流水体在枯水期时硝化作用强烈,且主要发生在中下游的支流地区,而部分河流丰水期时反硝化作用强烈(图5). 显然,我国大部分河流水系枯水期时生活和生产的污水排放等点源污染对水体硝态氮的影响较大(图8),而点源污染以铵态氮为主,往往导致河流水体中的硝化作用强烈^[36,62],同时,枯水期的低水温抑制了反硝化作用,而丰水期较



显著水平 $\alpha=0.01$,仅表示同一水系内部枯水期和丰水期之间的差异性比较,不同大写字母表示差异显著, n 为 $\rho(\text{NO}_3\text{-N})$ 数据量

图4 我国主要河流水系硝态氮浓度的季节性差异

Fig. 4 Seasonal differences in nitrate nitrogen concentrations in major river systems in China

高的温度导致部分河流反硝化作用强烈^[16,54],总体上看,我国大部分河流水系存在强烈的硝化作用,部分河流水系的反硝化作用明显.但是,由于影响硝态氮转化过程及其动态变化的机制十分复杂,特别是外源氮输入(如化肥、污水/粪便)的混合作用可能会掩盖同位素的反硝化信号^[63],密集的降雨,稀释和混合过程也可能掩盖同位素的硝化与反硝化信号^[35,64].因此,未来需要进一步结合水化学数据与氮转化相关微生物基因丰度等指标,以提高对水体硝化与反硝化作用强度识别结果的可靠性.

2.3 我国主要河流水系的硝态氮定量源解析

不同污染源对我国各大河流水系硝态氮的贡献率存在较大的差异(图6).珠江水系和黄河水系硝态氮的主要来源较为复杂,为污水/粪肥、土壤氮以及化肥的混合.污水/粪肥是珠江水系硝态氮的主要贡献来源(平均贡献率为36.3%),土壤氮和化肥的平均

贡献率分别为26.2%和20.8%.黄河水系的硝态氮主要来源于土壤氮(平均贡献率为37.8%),污水/粪肥和化肥的平均贡献率分别为30.3%和25.6%;淮河水系的硝态氮主要来源于污水/粪肥(平均贡献率为40.0%)和化肥(平均贡献率为32.4%);污水/粪肥是长江水系、海河水系、辽河水系以及东南诸河水系硝态氮的主要来源,其平均贡献率超过50%;松花江水系硝态氮主要来源于土壤氮源(平均贡献率为56.4%).以往的研究表明,海河、辽河水系河流硝态氮主要来源于污水/粪肥,而珠江、长江和黄河水系的硝态氮来源于污水/粪肥和土壤氮,松花江水系的硝态氮主要来源于土壤氮、化肥和污水/粪肥的混合^[16,19,20,28].与以往的研究相比,本研究结果与已有的研究结果虽有相同之处,但仍然存在不小的差异,这可能是由研究数据量,以及所选取的硝态氮潜在来源特征值的不同造成的.总体而言,我国河流水系硝

表4 我国主要河流水系水体硝化作用 $\delta^{18}\text{O-NO}_3^-$ 理论值与实测值¹⁾

Table 4 Theoretical and measured values of $\delta^{18}\text{O-NO}_3^-$ of nitrification in major river systems in China

水系	$\delta^{18}\text{O-NO}_3^-$ 理论值范围 ‰	$\delta^{18}\text{O-NO}_3^-$ 实测值范围 ‰	$\delta^{18}\text{O-NO}_3^-$ 实测值所占 理论值比例/%	文献
珠江	1.00~4.80	-7.49~73.0	67.5	[16,59]
长江	0.80~3.05	-13.1~53.73	34.1	[28,65~67]
淮河	1.14~6.46	-8.91~34.2	45.6	[31]
黄河	1.20~4.20	-21.4~55.8	52.3	[32,33]
海河	1.20~5.70	-17.6~39.2	65.7	[34,35,43]
辽河	1.50~5.40	-10.3~28.2	48.2	[14,36]
松花江	-2.66~5.38	-6.80~51.9	52.4	[39,48]

1) $\delta^{18}\text{O-NO}_3^-$ 理论值范围为已有研究中整理归纳得到的

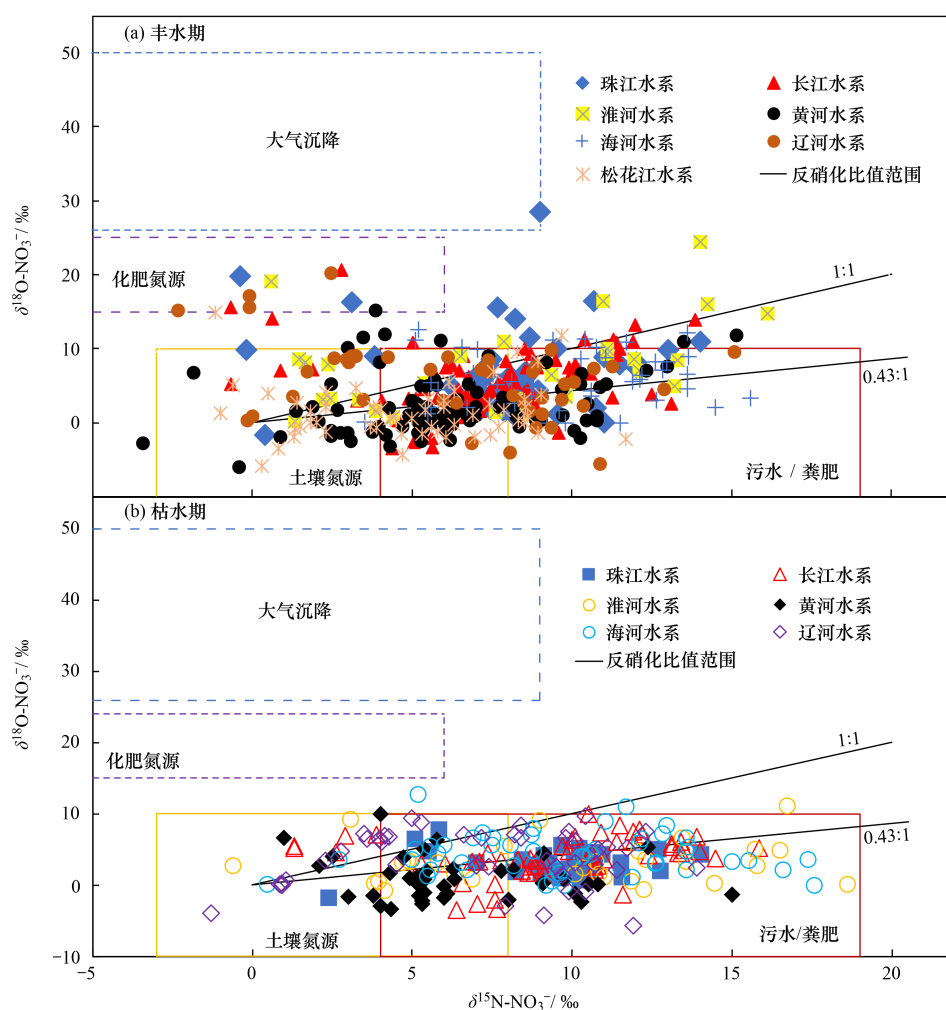


图5 不同氮源的 $\delta^{15}\text{N}-\text{NO}_3^-$ 和 $\delta^{18}\text{O}-\text{NO}_3^-$ 范围及我国主要河流水系丰水期和枯水期硝态氮的 $\delta^{15}\text{N}-\text{NO}_3^-$ 和 $\delta^{18}\text{O}-\text{NO}_3^-$ 特征值
Fig. 5 Ranges of $\delta^{15}\text{N}-\text{NO}_3^-$ and $\delta^{18}\text{O}-\text{NO}_3^-$ of different nitrogen sources and the characteristic values of $\delta^{15}\text{N}-\text{NO}_3^-$ and $\delta^{18}\text{O}-\text{NO}_3^-$ of nitrate nitrogen in major river systems in China during the wet season and dry season

态氮污染的主要来源包括污水/粪肥、土壤氮和化肥,但不同河流水系硝态氮的来源存在较大的差异性,这主要由于人类活动(如废水排放和施肥等)和自然要素影响.长江、海河、辽河和东南诸河流域的人口密度较大,工农业比较发达,畜禽养殖密度大,工业废水和生活污水排放强度较大(表1),使得污水/粪肥的点源污染对河流硝态氮污染的影响较大^[20,68,69],而点源污染以铵态氮为主,往往导致河流水体中的硝化作用强烈^[36],这也可能是海河水系、辽河水系和黄河水系中下游地区河流水体硝化作用强烈的原因.因此,长江、海河、辽河和东南诸河水系硝态氮污染的治理重点是控制生活和生产污水等点源污染,应加强完善城镇废水收集和集中处理系统建设,提高污水处理效率等^[70,71].淮河、黄河和松花江流域的农用地面积占比高,化肥施用量大,土壤中含氮量高^[55,72,73],使得土壤氮和化肥成为河流硝态氮的主要来源,因此,硝态氮污染的防治重点是控制化肥和土壤氮等流失造成的非点源污染,一方面可以

通过增加植被覆盖,减少农田径流,控制水土流失,另一方面要减少化肥氮施用,提高化肥利用率,减少农田氮非点源污染负荷^[55,74].

不同水系的干流与支流硝态氮来源组成存在较大差异(图7),即污水/粪肥对支流硝态氮贡献率总体大于干流的,土壤氮对干流硝态氮的贡献总体大于支流的.这是由于支流相较于干流来说流量较小,水体的稀释能力和自净能力相对较差,支流水体更容易受到密集人类生活活动以及大量工业废水排放影响,导致水体硝态氮的来源中污水/粪肥的贡献占比更高^[16,41,49].干流的高流量可能导致可观的土壤和化肥氮流失等面源污染^[75],但对生活和工业等点源污染具有较强的稀释作用.即点源污染对支流的作用信号较强,而对于干流的作用信号较弱.例如,土壤氮、化肥和污水/粪肥等三大污染源对珠江水系干流硝态氮的贡献较为接近(29.4%、22.2%和29.6%),但是污水/粪肥对支流的平均贡献率高达45.8%;污水/粪肥对长江水系干流硝态氮平均贡献为44.2%,而对支

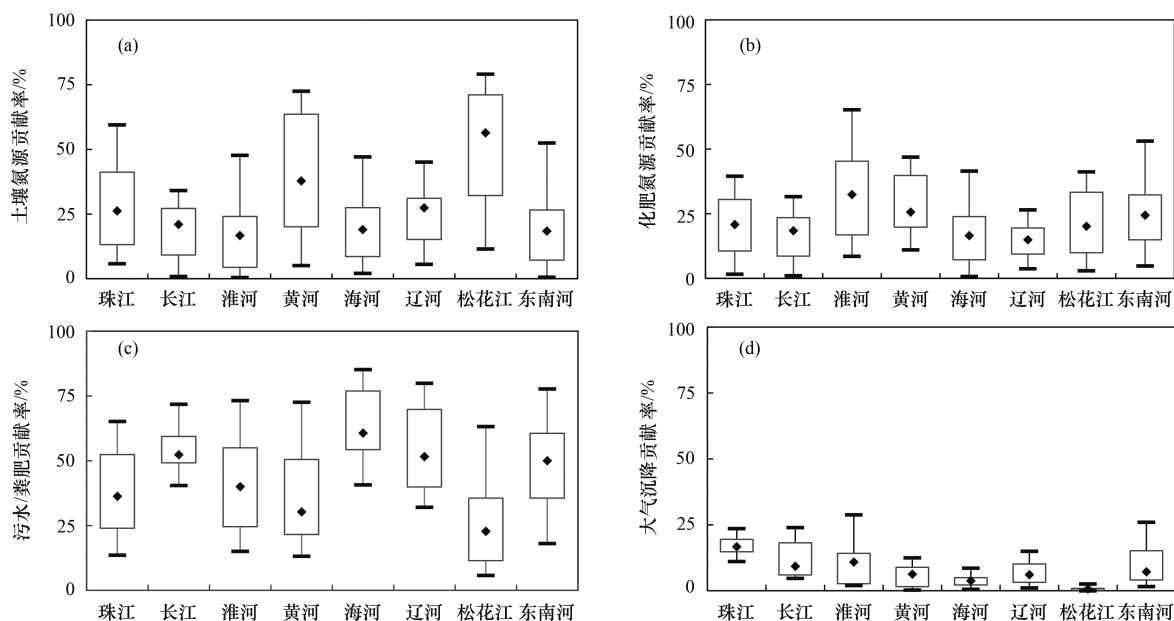


图6 我国主要河流水系硝态氮不同污染来源的贡献率

Fig. 6 Contribution of different sources to nitrate nitrogen pollution in major river systems in China

流的平均贡献率高达 59.6%。土壤氮对黄河水系干流硝态氮的平均贡献率为 42.4%,但对支流的贡献仅为 31.1%。与其他河流水系不同,污水/粪肥对松花江水系干流硝态氮的贡献率超过支流的,而土壤氮对支流的贡献率超过干流的,这主要是由于松花江水系支流的集水区森林覆盖度较高,而干流城镇密集,更容易受人类生产生活的影响^[20,48]。

不同水文期的水系硝态氮来源组成也存在较大差异(图8),丰水期土壤氮、化肥和大气沉降对河流硝态氮的贡献率高于枯水期,而枯水期污水/粪肥的贡献率高于丰水期。显然,土壤氮、化肥氮和大气沉降氮主要通过非点源污染形式作用于河流,因此,由于丰水期的降雨径流大,使得土壤氮、化肥氮和大气沉降氮的非点源污染负荷显著大于枯水期的^[54,76]。同时,由于雨热同期,丰水期的农业耕作活动强烈,使得农业非点源氮污染负荷大于枯水期的^[75]。因此,丰水期硝态氮的治理应重点考虑土壤氮和化肥氮等造

成的非点源污染治理。相反地,枯水期的非点源污染较弱,工业和生活等污水排放的点源污染影响显著增加^[19,77],因此,河流硝态氮污染的治理应重点控制生活和生产污水排放等点源污染。

尽管采用氮氧同位素方法定量识别了我国主要河流水系的来源组成,但是不可避免地存在较大不确定性。首先,由于河流硝态氮的来源和转化具有显著的时空变异性,因此,收集的氮氧同位素数据主要来源于低频采样,这可能会错过一些关键的同位素信息^[16];其次,不同氮源的氮氧同位素特征值区域存在一定的重叠,且不同地区的潜在硝态氮来源具有不同的氮氧同位素特征值^[14],这也可能导致源识别结果的不确定性;最后,硝态氮在陆域和水域的输移过程中存在同位素分馏效应,并且不同河流水系的分馏效应存在差异,从而可能导致源识别结果的不确定性^[44]。因此,未来应该加强对硝态氮污染较严重河流的氮氧同位素监测,协同开展氮源的同位素特

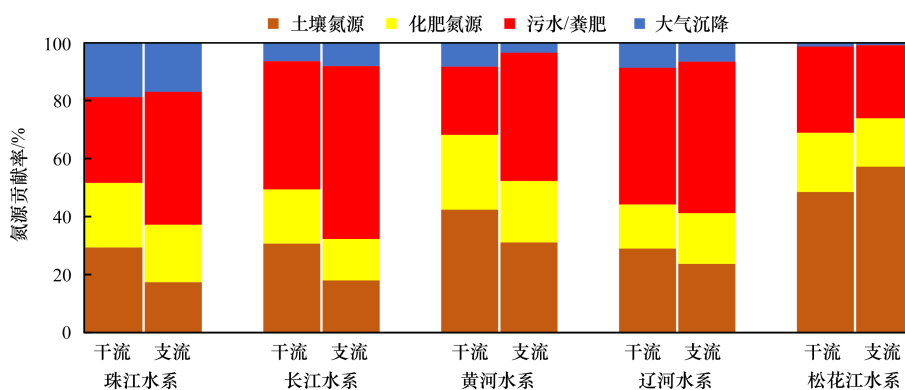


图7 我国主要河流水系干流与支流硝态氮不同污染来源的贡献率

Fig. 7 Contribution of different sources to nitrate nitrogen pollution in main stream and tributaries in major river systems in China

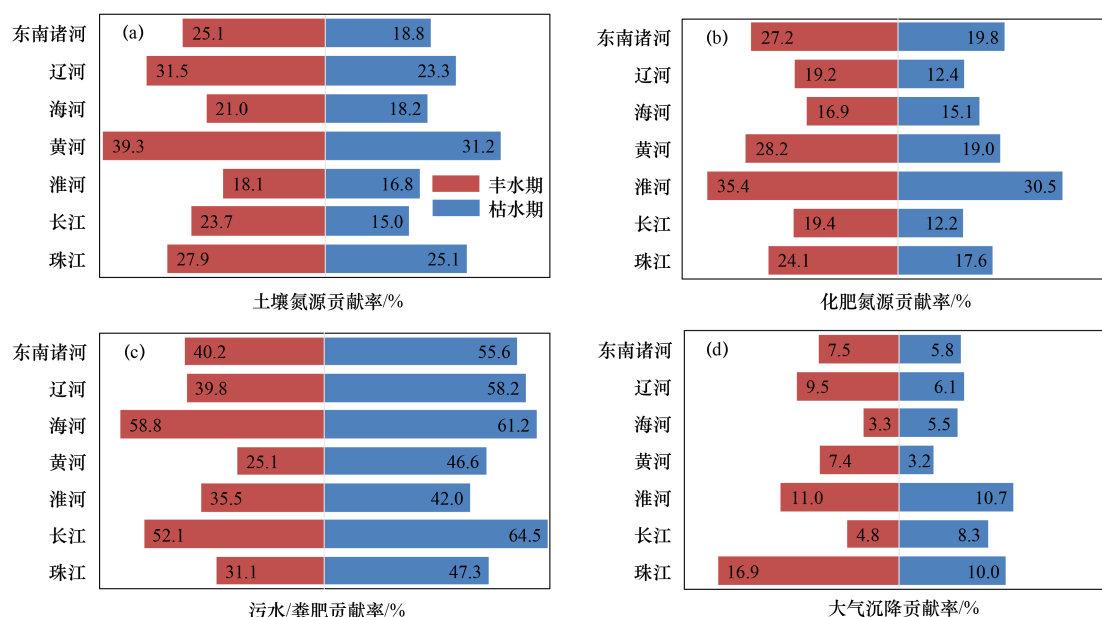


图8 我国主要河流水系硝态氮不同污染来源贡献率的季节性差异

Fig. 8 Seasonal differences in contributions of different sources to nitrate nitrogen pollution in major river systems in China

征值分析,构建全国河流水系氮氧同位素数据库,支撑河流氮污染科学研究.同时,将MixSIAR混合模型与多元统计方法和机器学习法等结合,提高河流硝态氮来源识别的精度;也可以与多同位素(如 $\delta^{15}\text{N}$)和微生物源跟踪标记(粪便指示菌等)等方法相结合^[19,45],进一步加强对基于氮氧同位素的溯源结果验证.

3 结论

(1) 2000~2022年期间,我国主要河流水系的 $\rho(\text{NO}_3\text{-N})$ 平均值为 $(4.54 \pm 3.99) \text{ mg} \cdot \text{L}^{-1}$,超标率为9.6%,长江、淮河、黄河、海河和辽河水系河流硝态氮浓度较高,需重点治理.各水系硝态氮浓度呈现出明显的时空变异性,支流水体硝态氮浓度整体上高于干流水体,而除黄河水系以外,其他河流水系枯水期的硝态氮浓度总体高于丰水期.

(2) 珠江水系、黄河水系中下游地区、辽河水系中游地区、松花江水系和海河水系河流水体存在显著的硝化作用;而长江水系、淮河水系和珠江水系下游地区河流水体存在反硝化作用;大部分河流水体在枯水期时存在显著的硝化作用,而部分河流水系丰水期时存在反硝化作用.

(3) 珠江水系和黄河水系硝态氮主要来源于污水/粪肥、土壤氮和化肥,淮河水系硝态氮主要来源于污水/粪肥和化肥,污水/粪肥是长江、海河、辽河和东南诸河水系硝态氮的主要来源,土壤氮是松花江水系硝态氮的主要来源.污水/粪肥对支流硝态氮的贡献率总体大于干流的,土壤氮对干流硝态氮的

贡献总体大于支流的.丰水期土壤氮、化肥和大气沉降对河流硝态氮的贡献率高于枯水期,而枯水期污水/粪肥的贡献率高于丰水期的.

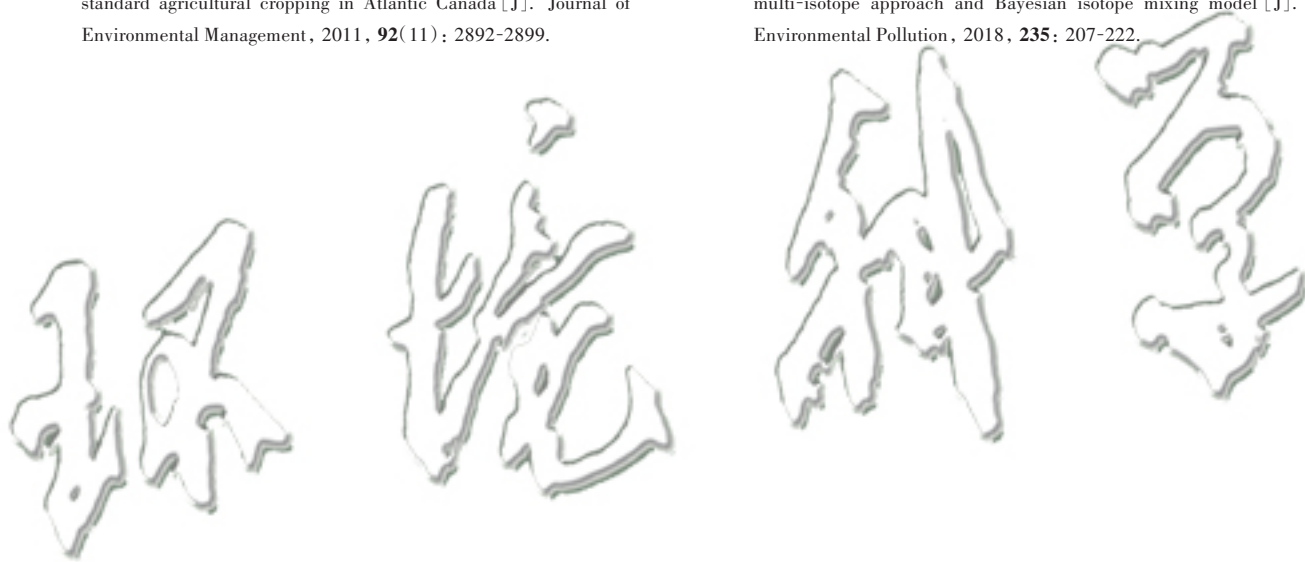
参考文献:

- [1] Niu C, Zhai T L, Zhang Q Q, *et al.* Research advances in the analysis of nitrate pollution sources in a freshwater environment using $\delta^{15}\text{N}\text{-NO}_3^-$ and $\delta^{18}\text{O}\text{-NO}_3^-$ [J]. *International Journal of Environmental Research and Public Health*, 2021, **18** (22), doi: 10.3390/ijerph182211805.
- [2] Wu S J, Tetzlaff D, Yang X Q, *et al.* Disentangling the influence of landscape characteristics, hydroclimatic variability and land management on surface water $\text{NO}_3\text{-N}$ dynamics: spatially distributed modeling over 30 yr in a lowland mixed land use catchment [J]. *Water Resources Research*, 2022, **58** (2), doi: 10.1029/2021WR030566.
- [3] Meghdadi A, Javar N. Quantification of spatial and seasonal variations in the proportional contribution of nitrate sources using a multi-isotope approach and Bayesian isotope mixing model [J]. *Environmental Pollution*, 2018, **235**: 207-222.
- [4] Ma P, Zhang L, Mitsch W J. Investigating sources and transformations of nitrogen using dual stable isotopes for Lake Okeechobee restoration in Florida [J]. *Ecological Engineering*, 2020, **155**, doi: 10.1016/j.ecoleng.2020.105947.
- [5] Cui Y H, Wang J, Hao S. Spatial variability of nitrate pollution and its sources in a hilly basin of the Yangtze River based on clustering [J]. *Scientific Reports*, 2021, **11** (1), doi: 10.1038/s41598-021-96248-0.
- [6] Ma T, Zhao N, Ni Y, *et al.* China's improving inland surface water quality since 2003 [J]. *Science Advances*, 2020, **6** (1), doi: 10.1126/sciadv. aau3798.
- [7] Stets E G, Sprague L A, Oelsner G P, *et al.* Landscape drivers of dynamic change in water quality of U. S. rivers [J]. *Environmental Science & Technology*, 2020, **54** (7): 4336-4343.
- [8] Chen D J, Hu M P, Dahlgren R A. A dynamic watershed model for determining the effects of transient storage on nitrogen export to rivers [J]. *Water Resources Research*, 2014, **50** (10): 7714-

- 7730.
- [9] Zhou J, Hu M P, Liu M, *et al.* Combining the multivariate statistics and dual stable isotopes methods for nitrogen source identification in coastal rivers of Hangzhou Bay, China [J]. *Environmental Science and Pollution Research*, 2022, **29**(55): 82903-82916.
- [10] Borah D K, Bera M. Watershed-scale hydrologic and nonpoint-source pollution models: review of applications[J]. *Transactions of the ASAE*, 2004, **47**(3): 789-803.
- [11] Hu M P, Yao M Y, Wang Y C, *et al.* Influence of nitrogen inputs, dam construction and landscape patterns on riverine nitrogen exports in the Yangtze River basin during 1980-2015[J]. *Journal of Hydrology*, 2023, 617, doi: 10.1016/j.jhydrol.2023.129109.
- [12] Yi Q T, Zhang Y, Xie K, *et al.* Tracking nitrogen pollution sources in plain watersheds by combining high-frequency water quality monitoring with tracing dual nitrate isotopes[J]. *Journal of Hydrology*, 2020, 581, doi: 10.1016/j.jhydrol.2019.124439.
- [13] 芮孝芳. 论流域水文模型[J]. *水利水电科技进展*, 2017, **37**(4): 1-7, 58.
- Rui X F. Discussion of watershed hydrological model [J]. *Advances in Science and Technology of Water Resources*, 2017, **37**(4): 1-7, 58.
- [14] Xue D M, Botte J, De Baets B, *et al.* Present limitations and future prospects of stable isotope methods for nitrate source identification in surface- and groundwater [J]. *Water Research*, 2009, **43**(5): 1159-1170.
- [15] Panno S V, Hackley K C, Kelly W R, *et al.* Isotopic evidence of nitrate sources and denitrification in the Mississippi River, Illinois [J]. *Journal of Environmental Quality*, 2006, **35**(2): 495-504.
- [16] Li C, Li S L, Yue F J, *et al.* Identification of sources and transformations of nitrate in the Xijiang River using nitrate isotopes and Bayesian model[J]. *Science of the Total Environment*, 2019, **646**: 801-810.
- [17] Xue D M, de Baets B, van Cleemput O, *et al.* Use of a Bayesian isotope mixing model to estimate proportional contributions of multiple nitrate sources in surface water [J]. *Environmental Pollution*, 2012, **161**: 43-49.
- [18] 孙文青, 陆光华, 薛晨旺, 等. 基于稳定同位素技术识别河流硝酸盐污染源研究进展[J]. *四川环境*, 2019, **38**(3): 193-198.
- Sun W Q, Lu G H, Xue C W, *et al.* Research progress on identification of nitrate pollution sources in rivers by using stable isotope technique [J]. *Sichuan Environment*, 2019, **38**(3): 193-198.
- [19] 张金兰, 蒯祖弘, 白文荣, 等. 利用整合分析方法探究我国不同土地利用类型区域河流硝酸盐的来源[J]. *农业资源与环境学报*, 2021, **38**(5): 746-754.
- Zhang J L, Lin Z H, Bai W R, *et al.* Sources of nitrate in rivers under different land-use types in China: a meta-analysis [J]. *Journal of Agricultural Resources and Environment*, 2021, **38**(5): 746-754.
- [20] Zhang X, Zhang Y, Shi P, *et al.* The deep challenge of nitrate pollution in river water of China [J]. *Science of the Total Environment*, 2021, 770, doi: 10.1016/j.scitotenv.2020.144674.
- [21] Stock B C, Semmens B X. Unifying error structures in commonly used biotracer mixing models[J]. *Ecology*, 2016, **97**(10): 2562-2569.
- [22] Stock B C, Jackson A L, Ward E J, *et al.* Analyzing mixing systems using a new generation of Bayesian tracer mixing models [J]. *PeerJ*, 2018, 6, doi: 10.7717/peerj.5096.
- [23] Zhang M, Zhi Y Y, Shi J C, *et al.* Apportionment and uncertainty analysis of nitrate sources based on the dual isotope approach and a Bayesian isotope mixing model at the watershed scale[J]. *Science of the Total Environment*, 2018, **639**: 1175-1187.
- [24] Stock B C, Semmens B. MixSIAR: Bayesian mixing models in R, version 3.1.12 [EB/OL]. <https://cran.r-project.org/web/packages/MixSIAR/index.html>, 2023-04-09.
- [25] Wang M, Lu B H, Wang J Q, *et al.* Using dual isotopes and a Bayesian isotope mixing model to evaluate nitrate sources of surface water in a drinking water source watershed, East China [J]. *Water*, 2016, **8**(8), doi: 10.3390/w8080355.
- [26] Liu C Q, Li S L, Lang Y C, *et al.* Using $\delta^{15}\text{N}$ - and $\delta^{18}\text{O}$ - values to identify nitrate sources in Karst ground water, Guiyang, Southwest China [J]. *Environmental Science & Technology*, 2006, **40**(22): 6928-6933.
- [27] Yue F J, Li S L, Liu C Q, *et al.* Sources and transport of nitrate constrained by the isotopic technique in a karst catchment: an example from Southwest China [J]. *Hydrological Processes*, 2015, **29**(8): 1883-1893.
- [28] Li S L, Liu C Q, Li J, *et al.* Assessment of the sources of nitrate in the Changjiang River, China using a nitrogen and oxygen isotopic approach [J]. *Environmental Science & Technology*, 2010, **44**(5): 1573-1578.
- [29] Zhong X S, Yan M J, Ning X Y, *et al.* Nitrate processing traced by nitrate dual isotopic composition in the early spring in the Changjiang (Yangtze River) Estuary and adjacent shelf areas [J]. *Marine Pollution Bulletin*, 2020, 161, doi: 10.1016/j.marpolbul.2020.111699.
- [30] Xia X H, Li S L, Wang F, *et al.* Triple oxygen isotopic evidence for atmospheric nitrate and its application in source identification for river systems in the Qinghai-Tibetan Plateau [J]. *Science of the Total Environment*, 2019, **688**: 270-280.
- [31] Ma P, Liu S X, Yu Q B, *et al.* Sources and transformations of anthropogenic nitrogen in the highly disturbed Huai River Basin, eastern China [J]. *Environmental Science and Pollution Research*, 2019, **26**(11): 11153-11169.
- [32] Liu T, Wang F, Michalski G, *et al.* Using ^{15}N , ^{17}O , and ^{18}O to determine nitrate sources in the Yellow River, China [J]. *Environmental Science & Technology*, 2013, **47**(23): 13412-13421.
- [33] Yue F J, Li S L, Liu C Q, *et al.* Tracing nitrate sources with dual isotopes and long term monitoring of nitrogen species in the Yellow River, China [J]. *Scientific Reports*, 2017, **7**(1), doi: 10.1038/s41598-017-08756-7.
- [34] Xue D, Li J, Wang Y, *et al.* Nitrate source distribution in rivers, estuaries and groundwater using a dual isotope approach and a Bayesian isotope mixing model [J]. *Applied Ecology and Environmental Research*, 2020, **18**(3): 4651-4668.
- [35] Liu J, Shen Z Y, Yan T Z, *et al.* Source identification and impact of landscape pattern on riverine nitrogen pollution in a typical urbanized watershed, Beijing, China [J]. *Science of the Total Environment*, 2018, **628-629**: 1296-1307.
- [36] Yue F J, Li S L, Liu C Q, *et al.* Using dual isotopes to evaluate sources and transformation of nitrogen in the Liao River, Northeast China [J]. *Applied Geochemistry*, 2013, **36**: 1-9.
- [37] Yue F J, Liu C Q, Li S L, *et al.* Analysis of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ to identify nitrate sources and transformations in Songhua River, Northeast China [J]. *Journal of Hydrology*, 2014, **519**: 329-339.
- [38] Huang H, Liu M Z, Wang J J, *et al.* Sources identification of

- nitrogen using major ions and isotopic tracers in Shenyang, China [J]. *Geofluids*, 2018, **2018**, 10.1155/2018/8683904.
- [39] Zhang H Y, Yang Y S, Zou J Y, *et al.* The sources and dispersal of nitrate in multiple waters, constrained by multiple isotopes, in the Wudalianchi region, Northeast China [J]. *Environmental Science and Pollution Research*, 2018, **25**(24): 24348-24361.
- [40] Hu M P, Liu Y M, Zhang Y F, *et al.* Coupling stable isotopes and water chemistry to assess the role of hydrological and biogeochemical processes on riverine nitrogen sources [J]. *Water Research*, 2019, **150**: 418-430.
- [41] Shang X, Huang H, Mei K, *et al.* Riverine nitrate source apportionment using dual stable isotopes in a drinking water source watershed of Southeast China [J]. *Science of the Total Environment*, 2020, **724**, doi: 10.1016/j.scitotenv.2020.137975.
- [42] 王婧宇. 应用氮氧同位素以及同位素模型识别天津水体中硝酸盐的潜在来源[D]. 天津: 天津师范大学, 2016.
Wang J Y. Nitrate sources apportionment in surface water and shallow groundwater in Tianjin (China) using a dual isotope approach and a Bayesian isotope mixing model [D]. Tianjin: Tianjin Normal University, 2016.
- [43] Li C, Jiang Y B, Guo X Y, *et al.* Multi-isotope (^{15}N , ^{18}O and ^{13}C) indicators of sources and fate of nitrate in the upper stream of Chaobai River, Beijing, China [J]. *Environmental Science: Processes & Impacts*, 2014, **16**(11): 2644-2655.
- [44] Li Z, Xiao J, Evaristo J, *et al.* Spatiotemporal variations in the hydrochemical characteristics and controlling factors of streamflow and groundwater in the Wei River of China [J]. *Environmental Pollution*, 2019, **254**, doi: 10.1016/j.envpol.2019.113006.
- [45] Carrey R, Ballesté E, Blanch A R, *et al.* Combining multi-isotopic and molecular source tracking methods to identify nitrate pollution sources in surface and groundwater [J]. *Water Research*, 2021, **188**, doi: 10.1016/j.watres.2020.116537.
- [46] Valiente N, Carrey R, Otero N, *et al.* A multi-isotopic approach to investigate the influence of land use on nitrate removal in a highly saline lake-aquifer system [J]. *Science of the Total Environment*, 2018, **631-632**: 649-659.
- [47] Weber G, Honecker U, Kubiniok J. Nitrate dynamics in springs and headwater streams with agricultural catchments in southwestern Germany [J]. *Science of the Total Environment*, 2020, **722**, doi: 10.1016/j.scitotenv.2020.137858.
- [48] Yue F J, Liu C Q, Li S L, *et al.* Analysis of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ to identify nitrate sources and transformations in Songhua River, Northeast China [J]. *Journal of Hydrology*, 2014, **519**: 329-339.
- [49] Zhen S C, Zhu W. Analysis of isotope tracing of domestic sewage sources in Taihu Lake—A case study of Meiliang Bay and Gonghu Bay [J]. *Ecological Indicators*, 2016, **66**: 113-120.
- [50] 李雨桓, 韦盼, 黄蓁, 等. 我国地表水环境质量现状及污染修复技术研究 [J]. *中国资源综合利用*, 2021, **39**(2): 195-197.
Li Y H, Wei P, Huang Z, *et al.* Research on the status quo of surface water environmental quality and pollution remediation technology [J]. *China Resources Comprehensive Utilization*, 2021, **39**(2): 195-197.
- [51] Yao Y Z, Tian H Q, Shi H, *et al.* Increased global nitrous oxide emissions from streams and rivers in the Anthropocene [J]. *Nature Climate Change*, 2020, **10**(2): 138-142.
- [52] Kendall C, Elliott E M, Wankel S D. Tracing anthropogenic inputs of nitrogen to ecosystems [A]. In: Michener R, Lajtha K (Eds.). *Stable Isotopes in Ecology and Environmental Science* (2nd ed.) [M]. Malden: Blackwell Pub, 2007. 375-449.
- [53] Shi P, Zhang Y, Song J X, *et al.* Response of nitrogen pollution in surface water to land use and social-economic factors in the Weihe River watershed, Northwest China [J]. *Sustainable Cities and Society*, 2019, **50**, doi: 10.1016/j.scs.2019.101658.
- [54] Jiang H, Liu W J, Zhang J Y, *et al.* Spatiotemporal variations of nitrate sources and dynamics in a typical agricultural riverine system under monsoon climate [J]. *Journal of Environmental Sciences*, 2020, **93**: 98-108.
- [55] Wang W J, Song X F, Ma Y. Identification of nitrate source using isotopic and geochemical data in the lower reaches of the Yellow River irrigation district (China) [J]. *Environmental Earth Sciences*, 2016, **75**(11), doi: 10.1007/s12665-016-5721-3.
- [56] Kool D M, Wragge N, Oenema O, *et al.* Oxygen exchange with water alters the oxygen isotopic signature of nitrate in soil ecosystems [J]. *Soil Biology and Biochemistry*, 2011, **43**(6): 1180-1185.
- [57] 赵文博, 解永新, 于英潭, 等. 不同生态复氧方式对城市河流溶解氧影响研究 [J]. *环境生态学*, 2019, **1**(3): 61-66.
Zhao W B, Xie Y X, Yu Y T, *et al.* Study on the influence of different ecological reoxygenation modes on dissolved oxygen in urban rivers [J]. *Environmental Ecology*, 2019, **1**(3): 61-66.
- [58] 苗迎, 章程, 肖琼, 等. 漓江段地表水体旱季硝酸盐动态变化特征及其来源 [J]. *环境科学*, 2018, **39**(4): 1589-1597.
Miao Y, Zhang C, Xiao Q, *et al.* Dynamic variations and sources of nitrate during dry season in the Lijiang river [J]. *Environmental Science*, 2018, **39**(4): 1589-1597.
- [59] Xuan Y X, Tang C Y, Cao Y J. Mechanisms of nitrate accumulation in highly urbanized rivers: evidence from multi-isotopes in the Pearl River Delta, China [J]. *Journal of Hydrology*, 2020, **587**, doi: 10.1016/j.jhydrol.2020.124924.
- [60] 康丽娟, 许海, 朱广伟, 等. 太湖主要环湖河道沉积物反硝化潜力及其控制因子 [J]. *环境科学学报*, 2021, **41**(4): 1393-1400.
Kang L J, Xu H, Zhu G W, *et al.* Sediment denitrification potential and its influencing factors in the main rivers of Lake Taihu [J]. *Acta Scientiae Circumstantiae*, 2021, **41**(4): 1393-1400.
- [61] Rivett M O, Buss S R, Morgan P, *et al.* Nitrate attenuation in groundwater: a review of biogeochemical controlling processes [J]. *Water Research*, 2008, **42**(16): 4215-4232.
- [62] Xia X H, Zhang S B, Li S L, *et al.* The cycle of nitrogen in river systems: sources, transformation, and flux [J]. *Environmental Science: Processes & Impacts*, 2018, **20**(6): 863-891.
- [63] Yi Q T, Chen Q W, Hu L M, *et al.* Tracking nitrogen sources, transformation, and transport at a basin scale with complex plain river networks [J]. *Environmental Science & Technology*, 2017, **51**(10): 5396-5403.
- [64] Xia Y Q, Li Y F, Zhang X Y, *et al.* Nitrate source apportionment using a combined dual isotope, chemical and bacterial property, and Bayesian model approach in river systems [J]. *Journal of Geophysical Research: Biogeosciences*, 2017, **122**(1): 2-14.
- [65] Zhao Y Y, Zheng B H, Jia H F, *et al.* Determination sources of nitrates into the Three Gorges Reservoir using nitrogen and oxygen isotopes [J]. *Science of the Total Environment*, 2019, **687**: 128-136.
- [66] Xu Y Y, Yuan Q Q, Zhao C F, *et al.* Identification of nitrate sources in rivers in a complex catchment using a dual isotopic approach [J]. *Water*, 2021, **13**(1), doi: 10.3390/w13010083.
- [67] Chen X J, Strokal M, Kroeze C, *et al.* Modeling the contribution of crops to nitrogen pollution in the Yangtze River [J]. *Environmental Science & Technology*, 2020, **54**(19): 11929-11939.

- [68] 马林, 卢洁, 赵浩, 等. 中国硝酸盐脆弱区划分与面源污染阻控[J]. 农业环境科学学报, 2018, **37**(11): 2387-2391.
Ma L, Lu J, Zhao H, *et al.* Nitrate vulnerable zones and strategies of non-point pollution mitigation in China [J]. *Journal of Agro-Environment Science*, 2018, **37**(11): 2387-2391.
- [69] Ye F, Ni Z X, Xie L H, *et al.* Isotopic evidence for the turnover of biological reactive nitrogen in the Pearl River Estuary, South China [J]. *Journal of Geophysical Research: Biogeosciences*, 2015, **120**(4): 661-672.
- [70] 赵楠芳, 李荣昉, 胡春华. 鄱阳湖地表水硝酸盐时空变异性及其来源研究[J]. 环境科学与技术, 2014, **37**(8): 93-98.
Zhao N F, Li R F, Hu C H, *et al.* Spatial and temporal variability and sources of nitrate in surface water in Poyang Lake [J]. *Environmental Science & Technology*, 2014, **37**(8): 93-98.
- [71] Xuan Y X, Tang C Y, Cao Y J. Mechanisms of nitrate accumulation in highly urbanized rivers: evidence from multi-isotopes in the Pearl River Delta, China[J]. *Journal of Hydrology*, 2020, **587**, doi: 10.1016/j.jhydrol.2020.124924.
- [72] Smith E L, Kellman L M. Examination of nitrate concentration, loading and isotope dynamics in subsurface drainage under standard agricultural cropping in Atlantic Canada [J]. *Journal of Environmental Management*, 2011, **92**(11): 2892-2899.
- [73] Xing M, Liu W G, Wang Z F, *et al.* Relationship of nitrate isotopic character to population density in the Loess Plateau of Northwest China[J]. *Applied Geochemistry*, 2013, **35**: 110-119.
- [74] 王响玲, 宋柏权. 氮肥利用率的研究进展[J]. 中国农学通报, 2020, **36**(5): 93-97.
Wang X L, Song B Q. Nitrogen fertilizer use efficiency: research progress [J]. *Chinese Agricultural Science Bulletin*, 2020, **36**(5): 93-97.
- [75] Liu S S, Wu F C, Feng W Y, *et al.* Using dual isotopes and a Bayesian isotope mixing model to evaluate sources of nitrate of Tai Lake, China[J]. *Environmental Science and Pollution Research*, 2018, **25**(32): 32631-32639.
- [76] 刘操, 马宁, 龚明波. 模拟降雨条件下北运河流域农田养分流失特征[J]. 农业资源与环境学报, 2016, **33**(3): 238-243.
Liu C, Ma N, Gong M B. Characteristic of soil nutrients loss in Beiyunhe Reservoir under the simulated rainfall [J]. *Journal of Agricultural Resources and Environment*, 2016, **33**(3): 238-243.
- [77] Meghdadi A, Javar N. Quantification of spatial and seasonal variations in the proportional contribution of nitrate sources using a multi-isotope approach and Bayesian isotope mixing model [J]. *Environmental Pollution*, 2018, **235**: 207-222.



CONTENTS

Impacts of Anthropogenic Emission Reduction on Urban Atmospheric Oxidizing Capacity During the COVID-19 Lockdown	ZHU Jian-lan, QIN Mo-mei, ZHU Yan-hong, <i>et al.</i> (617)
Transport and Potential Sources Regions of Double High Pollution in Nanjing by Different Synoptic Situations	QIN Yang, HU Jian-lin, KONG Hai-jiang (626)
Differences of Three Methods in Determining Ozone Sensitivity in Nanjing	CHEN Gan-yu, LI Xun, LI Lin, <i>et al.</i> (635)
Characteristics of Ozone Concentration in Shanghai and Its Associated Atmospheric Circulation Background During Summer Half-years from 2006 to 2021	ZHENG Qing-feng, LIANG Ping, DUAN Yu-sen, <i>et al.</i> (645)
Distribution Characteristics of Near Surface Ozone Volume Fraction in Shanxi Province Based on Atmospheric Composition Observation Network	LI Ying, WANG Shu-min, PEI Kun-ning, <i>et al.</i> (655)
Photochemical Mechanism and Control Strategy Optimization for Summertime Ozone Pollution in Yining City	WANG Wen-ting, GU Chao, LI Li-ming, <i>et al.</i> (668)
Characteristics, Sources, and Ozone-sensitive Species of VOCs in Four Seasons in Yuncheng	YIN Shi-jie, LIU Xin-gang, LIU Ya-fei, <i>et al.</i> (678)
Pollution Characteristics, Source Analysis, and Activity Analysis of Atmospheric VOCs During Winter and Summer Pollution in Zhengzhou	LAI Meng-jie, ZHANG Dong, YU Shi-jie, <i>et al.</i> (689)
Pollution Characteristics, Sources, and Secondary Generation of Organic Acids in PM _{2.5} in Zhengzhou	LI Zi-han, DONG Zhe, SHANG Lu-qi, <i>et al.</i> (700)
Nonlinear Variations in PM _{2.5} Concentration in the Three Major Urban Agglomerations in China	WU Shu-qi, GU Yang-yang, ZHANG Tian-yue, <i>et al.</i> (709)
Medium and Long-term Carbon Emission Projections and Emission Reduction Potential Analysis of the Lingang Special Area Based on the LEAP Model	WU Qiong, MA Hao, REN Hong-bo, <i>et al.</i> (721)
Dynamic Analysis on Carbon Metabolism of the Northern Region of China Under the Background of Carbon Emission Trading Policy	ZHENG Hong-mei, SHEN Fang, XU Guang-yao, <i>et al.</i> (732)
Carbon Reduction Analysis of Life Cycle Prediction Assessment of Hydrogen Fuel Cell Vehicles ; Considering Regional Features and Vehicle Type Differences	MA Jing, CAI Xu, ZHANG Chun-mei, <i>et al.</i> (744)
Nitrate Pollution Characteristics and Its Quantitative Source Identification of Major River Systems in China	WEI Ying-huai, HU Min-peng, CHEN Ding-jiang (755)
Effects of Land Use Structure and Spatial Pattern at Different Temporal and Spatial Scales on Water Quality in Suzhou Creek	TAN Juan, XIONG Li-jun, WANG Qing, <i>et al.</i> (768)
Spatial-temporal Variation in Water Quality of Rain-source Rivers in Shenzhen from 2015 to 2021 and Its Response to Rainfall	WEI Bi-ying, CHENG Jian-mei, SU Xiao-yu, <i>et al.</i> (780)
Chemical Characteristics of Shallow Groundwater in the Yellow River Diversion Area of Henan Province and Identification of Main Control Pollution Sources	WANG Shuai, REN Yu, GUO Hong, <i>et al.</i> (792)
Chemical Characteristics and Genetic Analysis of Karst Groundwater in the Beijing Xishan Area	GUO Gao-xuan, DAI Yin-dong, XU Liang, <i>et al.</i> (802)
Hydrochemical Characteristics and Its Origin of Surface Water and Groundwater in Dianbu River Basin	ZHENG Tao, QIN Xian-yan, WU Jian-xiong (813)
Hydrochemical Characteristics and Genesis Mechanism of Groundwater in the Dry Period in the Zhangjiakou Area	JIN Ai-fang, YIN Xiu-lan, LI Chang-qing, <i>et al.</i> (826)
Distribution of Typical Resistant Bacteria and Resistance Genes in Source Water of the Middle and Lower Reaches of the Yellow River	MIN Wei, GAO Ming-chang, SUN Shao-fang, <i>et al.</i> (837)
Contamination Characteristics, Detection Methods, and Control Methods of Antibiotic Resistance in Pharmaceutical Wastewater	PENG An-ping, GAO Hu, ZHANG Xin-bo (844)
Effect of Water Components on Aggregation and Sedimentation of Polystyrene Nano-plastics	TANG Duan-yang, ZHENG Wen-li, CHEN Guan-tong-yi, <i>et al.</i> (854)
Lead Removal from Water by Calcium-containing Biochar with Saturated Phosphate	LIU Tian, LÜ Si-lu, DU Xing-guo, <i>et al.</i> (862)
Preparation of Chitosan-modified Biochar and Its Adsorption Mechanism for Cd ²⁺ in Aqueous Solution	JIANG Ling, AN Jing-yue, YUE Xiao-qiong, <i>et al.</i> (873)
Efficacy and Mechanism of Tetracycline Adsorption by Boron-doped Mesoporous Carbon	ZOU Zhen, XU Lu, QIAO Wei, <i>et al.</i> (885)
Adsorption Properties of Magnetic Phosphorous Camellia Oleifera Shells Biochar to Sulfamethoxazole in Water	HAN Shuai-peng, TANG Li-wen, LIU Qin, <i>et al.</i> (898)
High Resolution Emission Inventory of Greenhouse Gas and Its Characteristics in Guangdong, China	LU Qing, TANG Ming-shuang, LIAO Tong, <i>et al.</i> (909)
Ecosystem CO ₂ Exchange and Its Environmental Regulation of a Restored Wetland in the Liaohe River Estuary	LIU Si-qi, CHEN Hong, XING Qing-hui, <i>et al.</i> (920)
Effects of Biochar Application Two Years Later on N ₂ O and CH ₄ Emissions from Rice-Vegetable Rotation in a Tropical Region of China	HU Yu-jie, TANG Rui-jie, HU Tian-yi, <i>et al.</i> (929)
Research and Application Progress of Biochar in Amelioration of Saline-Alkali Soil	WEI Ying, JIAO Le, ZHANG Peng, <i>et al.</i> (940)
Effect of No-tillage on Soil Aggregates in Farmland: A Meta Analysis	XU Yi-ping, RAO Yue-yue, MENG Yan, <i>et al.</i> (952)
Spatio-temporal Evolution and Multi-scenario Simulation of Carbon Storage in Karst Regions of Central Guizhou Province; Taking Puding County as An Example	LI Yue, LUO Hong-fen (961)
Effects of Different Modifiers on Aggregates and Organic Carbon in Acidic Purple Soil	LI Yue, XU Man, XIE Yong-hong, <i>et al.</i> (974)
Effect of Ca Modified Biochar on the Chemical Speciation of Soil Phosphorus and Its Stabilization Mechanism	ZHANG Chao, ZHAI Fu-jie, SHAN Bao-qing (983)
Plant Diversity Changes and Its Driving Factors of Abandoned Land at Different Restoration Stages in the Middle of the Qinling Mountains	YAN Cheng-long, XUE Yue, WANG Yi-fei, <i>et al.</i> (992)
Contamination Characteristics and Ecological Risk of Antibiotics in Contaminated Sites of Typical Pharmaceutical Factories in China	YANG Jiong-bin, HUANG Zheng, ZHAO Jian-liang, <i>et al.</i> (1004)
Pollution Characteristics and Risk Assessment of Polycyclic Aromatic Hydrocarbons in Soils of Guangzhou	ZOU Zi-hang, CHEN Lian, ZHANG Pei-zhen, <i>et al.</i> (1015)
Quantifying the Contribution of Soil Heavy Metals to Ecological and Health Risk Sources	PAN Yong-xing, CHEN Meng, WANG Xiao-tong (1026)
Potential Ecological Risk Characteristics and Source Apportionment of Heavy Metals in Farmland Soils around Typical Factories in Hunan Province Based on Monte-Carlo Simulation	LUO Hao-jie, PAN Jun, CHEN Xiao-xia, <i>et al.</i> (1038)
Health Risk Assessment for an Arsenic-contaminated Site Based on Monte Carlo Simulation and Parameters Optimization	YUAN Bei, LIU Hu-peng, DU Ping, <i>et al.</i> (1049)
Pollution Source Apportionment of Heavy Metals in Cultivated Soil Around a Red Mud Yard Based on APCS-MLR and PMF Models	SHEN Zhi-jie, LI Jie-qin, LI Cai-xia, <i>et al.</i> (1058)
Characteristics and Mechanism of Cd Release and Transport in Soil Contaminated with PE-Cd	WANG Di, XU Shao-hui, SHAO Ming-yan, <i>et al.</i> (1069)
Characterization of Reductive Dechlorination of Chlorinated Ethylenes by Anaerobic Consortium	LI Wei, LIU Gui-ping, LIU Jun, <i>et al.</i> (1080)
Analysis of Heavy Metal Pollution Evaluation and Correlation of Farmland Soil and Vegetables in Zhaotong City	ZHANG Hao, DONG Chun-yu, YANG Hai-chan, <i>et al.</i> (1090)
Safe Utilization Effect of Passivator on Mild to Moderate Cadmium Contaminated Farmland	WANG Xiao-jing, ZHANG Dong-ming, CAO Yang, <i>et al.</i> (1098)
Simultaneous Immobilization of Cadmium and Arsenic in Paddy Soils with Novel Fe-Mn Combined Graphene Oxide	YUAN Jing, WU Ji-zi, LIAN Bin, <i>et al.</i> (1107)
Effects of the Application of Irrigation Water Containing Zn at the Key Growth Period on the Uptake and Transport of Cd in Rice	ZHOU Xia, HU Yu-dan, ZHOU Hang, <i>et al.</i> (1118)
Effects of Exogenous Zinc on Growth and Root Architecture Classification of Maize Seedlings Under Cadmium Stress	ZHANG Hui-hong, WEI Chang, LIU Hai-tao, <i>et al.</i> (1128)
Mitigative Effect of Rare Earth Element Cerium on the Growth of Zinc-stressed Wheat (<i>Triticum aestivum</i> L.) Seedlings	ZHANG Jing-jing, XU Zheng-yang, JIAO Qiu-juan, <i>et al.</i> (1141)
Two-stage Inhibition Effects of <i>Burkholderia</i> sp. Y4 Application on Cadmium Uptake and Transport in Wheat	GUO Jia-jia, WANG Chang-rong, LIU Zhong-qi, <i>et al.</i> (1150)
Effects of Combined Stress of High Density Polyethylene Microplastics and Chlorimuron-ethyl on Soybean Growth and Rhizosphere Bacterial Community	HU Xiao-yue, HUA Zi-wei, YAO Lun-guang, <i>et al.</i> (1161)
Human Accumulation and Toxic Effects of Microplastics: A Critical Review	BAO Ya-bo, WANG Cheng-chen, PENG Wu-guang, <i>et al.</i> (1173)
Overview of the Application of Machine Learning for Identification and Environmental Risk Assessment of Microplastics	BAI Run-hao, FAN Rui-qi, LIU Qi, <i>et al.</i> (1185)
Research Process on the Combined Pollution of Microplastics and Typical Pollutants in Agricultural Soils	HOU Yu-qing, LI Bing, WANG Jin-hua, <i>et al.</i> (1196)
Research Progress in Electrochemical Detection and Removal of Micro/Nano Plastics in Water	ZHENG Wei-kang, LIU Zhen-zhong, XIANG Xiao-fang (1210)
Factors Influencing Willingness of Farmers to Pay for Agricultural Non-point Source Pollution Control Based on Distributed Cognitive Theory	GUO Chen-hao, LI Lin-fei, XIA Xian-li (1222)