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目 次

加速能源转型与产业结构调整的环境健康协同效益评估:以京津冀鲁地区为例 …… 杨玺,孙奕生,常世彦,李胜悦,郑昊天,王书肖,张希良(3627)
基于不同电力需求的中国减污降碳协同增效路径 …… 向梦宇,王深,吕连宏,张楠,白梓函(3637)
城市碳达峰碳中和行动评估方法与应用 …… 张保留,白梓函,张楠,吕连宏,阳平坚(3649)
基于可解释性机器学习的城市 O_3 驱动因素挖掘 …… 董佳奇,胡冬梅,闫雨龙,彭林,张鹏辉,牛月圆,段小琳(3660)
基于响应曲面法的臭氧生成敏感性分析 …… 朱禹寰,陈冰,张雅勉,刘晓,李光耀,舍静,陈强(3669)
基于WRF-CMAQ/ISAM模型的榆林市夏季 O_3 来源解析 …… 王伊凡,仝纪龙,陈羽翔,林鑫,刘永乐,敖丛杰,刘浩天(3676)
基于轻量级梯度提升机的南京大气臭氧浓度预测 …… 朱珈莹,安俊琳,冯悦政,贺婕,张玉欣,王俊秀(3685)
2022年广东省冬季一次臭氧污染过程的气象成因及潜在源区分析 …… 李婷苑,陈靖扬,龚宇,沈劲(3695)
夏季对流层臭氧辐射强迫对华北地区天气和空气质量的影响 …… 杜楠,陈磊,廖宏,朱佳,李柯(3705)
气象、本地光化学生成和外围传输对长沙市2018~2020年臭氧污染趋势变化影响的识别 …… 杨俊,杨雷峰,丁华,谢丹平,刘妍妍,余涛,吕明,袁自冰(3715)
2000~2021年成渝城市群 $PM_{2.5}$ 时空变化及驱动机制多维探测 …… 徐勇,郭振东,郑志威,戴强玉,赵纯,黄雯婷(3724)
基于GTWR-XGBoost模型的四川省 $PM_{2.5}$ 小时浓度估算 …… 吴迪,杜宁,王莉,吴宇宏,张少磊,周彬,张显云(3738)
2013~2021年邢台 $PM_{2.5}$ 重污染过程输送和增长特性 …… 江琪,盛黎,靳雨晨,王继康,尤媛,王飞(3749)
基于高分辨率在线观测数据分析上海市城区秋冬季大气有机气溶胶化学特征及污染来源 …… 朱书慧(3760)
机动车尾气碳质气溶胶排放因子及其稳定碳同位素特征 …… 于鸣媛,王谦,付明亮,戈畅,谢锋,曹芳,章炎麟(3771)
沈阳工业区夏季VOCs组成特征及其对二次污染形成的贡献 …… 关璐,苏枞枞,库盈盈,胡建林,于兴娜(3779)
疫情管控期间深圳市城区VOCs的变化特征及减排效果评估 …… 云龙,林楚雄,李成柳,邱志诚,古添发,李光程,张明棣,郭键锋(3788)
西安市冬季道路扬尘中有机质组成特征及其氧化潜势 …… 王擎雯,陈庆彩,王超,王瑞鹤,刘国瑞,李豪,李艳广(3797)
淮北孙疃矿区地表尘中多环芳烃类化合物的污染特征及致癌风险评价 …… 徐振鹏,钱雅慧,洪秀萍,罗钟庚,高秀龙,梁汉东(3809)
雄安建设初期白洋淀水质时空差异及其对降雨和土地利用的响应 …… 王子铭,杨丽虎,宋献方(3820)
流域尺度污染溯源模拟-优化防控方法:以铜陵市顺安河流域为例 …… 刘国王辰,陈磊,李佳奇,张钰晨,赵奕欣,刘妍琪,沈珍瑶(3835)
南昌市浅层地下水水质评价及监测指标优化 …… 郑紫吟,储小东,徐金英,马志飞(3846)
闪电河流域"三水"氢氧同位素特征及水体转换分析 …… 杨丽娜,贾德彬,高瑞忠,苏文旭,卢方园,郝玉胜(3855)
金沙江丰富类和稀有类浮游真核微生物的分布特征与影响因素 …… 燕炳成,崔戈,孙胜浩,王沛芳,王超,吴程,陈娟(3864)
嘉陵江不同干扰断面河道沉积物细菌群落多样性 …… 竹兰萍,张拓,李佳宁,王佳颖,郑伟亮,徐飞,张富斌(3872)
水库水深变化对不同浮游微生物群落及网络互作关键种的影响 …… 王洵,廖琴,王沛芳,袁秋生,胡斌,邢小蕾,徐浩森(3881)
典型工业区地下水细菌群落多样性特征与环境因子响应初探 …… 吴建强,张书源,王敏,陈敏,叶文娟,熊丽君,黄沈发(3892)
太湖流域肥料施用策略调整对典型作物系统氮磷流失的影响 …… 俞映倞,王逸之,杨梖,杨林章,段婧婧,韩雪梅,薛利红(3902)
基于机器学习的长江流域农田氮径流流失负荷估算 …… 张育福,潘哲祺,陈丁江(3913)
千岛湖地区上梧溪流域地表径流非点源氮污染分类识别 …… 俞珂,严琰,唐张轩,张方方,何圣嘉,姜培坤(3923)
沱江和涪江水系干支流氮磷营养盐的空间分布特征 …… 李子阳,周明华,徐鹏,陈露,刘祥龙,林洪羽,江南,任兵,张博文(3933)
外秦淮河底泥污染及疏浚效果 …… 张沐,任增谊,张曼,赵琼,尹洪斌(3945)
新污染物多环芳烃衍生物的来源、分布与光化学行为 …… 葛林科,王子宇,曹胜凯,车晓佳,朱超,张蓬,马宏瑞(3957)
镉砷在针铁矿界面共吸附的相互作用机制 …… 苏子贤,刘赛红,管玉峰,陶亮(3970)
环糊精键合Fe-TAML催化剂的制备及其活化 H_2O_2 氧化水中有机微污染物 …… 刘清泉,蔡本哲,蔡喜运(3978)
Co(Ⅱ)活化过一硫酸盐降解氨基三亚甲基膦酸的性能及反应机制 …… 朱敬林,汪舒(3990)
微塑料诱导下污泥造粒潜能变化及微生物富集特征 …… 谢晴帆,俞楠,张妮,谢周云,单珂欣,吴亦馨,唐力,夏静芬,杨国靖(3997)
中国农田土壤Cd累积分布特征及概率风险评价 …… 王静,魏恒,潘波(4006)
渝东南典型地质高背景区土壤重金属来源解析及污染评价 …… 蒋玉莲,余京,王锐,王佳彬,李瑜,余飞,张云逸(4017)
基于PMF模型的农田土壤重金属源暴露风险综合评价:以浙江省某电子垃圾拆解区为例 …… 方嘉,何影,黄乃涛,支裕优,傅伟军(4027)
乌鲁木齐市郊农田土壤及农作物中多环芳烃的污染特征及风险评价 …… 范悦,曹双瑜,艾力江·努尔拉,于苏云江·吗米提敏,阿不都艾尼·阿不里,谢宣宣,古丽斯坦·阿不都拉,刘河疆(4039)
荒漠绿洲土壤抗生素抗性基因分布特征及驱动机制 …… 黄福义,周曙仡聃,苏建强,朱永官(4052)
近15年我国土壤抗生素污染特征与生物修复研究进展 …… 赵晓东,乔青青,秦宵睿,李晓晶,李永涛(4059)
改性生物炭修复砷镉复合污染土壤研究进展 …… 吕鹏,李莲芳,黄晓雅(4077)
沼液还田对土壤-作物系统重金属累积的影响:Meta分析 …… 赵奇志,杨志敏,孔凡靖,熊海灵,朱康文,陈玉成(4091)
两种铁改性生物炭对微碱性砷镉污染土壤的修复效果 …… 梁欣冉,何丹,郑嬰华,付庆灵,胡红青,朱俊(4100)
秸秆离田对土壤Cd生物有效性及水稻Cd积累的影响 …… 王子钰,周航,周坤华,谭文韬,蒋毅,唐棋,伍港繁,辜娇峰,曾鹏,廖柏寒(4109)
秸秆与脱硫石膏配施改良黄河三角洲盐碱地的理化性质 …… 赵惠丽,于金艺,刘涛,王丽,赵英(4119)
基于环境DNA的复合污染土壤生物评价和胁迫诊断 …… 黄湘云,钟文军,刘训杰,毕婉娟,钱林皓,张效伟(4130)
地质高背景区土壤-玉米重金属综合质量评价 …… 张传华,王钟书,刘力,刘燕(4142)
浙江省蔬菜生产系统重金属污染生态健康风险 …… 张述敏,刘翠玲,杨桂玲,邓美华(4151)
生物炭配施磷肥对土壤养分、酶活性及紫花苜蓿养分吸收的影响 …… 刘鑫裕,王冬梅,张泽洲,张鹏,樊桐桐(4162)
低密度聚乙烯微塑料对空心菜生长和生理特征的影响 …… 周颖,蒋文婷,刘训悦,朱高荻,唐荣贵,章海波,蔡延江(4170)
沼液施用对麦稻茬口期土壤微生物群落结构特征及功能的影响 …… 乔宇颖,奚辉,李娜,陈国靖,沈阿林,喻曼(4179)
大同铅锌尾矿不同污染程度土壤细菌群落分析及生态功能特征 …… 刘泽勋,庄家尧,刘超,郑康,陈玲(4191)
马尾松采伐迹地火烧黑炭对土壤有机碳组分和碳转化酶活性的影响 …… 姚智,焦鹏宇,吴晓生,严强,刘先,胡亚林,王玉哲(4201)
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机动车尾气碳质气溶胶排放因子及其稳定碳同位素特征

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摘要: 机动车尾气是大气碳质气溶胶的重要人为来源,其排放因子与稳定碳同位素组成是重要的基础数据. 选取多辆不同类型在用机动车,进行多种工况、冷/热条件下启动的台架试验,收集各测试阶段尾气分析其碳质组分含量与稳定碳同位素比值,并探讨其影响因素. 结果表明,总碳排放因子大小为:重型柴油车 > 轻型柴油车 > 轻型汽油车,轻型天然气车虽然在低速与中速阶段排放因子极低,但高速行驶阶段可达到重型柴油车的排放水平. 各型车冷启动的排放因子均高于热启动,NEDC 工况的排放因子整体低于 WLTC 工况,应与其测试车速有关. 汽油车和天然气车各测试阶段排放有机碳(OC)均远高于元素碳(EC),柴油车 OC 与 EC 排放因子相近,各类车辆 OC/EC 都随测试车速的提高而上升. 稳定碳同位素 EC 重于 OC,同位素比值大小关系均呈现:汽油车 < 天然气车 < 轻型柴油车 < 重型柴油车,现有源解析的稳定碳同位素源谱较难反映汽油车与天然气车特征. 在排放治理与源解析工作中,应注意替代燃料的使用与机动车老化过程所造成的排放因子与同位素特征值的变化影响.

关键词: 机动车尾气; 排放因子; 有机碳(OC); 元素碳(EC); 稳定碳同位素; 底盘测功机
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Emission Factors of Carbonaceous Aerosol and Stable Carbon Isotope for In-use Vehicles

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Abstract: Vehicle exhaust is an important anthropogenic source of atmospheric carbonaceous aerosols; of which, the emission factors and stable carbon isotope composition are important basic data. In-use motor vehicles of different types were selected to conduct dynamometer tests using different test cycles and under cold/hot start conditions. The exhaust of each test stage was collected to analyze the carbonaceous components and stable carbon isotopes and to discuss the influencing factors. The total carbon emission factors follow the order: heavy-duty diesel vehicles > light-duty diesel vehicles > light-duty gasoline vehicles. Although the emission factors of light-duty natural gas vehicles were very low at the low- and medium-speed stages, they were similar to those of heavy-duty diesel vehicles at the high-speed stage. The emission factors of cold start were higher than those of hot start, and the emission factors of the NEDC test cycle were lower than those of WLTC (which should be related to the driving speed). The emission factors of organic carbon (OC) of gasoline and natural gas vehicles were much higher than those of elemental carbon (EC) in every test stage. The emission factors of OC and EC of diesel vehicles were similar. The OC/EC of all types of vehicles increased with the increase in driving speed. Stable carbon isotopes in EC were higher than those in OC. The stable carbon isotope in different vehicles follow the order: light-duty gasoline vehicles < light-duty natural gas vehicles < light-duty diesel vehicles < heavy-duty diesel vehicles. The results revealed that the source signatures of stable carbon isotope in vehicles used in current source apportionment could not well represent gasoline vehicles and natural gas vehicles. In future emission control and source apportionment, attention should be paid to the changes in emission factors and isotope signatures caused by the use of natural gas and the aging of motor vehicles.

Key words: motor vehicle exhaust; emission factor; organic carbon(OC); elemental carbon(EC); stable carbon isotope; chassis dynamometer

碳质气溶胶(total carbon, TC)包含有机碳(organic carbon, OC)和元素碳(elemental carbon, EC),是大气细颗粒物中的重要组分(约占 20% ~ 50%),对环境、气候以及人体健康均有重要影响[1~3]. 机动车尾气是碳质气溶胶的重要人为来源之一[4~6],准确识别机动车尾气中碳质气溶胶的构成,是优化排放清单、进行针对性管控以及开展大气污染物来源解析等的重要基础工作[7~9]. 碳质气溶胶是机动车尾气颗粒物中的首要组分(约占 50% ~ 90%)[10,11],然而其排放因子受到燃料类型、车辆类型、启动条件和行驶工况等因素影响[11~13]. 因而有必要针对各类影响因素测定排放因子,充实排放清单的基础数据库. 对于机动车尾气排放因子的测定,常用手段包括发动机测功[11,14]、道路试验[15,16]和隧道试验[17,18]等. 发动机测功方法尽管并非实际

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道路测试,但其测试条件更为可控,可得到特定车型、特定行驶工况以及特定环境下的排放因子,因而对于排放因子的精细化研究十分有利[19,20].

碳同位素技术是对碳质气溶胶进行来源解析的一项准确可靠的手段[21~26],该方法根据不同燃料的稳定同位素比值($\delta^{13}C$)的差异,以及不同的燃烧条件下产生的同位素分馏,解析出燃煤、机动车、C3 与 C4 植物对大气样品的贡献. 典型源排放碳质气溶胶的 $\delta^{13}C$ 值,是利用同位素技术展开解析的重要基础数据,因燃料特征、燃烧条件等存在地域差异,进行本土的源排放试验与测试工作对建立同位素数据库尤为重要[22,27,28]. 2007 年刘刚等[29]在杭州随机选取机动车,测试 OC 与 EC 的 $\delta^{13}C$ 值,陈颖军等[28]分析了不同车型排放 TC 与 EC 的 $\delta^{13}C$ 值,石磊等[30]与 Guo 等[31]分别测定了南京机动车 TC 与 EC 的 $\delta^{13}C$ 值,为排放源 $\delta^{13}C$ 数据库的建立提供了重要数据. 然而上述研究均通过采集机动车排气管内累积的烟尘进行同位素分析,尚缺乏可直接指示排放到大气中颗粒物 $\delta^{13}C$ 的证据,另外随着机动车排放标准与油品质量的改变,在用各类机动车的尾气颗粒物 $\delta^{13}C$ 数据库亟待更新.

针对以上现状,本研究选取不同燃料类型、重量类型与排放标准的在用车辆共 12 辆,采用发动机测功方法进行冷/热启动与多种工况测试,收集各个测试阶段尾气的细颗粒物($PM_{2.5}$)进行碳质气溶胶含量测定与稳定碳同位素分析,以期服务于排放清单的更新、机动车尾气排放的控制与气溶胶源解析研究.

1 材料与方法

1.1 试验车辆

本研究的台架试验于 2019 年 3 ~5 月在厦门进行,共选取 12 辆在用车辆,其中轻型汽油车、轻型柴油车和改装的天然气出租车各 2 辆,重型柴油车 6 辆,详细信息如表 1 所示. 两辆轻型汽油车符合国家第五阶段机动车污染物排放标准(简称国五标准),其他车辆均符合国四标准.

表 1 试验车辆基本信息

Table 1 Basic information of test vehicles

车辆编号	车辆类型	排放标准	发动机排量/L	整备质量/t	里程数/km
LG1	轻型汽油车	国五	1.6	1.2	30 207
LG2	轻型汽油车	国五	1.5	1.3	38 220
LD1	轻型柴油车	国四	2.8	1.7	65 731
LD2	轻型柴油车	国四	(信息缺失)	1.8	146 985
CT1	天然气出租车	国四	1.8	1.5	781 628
CT2	天然气出租车	国四	1.8	1.5	859 649
HD1	重型柴油车	国四	6.5	8.2	85 539
HD2	重型柴油车	国四	6.5	8.2	69 241
HD3	重型柴油车	国四	3.8	6.0	67 588
HD4	重型柴油车	国四	6.9	9.4	107 960
HD5	重型柴油车	国四	6.9	9.4	50 621
HD6	重型柴油车	国四	4.8	5.6	90 977

1.2 测试方法

所有车辆均采用底盘测功机和全流定容稀释(constant volume sampling, CVS)系统进行测试,测试系统如图 1 所示. 轻型车采用 AVL 48″ 4WD 底盘测功机和 AVL LE 汽油/柴油排放测试系统(AMA i60、CVS i60、PASS i60 和 BMD&DVE 150),重型车采用 Burke 75″ HD 底盘测功机和 AVL HD 分析系统(AMAi60HD、CVSi60 HD 和 PSSi60HD). 轻型车辆测试工况为全球轻型车统一测试循环(worldwide harmonized light-duty driving test cycle, WLTC)[32],该循环全程共1 800 s,包括低速阶段(589 s)、中速阶段(433 s)、高速阶段(455 s)和超高速阶段(323 s),依次标记为 L、M 和 H(高速和超高速阶段合并采样). 重型车测试工况为 C-WTVC (China worldwide transient vehicle cycle)[33],是以世界统一的重型商用车辆瞬态车辆循环(world transient vehicle cycle, WTVC)为基础,调整加速度和减速度形成的驾驶循环. 该循环全程共1 800 s,包含城区阶段(900 s)、乡村阶段(468 s)和高速阶段(432 s),依次标记为 p1、p2 和 p3,城区阶段存在频繁的加速与减速,高速阶段车速较为稳定. 采用 WLTC 工况和 C-WTVC 工况测试的车辆均分别在冷/热启动条件下进行重复测试与采样,以对比启动条件对碳质气溶胶排放的影响. 另为研究不同测试工况的差异,对于轻型车辆采用新欧洲驾驶循环(new European driving cycle, NEDC)在热启动条件下进行重复测试,该循环由城区阶段(ECE, p1)和市郊阶段(EUDC, p2)组成,全程共1 180 s.

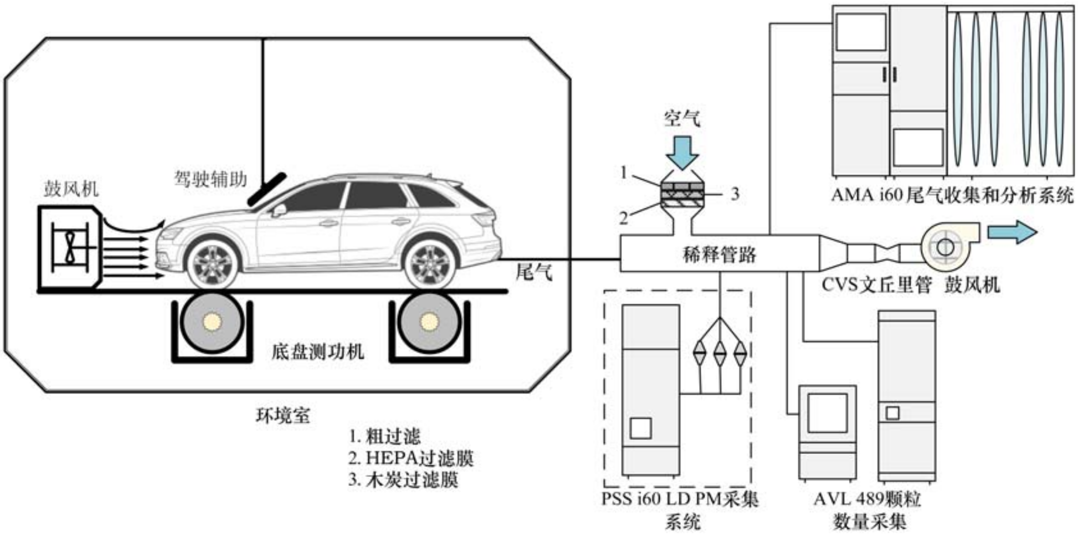

图 1　测试系统示意

Fig. 1　Schematic diagram of the test system

测试前 1 d 将测试车辆驶入恒温恒湿的环境室(室温 24℃,相对湿度 66%)中停放 12 h 以上,测试前使用标准气通入稀释管路对测试仪器校准. 测试时,将机动车排气管接通稀释管路,先后进行冷/热启动测试,冷/热启动测试之间进行管路清洗与后一工况的预热. 采样前将 42 mm 标准圆形石英滤膜放入马弗炉中 400 ℃烧 4 h 以控制空白,采用颗粒物采集系统分三路对各工况的尾气进行分阶段采集,采样流速 40 L·min^{-1},采样后将滤膜放入聚四氟乙烯膜盒中置于冰箱冷藏.

1.3　碳质组分与同位素分析方法

$PM_{2.5}$中的碳质组分采用美国 Sunset Lab 公司 Model-4 型全自动半连续式 OC/EC 分析仪进行测定,升温程序选用稍作改动后的 NIOSH 5040 方法. 测样开始前和结束后用标准蔗糖溶液进行外标校正,采用内外标联合校正确保仪器具有较高的精准度,其检出限为 0.5 μg.

选取一辆轻型汽油车(因含碳量过低及膜量不足等原因,只选取 LG2)、两辆天然气出租车、两辆轻型柴油车和两辆重型柴油车(HD1 和 HD6)进行稳定碳同位素分析. 分析方法采用 OC/EC 分析仪与光腔衰荡光谱(G2201-i, Picarro)联用,该方法曾用于测试北京环境大气样品,准确度优于 0.1‰[34].

2　结果与讨论

2.1　TC 排放因子

对于各个工况下的测试,收集各测试阶段尾气测定其碳质气溶胶含量,结合行驶里程获得对应的排放因子,然后对测试里程进行加权平均,得到该工况下的平均排放因子. TC 测试结果如表 2 所示,所有测试车辆中,轻型汽油车的 TC 排放因子最低,冷/热启动条件下分别为(6.1 ± 5.9) mg·km^{-1}和(2.0 ± 1.1) mg·km^{-1}. 轻型柴油车的 TC 排放因子[(40.6 ± 28.0) mg·km^{-1}和(34.6 ± 9.1) mg·km^{-1}]整体上低于重型柴油车[(52.3 ± 24.6) mg·km^{-1}和(45.7 ± 21.3) mg·km^{-1}],应与发动机排量和整车重量等有关. 与以往研究相比[10,11,35~39],本研究的柴油车排放因子略低,应与测试车辆对应的排放标准有关,而汽油车排放因子略高. 天然气车的尾气在以往研究中得到关注较少,对排放因子的测试更侧重于 CO 和 NO_x 等[40,41],本研究测试发现天然气车 TC 排放因子很高,热启动条件下[(43.1 ± 22.3) mg·km^{-1}]接近重型柴油车,冷启动条件下[(96.5 ± 12.8) mg·km^{-1}]远高于重型柴油车,在机动车排放控制工作中不可忽视.

表 2　各类车辆的 TC 排放因子/mg·km^{-1}

Table 2　Emission factors of TC/mg·km^{-1}

类型	冷启动	热启动	
	WLTC	WLTC	NEDC
LG	6.1 ± 5.9	2.0 ± 1.1	2.1 ± 0.5
LD	40.6 ± 28.0	34.6 ± 9.1	21.1 ± 15.7
CT	96.5 ± 12.8	43.1 ± 22.3	3.4 ± 2.1
HD	C-WTVC		C-WTVC
	52.3 ± 24.6		45.7 ± 21.3

图 2 和图 3 为测试车辆碳质气溶胶排放因子在测试工况各个阶段的分布. 两辆国五轻型汽油车的 TC 排放因子在各个阶段均维持较低水平,变化范围为 1.2 ~ 18.4 mg·km^{-1}. 两辆轻型柴油车 TC 排放因子差异较大,可能与里程有关:里程较短的 LD1 排放因子随测试速度加快而逐渐升高,里程较长的 LD2 在 3 个测试阶段中排放因子均比较高[42~44]. 天然气出租车的 TC 排放因子在 3 个测试阶段中变化最大,低速与中速阶段排放因子较低,高速与超高速阶段排

放因子显著升高,最高值可达 152.1 mg·km⁻¹.2 辆测试车辆均呈现此变化特征,说明并非由车辆故障或测试问题导致,推测可能与车辆较长的行驶里程引起的性能变化有关.6 辆重型柴油车在 3 个测试阶段的排放因子变化呈现相似的趋势,从城区阶段到郊区阶段再到高速阶段,随着行驶速度的逐渐提高,排放因子逐渐降低.在冷(热)启动条件下,3 个测试阶段的平均 TC 排放因子($mg·km^{-1}$)分别为 162.7 ± 71.2(143.1 ± 65.0)、46.3 ± 26.2(38.8 ± 14.8)和 24.9 ± 13.6(22.2 ± 14.9).重型柴油车在城区阶段的 TC 排放因子远高于其他两个阶段,应与该测试阶段频繁的刹车与启动有关.

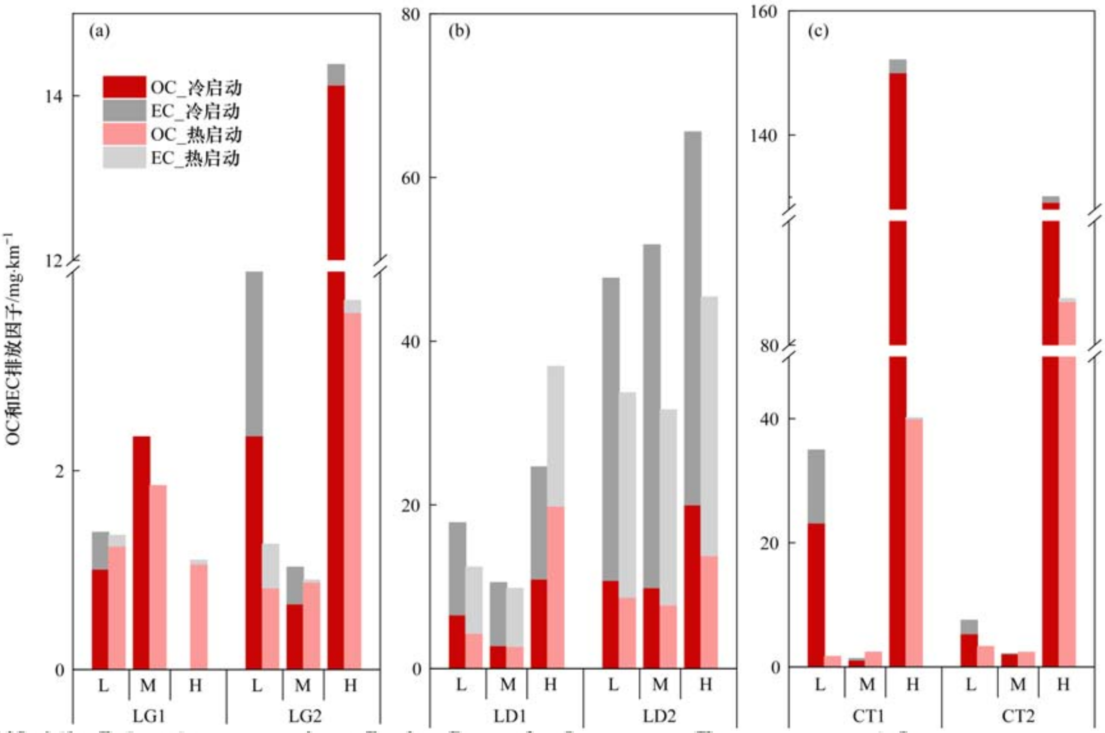

图 2　轻型车 WLTC 工况下各测试阶段碳质气溶胶排放因子

Fig. 2　Emission factors of carbonaceous aerosols for light-duty vehicles in each stage of the WLTC test cycle

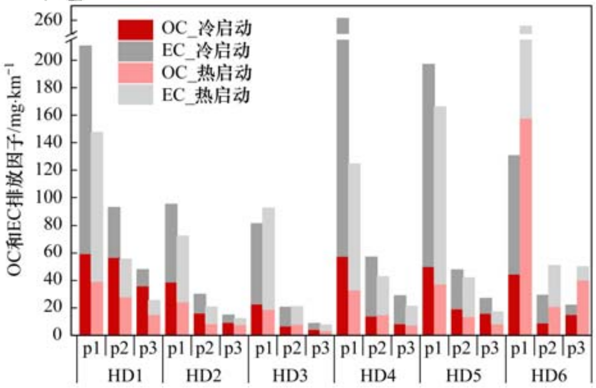

图 3　重型柴油车各测试阶段的碳质气溶胶排放因子

Fig. 3　Emission factors of carbonaceous aerosols for heavy-duty diesel vehicles in each stage of the C-WTVC test cycle

对于车辆的冷/热启动测试,冷启动测试后会进行相应测试工况的预热,预热后立即进行热启动测试.各测试车辆的各个测试阶段,TC 排放因子在冷启动阶段整体上高于热启动阶段,轻型汽油车、轻型柴油车、天然气出租车和重型柴油车冷启动排放因子分别约为热启动的 300%、118%、224% 和 115%.这与以往研究的结论较为一致[45],主要由两方面原因导致:①冷启动时燃料温度较低,燃烧室中燃料汽化不足从而产生不完全燃烧;②低温导致催化转化器的活性较低,无法高效对尾气进行处理.

对 6 辆轻型车重复进行 NEDC 工况下的热启动测试,结果如表 2 和图 4 所示.轻型汽油车在两种工况下的 TC 排放因子接近,轻型柴油车的 TC 排放因子在 NEDC 工况下约为 WLTC 工况下的 60%,天然气出租车的 TC 排放因子在两种工况下差别最大,NEDC 工况下低于 WLTC 工况的 10%.这是因为对于 WLTC 工况,各类测试车辆 TC 排放因子的最高值均出现在测试速度最高的第 3 阶段,而 NEDC 两个测试工况的行驶速度均低于 WLTC 的第 3 阶段,因而无法反映对应的排放状况.说明 NEDC 工况的测试会低估污染物的排放,尤其是对于高速行驶阶段的排放.

2.2　OC 和 EC 排放因子

图 2 和图 3 中的 TC 排放因子也包含了不同碳质组分的信息.轻型汽油车的 TC 主要由 OC 构成(占比超过 95%),而 EC 排放量非常低,WLTC 工况的冷热启动条件下分别为(0.3 ± 0.3)$mg·km^{-1}$ 和

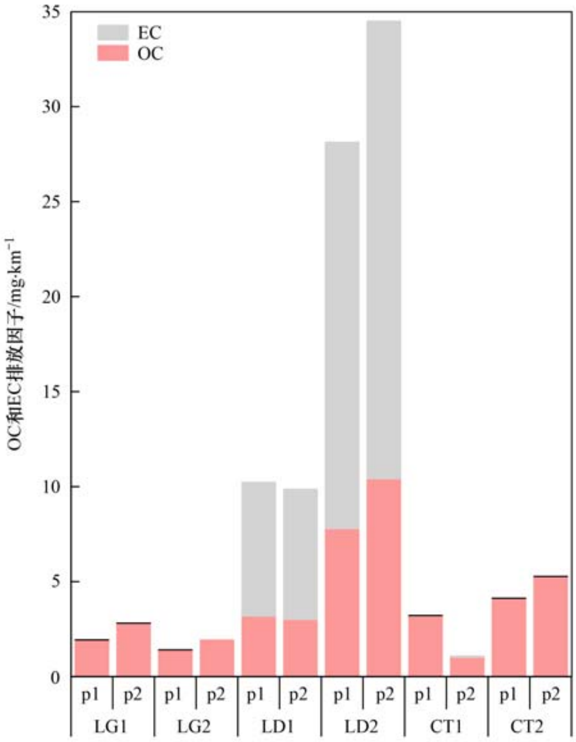

图 4　轻型车 NEDC 工况下各测试阶段碳质气溶胶排放因子

Fig. 4　Emission factors of carbonaceous aerosols for light-duty vehicles in each stage of the NEDC test cycle

(0.1 ±0.1) mg·km^{-1}, NEDC 工况下无 EC 检出. 天然气出租车所排放的 OC 在 TC 中占比超过 98%, EC 排放因子在 WLTC 工况的冷热启动条件下分别为(1.9 ±1.5) mg·km^{-1} 和(0.2 ±0.1) mg·km^{-1}, NEDC 工况下低于 0.1 mg·km^{-1}. 与汽油车和天然气出租车不同,柴油车的 OC 排放因子整体上低于 EC 排放因子. 轻型柴油车在两种工况下的 EC 排放因子均约为 OC 的两倍,重型柴油车在冷启动条件下 EC 排放因子高出 OC 约 50% [(31.0 ±15.8) mg·km^{-1}和(21.3 ±13.0) mg·km^{-1}],热启动条件下 OC 排放因子变化不大[(20.2 ±16.6) mg·km^{-1}],而 EC 排放因子降低[(25.4 ±8.0) mg·km^{-1}],二者差距从而缩小.

整体而言,测试车辆的 OC 排放因子从高到低依次为:天然气出租车 > 重型柴油车 > 轻型柴油车 > 轻型汽油车,EC 排放因子从高到低依次为:重型柴油车 > 轻型柴油车≫天然气出租车 > 轻型汽油车,冷启动与热启动条件下均呈现此分布规律. OC/EC 比值如表 3 所示,柴油车 OC/EC 均低于 1(EC 占主导);轻型汽油车 OC/EC 较高,但较低的 EC 检出量可能导致该数据误差较大;天然气出租车的 OC/EC 最高,冷启动条件下与 Wang 等[46]研究的结果相近,热启动条件下的值高于现有大部分研究,应由 EC 排放量较少、且高速与超高速阶段排放大量 OC 所导致.

表 3　各类车辆尾气的 OC/EC 值

Table 3　OC-to-EC ratios of each type of vehicle

类型	冷启动	热启动
LG	15.5 ±4.2	24.1 ±7.2
LD	0.5 ±0.2	0.7 ±0.4
CT	65.9 ±44.7	178.8 ±4.8
HD	0.7 ±0.3	0.8 ±0.5

对于不同的测试阶段, WLTC 工况下的测试车辆的 OC 和 EC 排放因子从低速到高速阶段均呈现上升趋势(EC 含量过低的除外), 重型柴油车的 OC 和 EC 排放因子在 3 个测试阶段中均呈现下降趋势, 这与 TC 排放因子的变化趋势一致. 对于重型柴油车, 从城区阶段到高速阶段, OC 与 EC 比值(OC/EC)均呈现上升趋势, 冷启动条件下从(0.11 ±0.15) ~ (0.69 ±0.92), 热启动条件下从(0.35 ±0.52) ~ (0.91 ±1.34), 说明与高速阶段相比,城区阶段存在较多的燃料不完全燃烧情况. 另外, 采样期间未考虑环境空气中挥发性有机物的影响, 可能为 OC 的测量引入误差.

2.3　碳质气溶胶中的稳定同位素构成

测试车辆的 $\delta^{13}C$ 分析结果如图 5 所示,TC、OC 和 EC 的$\delta^{13}C$值分别为(−27.89 ±4.09)‰(范围: −35.19‰ ~ −19.78‰, 下同)、(−28.13 ±3.83)‰(−34.58‰ ~ −19.49‰)和 −25.25‰ ±3.02‰(−32.04‰ ~ −20.72‰),TC 与 OC 的 $\delta^{13}C$ 值范围相近,而 EC 的 $\delta^{13}C$ 偏重. Widory 等[47]测定液体燃料燃烧排放 TC 的 $\delta^{13}C$ 为 −28‰ ~ −26‰, 陈颖军等[28]分析了汽油车与柴油车的尾气管内壁烟尘样品,TC 与 EC 的 $\delta^{13}C$ 分别为(−25.75 ±0.32)‰和(−25.17 ±0.40)‰,石磊等[30]采用相似的方法测定 TC 的 $\delta^{13}C$ 范围为 −26.32‰ ~ −23.57‰. 本研究的分析结果与之相比,同位素均值较为接近但数值范围更大,这一方面说明通过分析尾气管烟尘的方法可以一定程度上获得可信的稳定碳同位素结果,同时也表明燃料、车型、行驶方式等因素的确会对尾气稳定碳同位素比值造成不可忽视的影响.

燃料类型的差异对尾气碳质气溶胶的 $\delta^{13}C$ 影响显著,以 TC 为例,汽油车 $\delta^{13}C$ 值最低,为(−32.25 ±2.27)‰, 其次为天然气车[(−30.69 ±2.44)‰],柴油车 $\delta^{13}C$ 值较高,轻型柴油车和重型柴油车的 $\delta^{13}C$ 分别为(−25.85 ±2.55)‰和(−23.45 ±1.68)‰. OC 与 EC 的 $\delta^{13}C$ 值呈现相似分布,尽管由于汽油车和天然气车 EC 排放量较低而导致部分样品无法满足同位素分析需求,依然呈

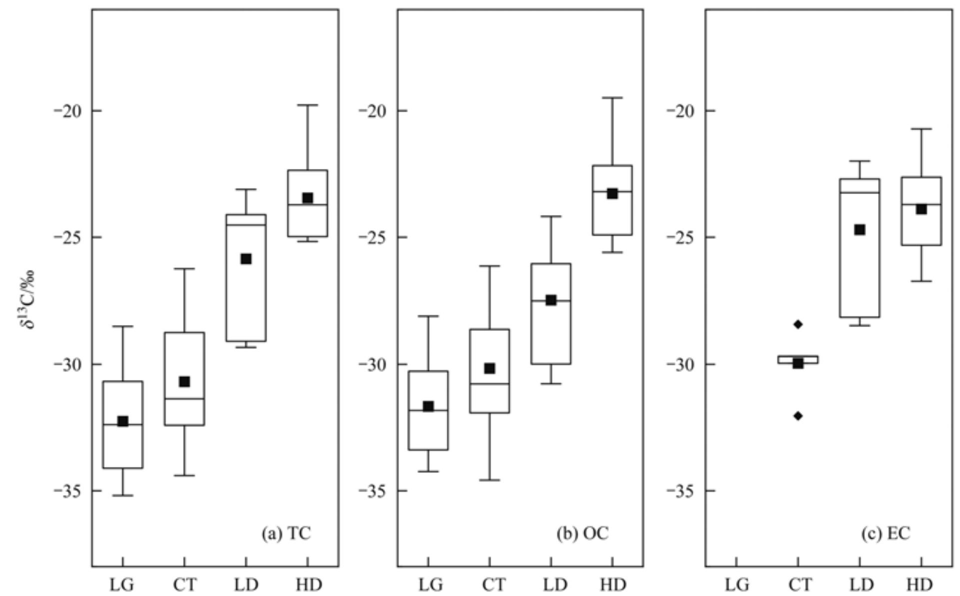

图 5　各类测试车辆排放 TC、OC 和 EC 中的稳定碳同位素比值

Fig. 5　Stable carbon isotope in TC, OC, and EC for each type of vehicle

现柴油车的 $\delta^{13}C$ 显著高于其他车型的分布特征. 目前利用碳同位素进行源解析研究中[21,27]，所采用的机动车源稳定同位素特征值与本研究测试的柴油车相近，高于汽油车和天然气车，其解析结果应存在一定的误差.

各工况各测试阶段 OC 和 EC 的 $\delta^{13}C$ 分析结果如图 6 所示，基本特征为：①同车型、同测试阶段内，EC-$\delta^{13}C$ 略重于 OC-$\delta^{13}C$；②随着测试车速的增加，$\delta^{13}C$ 逐渐变重；③冷/热条件下启动对尾气 $\delta^{13}C$ 几乎无影响. 与其他化石燃料相比，天然气的 $\delta^{13}C$ 最低[47]，一定程度上可以解释较轻的天然气车尾气，然而在不同的测试阶段中（尤其是 WLTC 工况的高速阶段）OC-$\delta^{13}C$ 出现较大变动，应与排放因子的急剧增长（图 2）相关. 两辆轻型柴油车中，LD2 的 $\delta^{13}C$ 整体高于 LD1，推测与行驶里程有关，LD2 里程较长，发动机性能与尾气处理装置有效性均劣于 LD1，因而碳质气溶胶排放量与 $\delta^{13}C$ 均接近重型柴油车. 两辆重型柴油车 $\delta^{13}C$ 特征相近. 尽管轻型汽油车和天然气车的有效样品数量较少且 $\delta^{13}C$ 值跨度较大，但其 OC 和 EC 的 $\delta^{13}C$ 值均低于柴油车，因而进行环境样品（尤其是城市环境样品）源解析时应考虑这一因素.

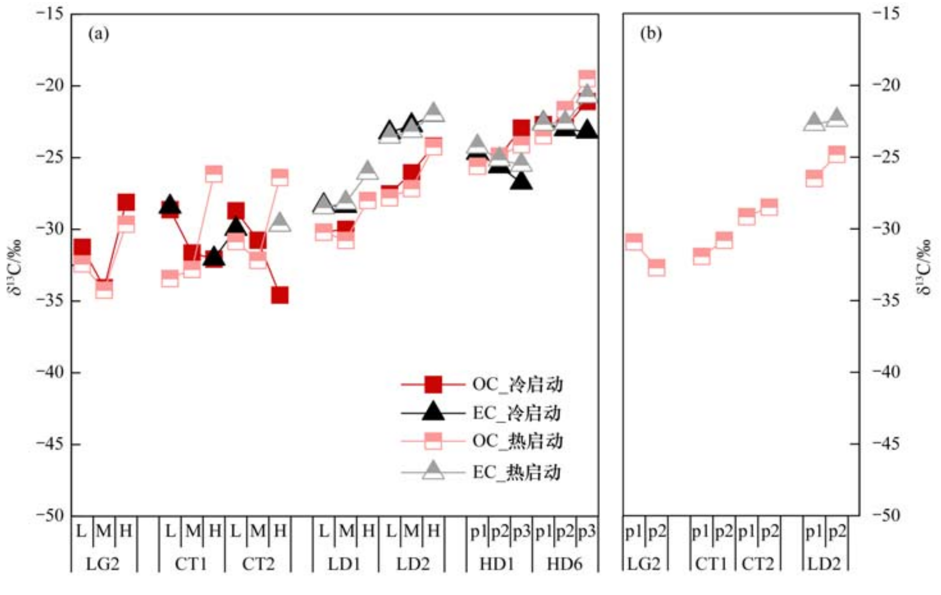

图 6　测试车辆各测试阶段排放的 OC 和 EC 中稳定碳同位素比值

Fig. 6　Stable carbon isotope in OC and EC in each stage of test cycles

3 结论

(1)总碳排放因子呈现重型柴油车 > 轻型柴油车 > 轻型汽油车的特征,轻型天然气车虽然在低速与中速阶段排放因子极低,但高速行驶阶段可达到重型柴油车的排放水平. 各型车冷启动的排放因子均高于热启动,NEDC 工况的排放因子整体低于 WLTC 工况.

(2)汽油车和天然气车各测试阶段排放 OC 均远高于元素碳 EC(OC/TC > 95%),柴油车 OC 与 EC 排放因子相近,各类车辆 OC/EC 都随测试车速的提高而上升.

(3) TC 稳定碳同位素比值大小为:汽油车[(-32.25 ± 2.27)‰] < 天然气车[(-30.69 ± 2.44)‰] < 轻型柴油车[(-25.85 ± 2.55)‰] < 重型柴油车[(-23.45 ± 1.68)‰]. EC 与 OC 分布特征与此类似,EC 重于 OC. 另外,车辆老化对碳质气溶胶排放因子和稳定碳同位素特征均有影响.

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位素组成及来源变化[J]. 矿物岩石地球化学通报, 2019, **38**(6): 1073-1080.
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HUANJING KEXUE
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CONTENTS

Assessing the Environmental and Health Co-benefits of Accelerated Energy Transition and Industrial Restructuring: A Case Study of the BTHS Region ······ YANG Xi, SUN Yi-sheng, CHANG Shi-yan, *et al.* (3627)
Synergistic Paths of Reduced Pollution and Carbon Emissions Based on Different Power Demands in China ······ XIANG Meng-yu, WANG Shen, LÜ Lian-hong, *et al.* (3637)
Evaluation Method and Application for Urban Carbon Peaking & Neutrality Performance ······ ZHANG Bao-liu, BAI Zi-han, ZHANG Nan, *et al.* (3649)
Revealing Driving Factors of Urban O_3 Based on Explainable Machine Learning ······ DONG Jia-qi, HU Dong-mei, YAN Yu-long, *et al.* (3660)
Sensitivity Analysis of Ozone Formation Using Response Surface Methodology ······ ZHU Yu-huan, CHEN Bing, ZHANG Ya-ru, *et al.* (3669)
Analysis of O_3 Sources in Yulin City in Summer Based on WRF-CMAQ/ISAM Model ······ WANG Yi-fan, TONG Ji-long, CHEN Yu-xiang, *et al.* (3676)
Atmospheric Ozone Concentration Prediction in Nanjing Based on LightGBM ······ ZHU Jia-ying, AN Jun-lin, FENG Yue-zheng, *et al.* (3685)
Meteorological Formation Mechanisms and Potential Sources of an Ozone Pollution Process in Winter of 2022 in Guangdong Province ······ LI Ting-yuan, CHEN Jing-yang, GONG Yu, *et al.* (3695)
Impact of Summer Tropospheric Ozone Radiative Forcing on Meteorology and Air Quality in North China ······ DU Nan, CHEN Lei, LIAO Hong, *et al.* (3705)
Identification of Impacts from Meteorology and Local and Transported Photochemical Generation on Ozone Trends in Changsha from 2018 to 2020 ······ YANG Jun, YANG Lei-feng, DING Hua, *et al.* (3715)
Spatio-temporal Variation and Multi-dimensional Detection of Driving Mechanism of $PM_{2.5}$ Concentration in the Chengdu-Chongqing Urban Agglomeration from 2000 to 2021 ······ XU Yong, GUO Zhen-dong, ZHENG Zhi-wei, *et al.* (3724)
Estimation of $PM_{2.5}$ Hourly Concentration in Sichuan Province Based on GTWR-XGBoost Model ······ WU Di, DU Ning, WANG Li, *et al.* (3738)
Transmission and Growth Characteristics of Severe $PM_{2.5}$ Pollution Events from 2013 to 2021 in Xingtai, Hebei ······ JIANG Qi, SHENG Li, JIN Yu-chen, *et al.* (3749)
Chemical Characteristics and Source Apportionment of Organic Aerosols in Urban Shanghai During Cold Season Based on High Time-resolution Measurements of Organic Molecular Markers ······ ZHU Shu-hui (3760)
Emission Factors of Carbonaceous Aerosol and Stable Carbon Isotope for In-use Vehicles ······ YU Ming-yuan, WANG Qian, FU Ming-liang, *et al.* (3771)
Composition Characteristics of Volatile Organic Compounds and Associated Contributions to Secondary Pollution in Shenyang Industrial Area in Summer ······ GUAN Lu, SU Cong-cong, KU Ying-ying, *et al.* (3779)
Characteristics of VOCs and Assessment of Emission Reduction Effect During the Epidemic Lockdown Period in Shenzhen Urban Area ······ YUN Long, LIN Chu-xiong, LI Cheng-liu, *et al.* (3788)
Characteristics of Organic Matter Composition and Oxidation Potential in Road Dust in Winter in Xi'an ······ WANG Qing-wen, CHEN Qing-cai, WANG Chao, *et al.* (3797)
Contamination Characteristics and Risk Assessment of Polycyclic Aromatic Compounds in Surface Dust of Suntuan Mining Area in Huaibei ······ XU Zhen-peng, QIAN Ya-hui, HONG Xiu-ping, *et al.* (3809)
Spatial-temporal Variation in Water Quality and Its Response to Precipitation and Land Use in Baiyangdian Lake in the Early Stage of the Construction of Xiong'an New Area ······ WANG Zi-ming, YANG Li-hu, SONG Xian-fang (3820)
Simulation of Pollution Apportionment and Optimization of Control Methods in Watershed Scale: A Case Study of the Shun'an Watershed in Tongling City ······ LIU Guo-wangchen, CHEN Lei, LI Jia-qi, *et al.* (3835)
Evaluation of Shallow Groundwater Quality and Optimization of Monitoring Indicators in Nanchang ······ ZHENG Zi-yin, CHU Xiao-dong, XU Jin-ying, *et al.* (3846)
Hydrogen and Oxygen Isotopic Characteristics and Influencing Factors of "Three Waters" in Shandian River Basin ······ YANG Li-na, JIA De-bin, GAO Rui-zhong, *et al.* (3855)
Distribution Characteristics and Influencing Factors of Abundant and Rare Planktonic Microeukaryotes in Jinsha River ······ YAN Bing-cheng, CUI Ge, SUN Sheng-hao, *et al.* (3864)
Bacterial Community Diversity in Channel Sediments of Different Disturbance Sections of the Jialing River ······ ZHU Lan-ping, ZHANG Tuo, LI Jia-ning, *et al.* (3872)
Effects of Reservoir Water Depth on Different Plankton Communities and Keystone Species of Network Interaction ······ WANG Xun, LIAO Qin, WANG Pei-fang, *et al.* (3881)
Correlation Between the Diversity Characteristics of Groundwater Bacterial Community and Environmental Factors in Typical Industrial Areas ······ WU Jian-qiang, ZHANG Shu-yuan, WANG Min, *et al.* (3892)
Effects of Fertilizer Application Strategy Adjustments on Nitrogen and Phosphorus Loss from Typical Crop Systems in Taihu Lake Region ······ YU Ying-liang, WANG Yi-zhi, YANG Bei, *et al.* (3902)
Estimation of Cropland Nitrogen Runoff Loss Loads in the Yangtze River Basin Based on the Machine Learning Approaches ······ ZHANG Yu-fu, PAN Zhe-qi, CHEN Ding-jiang (3913)
Classification and Identification of Non-point Source Nitrogen Pollution in Surface Flow of the Shangwu River Watershed in the Qiandao Lake Region ······ YU Ke, YAN Yan, TANG Zhang-xuan, *et al.* (3923)
Spatial Distribution of Nitrogen and Phosphorus Nutrients in the Main Stream and Typical Tributaries of Tuojiang River and Fujiang River ······ LI Zi-yang, ZHOU Ming-hua, XU Peng, *et al.* (3933)
Sediment Pollution and Dredging Effect of Waiqinhuai River ······ ZHANG Mu, REN Zeng-yi, ZHANG Man, *et al.* (3945)
Critical Review on Environmental Occurrence and Photochemical Behavior of Substituted Polycyclic Aromatic Hydrocarbons ······ GE Lin-ke, WANG Zi-yu, CAO Sheng-kai, *et al.* (3957)
Cadmium and Arsenic Interactions During Co-adsorption onto Goethite ······ SU Zi-xian, LIU Sai-hong, GUAN Yu-feng, *et al.* (3970)
Preparation of Catalyst Cyclodextrin-Fe-TAML to Activate H_2O_2 and Oxidize Organic Micropollutants in Water ······ LIU Qing-quan, CAI Ben-zhe, CAI Xi-yun (3978)
Performance and Reaction Mechanism of Co(Ⅱ) Mediated Activation of Peroxymonosulfate for Degrading Nitrilotris (Methylene Phosphonic Acid) ······ ZHU Jing-lin, WANG Shu (3990)
Change in Granulation Potential and Microbial Enrichment Characteristics of Sludge Induced by Microplastics ······ XIE Qing-fan, YU Nan, ZHANG Ni, *et al.* (3997)
Accumulation Characteristics and Probabilistic Risk Assessment of Cd in Agricultural Soils Across China ······ WANG Jing, WEI Heng, PAN Bo (4006)
Source Analysis and Pollution Assessment of Soil Heavy Metals in Typical Geological High Background Area in Southeastern Chongqing ······ JIANG Yu-lian, YU Jing, WANG Rui, *et al.* (4017)
Integrated Analysis on Source-exposure Risk of Heavy Metals in Farmland Soil Based on PMF Model: A Case Study in the E-waste Dismantling Area in Zhejiang Province ······ FANG Jia, HE Ying, HUANG Nai-tao, *et al.* (4027)
Pollution Characteristics and Risk Assessment of Polycyclic Aromatic Hydrocarbons in Farmland Soil and Crops in the Suburbs of Urumqi ······ FAN Yue, CAO Shuang-yu, Nuerla Ailijiang, *et al.* (4039)
Distribution and Driving Mechanisms of Antibiotic Resistance Genes in Desert-Oasis Continuum ······ HUANG Fu-yi, ZHOU Shu-yidan, SU Jian-qiang, *et al.* (4052)
Characteristics of Antibiotic Contamination of Soil in China in Past Fifteen Years and the Bioremediation Technology: A Review ······ ZHAO Xiao-dong, QIAO Qing-qing, QIN Xiao-rui, *et al.* (4059)
Modified Biochar for Remediation of Soil Contaminated with Arsenic and Cadmium: A Review ······ LÜ Peng, LI Lian-fang, HUANG Xiao-ya (4077)
Effect of Biogas Slurry Return to Field on Heavy Metal Accumulation in Soil-crop System: A Meta-analysis ······ ZHAO Qi-zhi, YANG Zhi-min, KONG Fan-jing, *et al.* (4091)
Remediation Effect of Two Iron-modified Biochars on Slightly Alkaline Arsenic and Cadmium Contaminated Soil ······ LIANG Xin-ran, HE Dan, ZHENG Zhao-hua, *et al.* (4100)
Effects of Straw Removal Measure on Soil Cd Bioavailability and Rice Cd Accumulation ······ WANG Zi-yu, ZHOU Hang, ZHOU Kun-hua, *et al.* (4109)
Application of Desulphurized Gypsum with Straw to Improve Physicochemical Properties of Saline-alkali Land in Yellow River Delta ······ ZHAO Hui-li, YU Jin-yi, LIU Tao, *et al.* (4119)
Biological Evaluation and Key Stress Factor Diagnosis of Compound Contaminated Soil Based on Environmental DNA ······ HUANG Xiang-yun, ZHONG Wen-jun, LIU Xun-jie, *et al.* (4130)
Comprehensive Quality Assessment of Soil-Maize Heavy Metals in High Geological Background Area ······ ZHANG Chuan-hua, WANG Zhong-shu, LIU Li, *et al.* (4142)
Ecological Risk and Health Risk of Heavy Metal Pollution in Vegetable Production System of Zhejiang Province ······ ZHANG Shu-min, LIU Cui-ling, YANG Gui-ling, *et al.* (4151)
Effect of Biochar with Phosphorus Fertilizer on Soil Nutrients, Enzyme Activity, and Nutrient Uptake of *Alfalfa* ······ LIU Xin-yu, WANG Dong-mei, ZHANG Ze-zhou, *et al.* (4162)
Effects of Low-density Polyethylene Microplastics on the Growth and Physiology Characteristics of *Ipomoea aquatica* Forsk ······ ZHOU Ying, JIANG Wen-ting, LIU Xun-yue, *et al.* (4170)
Effects of Biogas Slurry Application on Soil Microbial Communities Structure and Function During Wheat-rice Stubble Period ······ QIAO Yu-ying, XI Hui, LI Na, *et al.* (4179)
Analysis of Soil Bacterial Community Structure and Ecological Function Characteristics in Different Pollution Levels of Lead-zinc Tailings in Datong ······ LIU Ze-xun, ZHUANG Jia-yao, LIU Chao, *et al.* (4191)
Effect of Fire-deposited Charcoal on Soil Organic Carbon Pools and Associated Enzyme Activities in a Recently Harvested *Pinus massoniana* Plantation Subjected to Broadcast Burning ······ YAO Zhi, JIAO Peng-yu, WU Xiao-sheng, *et al.* (4201)