

目次

中国省域差异化碳达峰评价方法与应用刘润璞, 彭栓, 陈玉烁, 陈民, 张楠, Nihed Benani, 吕连宏, 阳平坚 (1233)

全生命周期视角下中国建筑碳排放空间关联网络演化及影响因素分析任晓松, 李昭睿 (1243)

京津冀地区城市三生空间碳代谢效率特征及演进模式田超, 程琳琳, 邵盈钊 (1254)

太原市“十四五”规划大气污染防治政策的CO₂协同效益评估肖婷玉, 束赕, 李慧, 王涵, 李俊宏, 严沁, 张文杰, 姜华 (1265)

湖南省工业领域碳减排与空气质量改善协同李楠, 刘弯弯, 朱书涵, 邢晓雯, 汤克勤, 王松伟, 白露 (1274)

“双碳”背景下河南省电力行业中长期控煤降碳路径张静, 杨萌, 张伟, 曹东, 赵静, 李勃, 薛英岚, 蒋洪强 (1285)

郑州市公交车队电动化减排降碳环境效益邹超, 汪亚男, 吴琳, 何敬, 倪经纬, 毛洪钧 (1293)

长江中游城市群城市化对PM_{2.5}浓度的多尺度驱动机制张政, 周廷刚, 周志衡, 昌悦 (1304)

天山北坡城市群PM_{2.5}浓度时空分布特征及影响因素分析王相男, 张喆, 刘方青 (1315)

天津市PM_{2.5}碳组分空间差异性来源解析武甫亮, 吴建会, 戴启立, 肖致美, 冯银厂 (1328)

贵阳市花溪城区大气PM_{2.5}中碳质气溶胶的变化特征及来源解析桂佳群, 杨员, 王显钦, 李云武, 闫广轩, 徐鹏 (1337)

新乡市大气PM_{2.5}中水溶性离子的污染特征、来源解析及气象影响分析刘桓嘉, 李岚清, 李焕莉, 任言, 许梦源, 贾梦珂, 刘恒志, 杨莹, 宋天颂, 洪启航 (1349)

2017~2018年冬季菏泽大气PM_{2.5}中金属元素特征及健康风险评估杜虹萱, 任丽红, 赵明升, 韩慧霞, 徐义生 (1361)

北京市臭氧污染跳变型特征及影响因素分析潘锦秀, 安欣欣, 刘保献, 李云婷, 李倩, 孙峰, 张章, 邱启鸿, 陈阳 (1371)

南京夏季大气臭氧光化学特征与敏感性分析罗丽彤, 章炎麟, 林煜棋, Ahsan Mozaffar, 曹梦瑶 (1382)

苏州市初夏臭氧污染成因及年际变化吴也正, 张鑫, 顾韵, 缪青, 魏恒, 熊宇, 杨倩, 吴斌, 沈文渊, 马强 (1392)

长江中游典型湖泊沉积物重金属分布特征、生态风险评估及溯源卢洪斌, 卢少勇, 李响, 张霖霖, 黄张根 (1402)

基于PCA-APCS-MLR模型的乌梁素海表层沉积物重金属时空分布及来源解析崔志谋, 史小红, 赵胜男, 卢俊平, 张昊, 刘莹慧, 郭鑫, 王彦隽 (1415)

重庆市长寿湖水表层水体重金属时空分布及风险评价张瑞溪, 刘娅君, 罗泳楠, 李杰芹, 李彩霞, 李佳佳, 张成 (1428)

长江流域微塑料污染特征及生态风险评估李思琼, 王华, 储林佑, 曾一川, 闫雨婷 (1439)

宜昌市东山运河微塑料污染评估及年排放量估算丁爽, 李卫明, 张续同, 刘子健, 高雅坤, 李映成, 王芳炜 (1448)

汜水河(荥阳段)入河排污口水体微塑料赋存特征及风险评估赵长民, 和兵, 李和通, 张瑞琪, 李银月, 张发文, 桂新, 马丽 (1457)

宁夏入黄排水沟中药物和个人护理品的污染特征与生态风险评估高礼, 李凌云, 郑兰香, 吴海娟, 陶红, 刘邓超 (1468)

浙南瓯江流域水体抗生素污染特征及风险评估钟奕昕, 李立湘, 吴鑫, 周施阳, 姚飞延, 董好刚 (1480)

鄱阳湖沉积物中多环芳烃的时空分布及源解析马妍, 孙晨, 毕茹乐, 张波涛, 刘艳, 邵鹏, 刘统, 王圣瑞, 钟文军 (1492)

杭州湾南岸20a水质净化功能变化及预测王珊珊, 曹公平, 徐明伟, 黄君宝, 曾剑 (1502)

不同缓冲区的土地利用方式对地表水水质的影响:以海河流域天津段为例代孟均, 张兵, 杜倩倩, 孙季琰, 田蕾, 王义东 (1512)

长江流域安庆段浅层地下水水化学特征及控制因素刘海, 宋阳, 李迎春, 魏伟, 赵国红, 王旭东, 黄健敏 (1525)

富营养化湖泊藻华腐解产生的溶解性有机质动态变化及其环境效应张瑾, 陈明滢, 郝智能, 钟寰, 何欢, 雷沛 (1539)

紫外光活化亚硫酸盐降解水中卡马西平林涛, 苑宇杰 (1553)

再生水消毒副产物的检测、生成与控制廖雨枫, 王正, 潘昀, 李爱民 (1561)

3种人工湿地基质材料对氨氮的吸附特性何强, 陈博文, 杨雨静, 周全, 刘彦君, 王志刚, 程呈 (1577)

基于改进遥感生态指数的青藏公路那(曲)安(多)段生态环境评估及驱动机制分析傅楷翔, 贾国栋, 余新晓, 王旭 (1586)

基于AWRSEI的岱海流域生态环境质量时空演变及驱动因子分析赵嘉丽, 李兴, 孙冰 (1598)

定量评估气候变化对长江中下游地区植被GPP_{GS}变化的影响徐勇, 盘钰春, 邹滨, 郑志威, 郭振东 (1615)

基于Meta分析的煤矿区植被恢复对土壤有机碳储量的影响李健明, 康雨欣, 蒋福祺, 宋明丹, 祁凯斌, 卢素锦, 李正鹏 (1629)

连续周年轮作休耕对土壤团聚体稳定性及有机碳的影响鲁泽让, 李永梅, 杨春怀, 夏梓泰, 程伟威, 王自林, 赵吉霞, 范茂攀 (1644)

4种改良剂对酸性紫色土肥力及活性有机碳组分的影响丁馨茹, 严宁珍, 王子芳, 李志琦, 黄容, 王洋, 代文才, 高明 (1655)

不同植茶年限土壤氮素组分变化及其与环境因子关系邵奇, 吴涛, 解雪峰, 徐梓晴, 李文琦, 蒋国俊, 张建珍, 徐飞 (1665)

黄河下游典型湿地土壤养分及其生态化学计量特征王传盈, 王凯月, 王浩然, 张梦迪, 周云凯 (1674)

覆膜年限和有机肥施用对花生田耕层土壤微塑料赋存特征的影响宋宁宁, 李梦佳, 王学霞, 刘君, 王芳丽, 宗海英, 黄小丽, 王斌, 梁丽娜 (1684)

秸秆还田和添加生物炭对热带地区稻菜轮作体系中淹水后土壤温室气体排放的影响胡天怡, 车佳玥, 胡煜杰, 陈琦琦, 张冬明, 雷菲, 曾建华, 汤水荣, 伍延正, 孟磊 (1692)

耕作深度调控秸秆还田对农田土壤呼吸的影响陈曦, 张彦军, 邹俊亮, 李天姿, 于媛, 李晶 (1702)

基于遥感时-空-谱特征及随机森林模型的土壤重金属空间分布预测王泽强, 张冬有, 徐夕博, 王兆鹏, 杨东宇, 宋晓宁 (1713)

黄河流域农田土壤重金属污染特征及其优先控制源分析李军, 李旭, 李开明, 焦亮, 台喜生, 臧飞, 曹素珍 (1724)

广西贺州市典型矿区周边耕层土壤Cd通量特征杨烨宇, 李程, 杨忠芳, 张起钻, 邹胜章, 宋淑娟, 蔡贺清 (1739)

基于信息扩散模型的沔东新区土壤重金属潜在生态风险评估杨楠楠, 韩玲, 刘明 (1749)

湘西地区土壤重金属污染溯源分析及环境质量评价肖凯琦, 徐宏根, 甘杰, 戴亮亮, 李毅, 李凯, 许青阳, 张俊, 邓世民, 李颖 (1760)

典型行业再利用土壤重金属含量分布、来源解析及生态风险评估沈城, 王文娟, 沙晨燕, 谢雨晴, 王敏, 吴健 (1769)

省级尺度土壤As迁移转化与水稻安全种植区划:以贵州省为例董心月, 吴勇, 周子寒, 王佛鹏, 张云霞, 宋波 (1781)

谷壳灰硅肥改善土壤质量降低水稻镉累积的效应易轩韬, 欧阳坤, 辜娇峰, 李倩, 游萍, 周航, 廖柏寒 (1793)

EDDS对土壤铜镉有效性及蓖麻吸收转运的影响刘文英, 吴刚, 胡红青 (1803)

叶面凋制剂对复合污染农田小麦籽粒Cd、As和Pb累积的阻控效应肖冰, 王秋实, 高培培, 赵全利, 杨威, 王钊, 刘文菊, 薛培英 (1812)

民勤绿洲退耕地土壤微生物群落结构与功能多样性特征李常乐, 张富, 王理德, 赵赫然, 赵学成, 张恒平 (1821)

宏基因组揭示紫色土中邻苯二甲酸酯去除的微生物学机制李雨桐, 余海, 刘坤, 柏宏成, 汪军, 朱正杰 (1830)

养鸡场空气微生物污染及工人呼吸暴露风险白渔樵, 孙兴滨, 仇天雷, 郭雅志, 高敏, 王旭明 (1840)

玛瑙河多环境介质和铜锈环螺体内微塑料的赋存特征高雅坤, 李卫明, 张续同, 刘子健, 李映成, 丁爽, 王芳炜, 刘流 (1849)

不同官能团微塑料对斑马鱼胚胎菌群和代谢功能的胁迫效应闫振华, 张燕, 包旭辉, 朱培元, 陈玉芳 (1859)

2017~2018年冬季菏泽大气PM_{2.5}中金属元素特征及健康风险评估

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摘要: 于山东省菏泽市采集了2017年10月15日至2018年1月31日期间菏泽学院、华润制药和污水处理厂共3个采样点的大气PM_{2.5}样品, 利用电感耦合等离子体质谱仪(ICP-MS)测定PM_{2.5}中21种金属元素的浓度, 并讨论元素富集程度、评估重金属的健康风险和潜在生态风险. 结果表明, 采样期间3个采样点中 $\rho(\text{PM}_{2.5})$ 范围为26.7~284.1 $\mu\text{g}\cdot\text{m}^{-3}$ 且浓度值差别不大, 均处于较高污染水平; 3个采样点金属元素中K浓度最高, 分别占总量的31.03%、39.47%和38.43%, 主要由于菏泽作为较大农业城市, 其秋冬季生物质燃烧贡献率较高; 3个采样点微量元素中 $\rho(\text{Zn})$ 最高, 分别为89.70、84.21和67.68 $\text{ng}\cdot\text{m}^{-3}$. 富集因子结果表明, Zn、Pb、Sn、Sb、Cd和Se的富集因子值均高于100, 其中Cd和Se的富集因子分别高于2000和4000, 受人为活动影响显著, 可能与工业生产、金属冶炼、道路源和燃煤排放等有关. 健康风险结果表明, As存在一定的潜在非致癌风险(儿童和成人HQ>0.1), 3个采样点对儿童和成人均存在综合潜在非致癌风险(HI>0.1)和一定的潜在致癌风险(CRT>1 $\times 10^{-6}$), 其中污水处理厂对成人的致癌风险较为显著(CRT>1 $\times 10^{-4}$), 成人的致癌风险略高于儿童可能与成人室外活动时间较长和PM_{2.5}暴露量更高有关. 潜在生态风险值最高的元素为Cd、As和Pb, 其中Cd表现为极高的潜在生态风险, 应引起重视; 3个采样点均呈现出极高的综合潜在生态风险, 强度在空间上表现为: 菏泽学院>华润制药>污水处理厂.

关键词: PM_{2.5}; 重金属; 富集因子; 健康风险评估; 潜在生态风险

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Characterization of Metal Elements in Atmospheric PM_{2.5} and Health Risk Assessment in Heze in Winter from 2017 to 2018

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Abstract: Atmospheric PM_{2.5} samples were collected in Heze, Shandong Province, from a total of three sampling sites at Heze College, Huarun Pharmacy, and a wastewater treatment plant between October 15, 2017 and January 31, 2018, to determine the concentrations of 21 metal elements in PM_{2.5} using inductively coupled plasma mass spectrometry (ICP-MS). The degree of elemental enrichment was also discussed, the health risks and potential heavy metal ecological risks were assessed. The results showed that $\rho(\text{PM}_{2.5})$ ranged from 26.7 to 284.1 $\mu\text{g}\cdot\text{m}^{-3}$ at the three sampling sites during the sampling period, and the concentration values did not differ significantly, all of which were at high pollution levels. The highest concentrations of K were found in the three sampling sites, accounting for 31.03%, 39.47%, and 38.43% of the total, respectively, mainly due to the high contribution of biomass burning in autumn and winter in Heze, a large agricultural city. The highest concentrations of Zn, 89.70, 84.21, and 67.68 $\text{ng}\cdot\text{m}^{-3}$, were found in the trace elements at the three sampling sites, respectively. The enrichment factor results showed that the enrichment factor values of Zn, Pb, Sn, Sb, Cd, and Se were higher than 100, among which the enrichment factors of Cd and Se were higher than 2000 and 4000, respectively, which were significantly influenced by anthropogenic activities and might have been related to industrial production, metal smelting, road sources, and coal combustion emissions. The health risk results showed that there was some potential non-carcinogenic risk (HQ > 0.1 for children and adults) for As and a combined potential non-carcinogenic risk (HI > 0.1) and some potential carcinogenic risk (CRT > 1 $\times 10^{-6}$) for both children and adults at the three sampling sites. There was a more significant carcinogenic risk (CRT > 1 $\times 10^{-4}$) for adults at the wastewater treatment plant, and the slightly higher carcinogenic risk for adults than that for children may have been related to the longer outdoor activity and higher PM_{2.5} exposure for adults. The elements with the highest potential ecological risk values were Cd, As, and Pb, with Cd exhibiting a very high potential ecological risk that should be taken seriously. All three sampling sites showed a very high combined potential ecological risk, with the intensity spatially expressed as Heze College > Huarun Pharmacy > wastewater treatment plant.

Key words: PM_{2.5}; heavy metals; enrichment factor; health risk assessment; potential ecological risks

近年来,随着中国经济的快速发展,颗粒物成为许多城市大气环境中的首要污染物,特别是对人体健康、大气能见度和辐射平衡有较大影响的细颗粒物(PM_{2.5}),已经成为社会关注的焦点^[1-4]. 作为PM_{2.5}的一个重要组成部分,金属元素可以通过自然和人为来源进入大气^[5],其中重金属毒性大、难降解且易富集,在生物体内累积会对心肺等部位产生危害^[6,7].

因此,研究重金属元素对于PM_{2.5}的溯源,了解其迁移机制、变化规律以及保护人体健康都具有重要意义.

PM_{2.5}中重金属元素污染特征和健康风险引起了

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学者的关注,并进行了研究.徐静等^[8]研究指出,北京城区 Cu、Zn、As、Cd 和 Pb 中度污染且较大程度受到人类活动的影响;刘佳等^[9]对重庆雾-霾期 PM_{2.5} 中 23 种金属元素的研究证实了其中主要的无机元素是 K、Fe、Ca、Zn、Pb、Mn、Ba 和 As;杨毅红等^[10]研究表明珠海市大气 PM_{2.5} 中致癌风险贡献最大的金属元素是 Cr 和 Pb,气候、地理位置和经济发展使不同地区大气颗粒物中重金属的分布特征存在明显差异.秦伟等^[11]研究发现石家庄市采暖季 PM_{2.5} 中地壳元素无富集,重金属元素呈现一定程度的富集,Cr 元素富集程度较高;李雪梅等^[12]研究表明 2017 年山西大学城 PM_{2.5} 中 Ca 浓度最高,且 Cr 浓度超过我国环境空气质量标准年平均浓度阈值的 104 倍.以上大部分对 PM_{2.5} 载带金属元素的研究主要集中在元素特征上,针对生态健康风险及同一城市多个采样点同步开展的研究还相对缺乏.

本研究在山东省菏泽市的菏泽学院、华润制药和污水处理厂共 3 个采样点于 2017~2018 年冬季期间同步采集和分析大气 PM_{2.5} 样品,从富集程度等方面探究其中金属元素的分布特征;同时,采用美国环保署开发的健康风险评估模型探究重金属元素通过呼吸途径对菏泽市不同人群产生的健康风险并评估

其潜在生态风险.本研究结果可为提高认知环境空气中重金属的污染特征和健康影响提供参考,并为进一步的污染防治和保护公众健康提供科学依据.

1 材料与方法

1.1 采样地位信息

菏泽市位于山东省西南部(34°39'~35°52'N, 114°45'~116°25'E),有研究统计了 2015~2016 年菏泽市区 $\rho(\text{PM}_{2.5})$ 均值为 $100.9 \mu\text{g}\cdot\text{m}^{-3}$ ^[13],明显高于我国 PM_{2.5} 环境空气质量标准(GB 3095-2012)二级阈值($75 \mu\text{g}\cdot\text{m}^{-3}$)^[14].本研究参照《环境空气质量监测规范(试行)》及相关要求、原则和质控标准,并综合考虑城市功能区和人口密度等,在菏泽市设立了菏泽学院、华润制药和污水处理厂共 3 个环境采样点同步采集 PM_{2.5} 滤膜样品.采样点位分布及周边情况见图 1 和表 1.

表 1 菏泽市环境样品采样点位置及周边情况
Table 1 Location and surroundings of environmental sampling sites in Heze

采样点位	经纬度	周边情况
菏泽学院	35°16.16'N; 115°27.60'E	教育、住宅和商业区
华润制药	35°15.38'N; 115°30.95'E	工业、交通和住宅
污水处理厂	35°13.35'N; 115°31.60'E	工业、住宅和交通



图 1 菏泽采样点的位置示意

Fig. 1 Locations of the sampling sites in Heze

1.2 样品采集和分析

在菏泽学院、华润制药和污水处理厂这 3 个采样点采用直径为 47mm 的 Teflon 滤膜和 TH-16A 型四通道大气颗粒物智能采样仪(武汉天虹,中国)同步采集 PM_{2.5},采样流量为 $16.7 \text{ L}\cdot\text{min}^{-1}$;采样日期为 2017 年 10 月 15 日至 2018 年 1 月 31 日,采样时段为每日 10:00 至次日 09:00,共 23 h;排除设备故障等突发状况后的 3 个采样点有效样品数分别为 92、99 和 100

个.采样前后,将滤膜置于 $(20\pm 2.5)^\circ\text{C}$ 恒温 and $(50\pm 5)\%$ 恒湿条件下平衡 48 h,采用十万分之一分析天平(精度 0.01mg , Sartorius, 德国)称量滤膜质量,储存在密封袋中备用.剪取 1/2 滤膜样品置于聚四氟乙烯消解管中,加入 3 mL 优级纯 HNO_3 、1 mL 优级纯 HClO_4 和 1 mL MOS 级 HF 常温预消解 45 min,然后升温进行三段微波消解程序(120°C 保持 2 min、 170°C 保持 8 min、 190°C 保持 50 min),消解完成后在 180°C 的条件

下赶酸3 h,冷却至室温后用超纯水定容并静置避光保存以待分析. 大气颗粒物样品中的Na、Mg、Al、K、Ca、Ti、V、Cr、Mn、Fe、Co、Ni、Cu、Zn、As、Se、Cd、Sn、Sb、Ba和Pb共21种金属元素的浓度采用电感耦合等离子体质谱仪(ICP-MS, Agilent 7800, 美国)测定. 每10个样品分析一个空白膜,做平行样品和标准样品分析,需控制平行样品相对误差小于20%、标准样品回收率在80%~120%之间,以减小误差影响^[15-17].

1.3 统计分析 with 计算

1.3.1 富集因子

颗粒物中元素的来源和形成机制不同、富集程度也不同,富集因子(enrichment factor, EF)常用于研究元素在大气颗粒物中的富集情况并判断和评估其来源^[18]. 本研究采用菏泽地区母质层测定的土壤均值作为背景值,并选取性质稳定的常用保守元素Al作为参考元素^[19],计算公式如下^[20]:

$$EF = \frac{(c/c_{ref})_{PM_{2.5}}}{(c/c_{ref})_{soil}} \quad (1)$$

式中,EF为富集因子值; c 为元素浓度, c_{ref} 为参考元素浓度, $\mu\text{g}\cdot\text{m}^{-3}$; $(c/c_{ref})_{PM_{2.5}}$ 和 $(c/c_{ref})_{soil}$ 分别为样品和土壤背景中研究元素与参考元素的比值. 相关研究表明,

表2 不同人群重金属健康风险参数取值^[28,29]

Table 2 Health risk parameters for heavy metals in different populations

参数	符号	单位	人群	
			儿童	成人
暴露频率	EF	$\text{d}\cdot\text{a}^{-1}$	180	180
暴露年限	ED	a	6	24
暴露时间	ET	$\text{h}\cdot\text{d}^{-1}$	24	24
平均寿命	AT_n	h	ED×365×24(非致癌作用)	ED×365×24(非致癌作用)
			70×365×24(致癌作用)	70×365×24(致癌作用)

非致癌健康风险以危险商值(HQ)衡量,危险指数(HI)用于评估金属元素的综合非致癌风险;致癌健康风险以危险商值(CR)衡量,危险指数(CRT)用于评估金属元素的综合致癌风险. 公式如下^[26]:

$$HI = \sum HQ = \frac{EC}{RfC \times 1000} \quad (4)$$

$$CRT = \sum CR = EC \times IUR \quad (5)$$

式中,RfC为单一污染物的参考剂量, $\text{mg}\cdot\text{m}^{-3}$;IUR为吸入单位风险, $(\mu\text{g}\cdot\text{m}^{-3})^{-1}$,取值见表3. 通常来说,当 $HQ < 0$ 时不存在非致癌风险; $0.1 \leq HQ(HI) \leq 1$ 时存在潜在非致癌风险; $HQ(HI) > 1$ 时存在显著非致癌风险^[30]. 当 $CR(CRT) < 10^{-6}$ 时,该金属元素不存在致癌风险; $10^{-6} \leq CR(CRT) \leq 10^{-4}$ 时存在潜在致癌风险; $CR(CRT) > 10^{-4}$ 时存在显著的致癌风险,有必要采取措施降低风险^[20].

EF≤10表示该元素主要源于自然源,表现为轻度富集或无富集;10<EF≤100表示主要源于人为源,表现为中度富集;EF>100表示受到较严重的人为活动影响,EF值越大影响越显著,表现为高度富集或超富集^[21].

1.3.2 健康风险评估

一般来说,重金属可以通过呼吸系统、皮肤接触和手口摄入这3种途径进入人体^[22,23],且其健康风险可分为致癌风险和非致癌风险^[24]. 由于PM_{2.5}主要通过呼吸影响人体健康,因此本研究仅对吸入途径产生的健康风险展开评估,考虑到毒性参数的可获取性,采用美国EPA健康风险评估模型对3个采样点大气PM_{2.5}载带的As、Cd、Cr、Ni、V、Cu、Pb和Zn共8种金属元素进行计算,量化其致癌和非致癌风险. 其中,Cu、Pb、V和Zn构成非致癌风险,而As、Cd、Cr和Ni构成致癌风险^[25]. 暴露量计算公式如下^[26]:

$$EC = c \times \frac{ET \times EF \times ED}{AT_n} \quad (3)$$

式中,EC为吸入接触浓度, $\mu\text{g}\cdot\text{m}^{-3}$;c为重金属元素浓度, $\mu\text{g}\cdot\text{m}^{-3}$;ET为暴露时间, $\text{h}\cdot\text{d}^{-1}$;EF为暴露频率, $\text{d}\cdot\text{a}^{-1}$;ED为暴露年限,a; AT_n 为平均寿命,h. 由于不同价态Cr元素的健康影响不同,本研究参考大气中Cr(VI)浓度是Cr的1/7的比例系数进行转化^[27],其他参数取值见表2.

表3 金属元素通过呼吸途径进入人体的反应参数^[17]

Table 3 Reaction parameters for the entry of elemental metals into the body via the respiratory route

重金属种类	RfC/ $\text{mg}\cdot\text{m}^{-3}$	IUR/ $(\mu\text{g}\cdot\text{m}^{-3})^{-1}$
Pb	3.52×10^{-3}	0.000 012
As	1.50×10^{-5}	0.0043
Zn	3.01×10^{-1}	— ¹⁾
Cu	4.02×10^{-2}	—
V	1.00×10^{-4}	—
Cr	1.00×10^{-4}	0.084
Ni	9.00×10^{-5}	0.000 26
Cd	1.5×10^{-5}	0.001 8

1)“—”表示没有相关数据

1.3.3 潜在生态风险评估

潜在生态风险指数法可以评价金属元素对生态系统的潜在风险^[31],潜在的生态风险指数(RI)用于

评估颗粒物中重金属污染的程度^[20,32,33],计算公式如下^[33]:

$$E_r = T_r \times \frac{c}{c_{soil}} \quad (6)$$

$$RI = \sum E_r \quad (7)$$

式中, E_r 为潜在生态风险系数; T_r 为毒性系数,参考相关研究,Pb、As、Zn、Cu、V、Cr、Ni和Cd的毒性系数分别取为5、10、1、5、2、2、5和30^[31,34]; c 为元素浓度, c_{soil} 为元素在土壤背景中的浓度, $\mu\text{g}\cdot\text{m}^{-3}$. 潜在生态风险评价指标和等级划分见表4.

表4 潜在生态风险评价分级^[35]

Table 4 Potential ecological risk evaluation grading

E_r	RI	生态风险等级
<40	<150	I 低风险
40~80	150~300	II 中等风险
80~160	300~600	III 较高风险
160~320	600~1 200	IV 高风险
>320	>1 200	V 极高风险

2 结果与讨论

2.1 $\text{PM}_{2.5}$ 及金属元素浓度

2.1.1 $\text{PM}_{2.5}$ 浓度

由图2可知,2017~2018冬季,菏泽学院、华润制药和污水处理厂中 $\rho(\text{PM}_{2.5})$ 范围分别为28.6~260.4、28.7~284.1和26.7~265.1 $\mu\text{g}\cdot\text{m}^{-3}$;均值为(101.8±57.0)、(104.1±55.4)和(105.2±57.4) $\mu\text{g}\cdot\text{m}^{-3}$,是我国 $\text{PM}_{2.5}$ 环境空气质量标准二级阈值(75 $\mu\text{g}\cdot\text{m}^{-3}$)^[14]的1.36、1.39和1.40倍,是世界卫生组织标准值(15 $\mu\text{g}\cdot\text{m}^{-3}$)的6.79、6.94和7.01倍^[36],表明冬季菏泽地区 $\text{PM}_{2.5}$ 处于较高的污染水平.除此之外,各采样点的 $\text{PM}_{2.5}$ 浓度差别不大,且逐日变化趋势呈现出高度的一致性,表明菏泽大气中 $\text{PM}_{2.5}$ 污染在一定程度上具有区域特征.与国内典型城市相比,菏泽市的 $\text{PM}_{2.5}$ 浓度值远高于厦门^[37](21.9 $\mu\text{g}\cdot\text{m}^{-3}$)和珠江三角洲^[38]

(50.0 $\mu\text{g}\cdot\text{m}^{-3}$);与郑州^[39](108.0 $\mu\text{g}\cdot\text{m}^{-3}$)相当.

2.1.2 金属元素浓度

由图3和图4可知,2017~2018年冬季菏泽学院、华润制药和污水处理厂的大气 $\text{PM}_{2.5}$ 中21种金属元素累计浓度分别为7.30、6.01和5.47 $\mu\text{g}\cdot\text{m}^{-3}$,占 $\text{PM}_{2.5}$ 浓度总量的7.17%、5.77%和5.20%.与国内其他城市相比,金属元素在 $\text{PM}_{2.5}$ 中的占比接近天津(5.8%)、略高于青岛(3.6%)^[17].金属元素浓度排序依次为:K>Fe>Ca>Al>Na>Mg>Zn>Ti>Pb>Mn>Cu>Ba>Sn>As>Cr>Se>Sb>Ni>Cd>V>Co(菏泽学院)、K>Ca>Al>Fe>Na>Mg>Zn>Ti>Pb>Mn>Cu>Ba>Sn>As>Cr>Sb>Se>Ni>Cd>V>Co(华润制药)和K>Ca>Fe>Al>Na>Zn>Mg>Pb>Mn>Ti>Cu>Ni>Ba>Cr>Sn>As>Sb>Se>Co>Cd>V(污水处理厂).菏泽学院、华润制药和污水处理厂K浓度最高,分别占总元素浓度的31.03%、39.47%和38.43%,主要源于菏泽市是一个较大的农业城市,秋冬季生物质燃烧的贡献率较高,除了家庭使用秸秆外,收获季节露天焚烧秸秆的情况有所增加^[13].其次,菏泽学院、华润制药和污水处理厂的地壳元素(Na、Mg、Al、K、Ca和Fe)的占比也较高,占金属元素总量的64.05%、61.52%和63.23%.

重金属元素虽然浓度值较低,但其对人体健康影响较大,应当引起重视.重金属元素中以Zn和Pb浓度最高,其中菏泽学院、华润制药和污水处理厂的 $\rho(\text{Zn})$ 为89.70、84.21和67.68 $\text{ng}\cdot\text{m}^{-3}$,明显低于沈阳(206.4 $\text{ng}\cdot\text{m}^{-3}$)^[40];其次, $\rho(\text{Pb})$ 分别为56.60、59.35和49.54 $\text{ng}\cdot\text{m}^{-3}$,与沈阳(58.4 $\text{ng}\cdot\text{m}^{-3}$)^[40]和珠海(59.3 $\text{ng}\cdot\text{m}^{-3}$)^[10]相当、远低于郑州(136.6 $\text{ng}\cdot\text{m}^{-3}$)^[41].由于Zn和Pb主要来源于工业铅锌冶炼和机动车尾气排放,表明冶金工业和交通排放对菏泽地区的影响不容忽视.我国环境空气质量标准对大气中 $\rho(\text{Cd})$ 、 $\rho(\text{As})$ 和 $\rho[\text{Cr}(\text{VI})]$ 的阈值分别为5、6和0.025 $\text{ng}\cdot\text{m}^{-3}$.从3个采样点的 $\text{PM}_{2.5}$ 中重金属浓度可知,荷

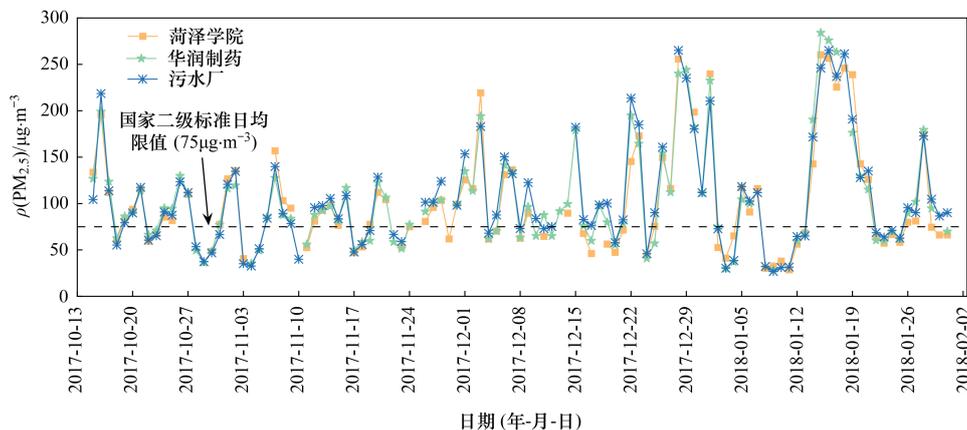


图2 菏泽学院、华润制药和污水处理厂 $\text{PM}_{2.5}$ 浓度

Fig. 2 $\text{PM}_{2.5}$ concentrations in Heze College, Huarun Pharmacy, and Wastewater Treatment Plant

泽学院、华润制药和污水处理厂的 $\rho(\text{Cd})$ 均小于环境空气质量标准阈值($5 \text{ ng}\cdot\text{m}^{-3}$)^[42],对采样点的污染影响较小;菏泽学院和华润制药 $\rho(\text{As})$ 均值达到了空气质量标准阈值($6 \text{ ng}\cdot\text{m}^{-3}$)^[42],污水处理厂As浓度均低于阈值;此外,菏泽学院、华润制药和污水处理厂 $\rho(\text{Cr})$ 均值超过了空气质量标准阈值($0.025 \text{ ng}\cdot\text{m}^{-3}$)^[42].由此说明菏泽地区As和Cr重金属污染应该引起重视,可能与菏泽的一些与金属制造业有关的企业,如菏泽冶炼厂、菏泽安兴铁厂等排放有关.因此,工业冶金和燃煤活动对菏泽大气PM_{2.5}中重金属的影响较大.与国内其他城市相比,3个采样点的 $\rho(\text{As})$ 均远低于佛山的($76.6 \text{ ng}\cdot\text{m}^{-3}$)^[43],与南京的($7.1 \text{ ng}\cdot\text{m}^{-3}$)^[44]相当; $\rho(\text{Cr})$ 与沈阳的($6.6 \text{ ng}\cdot\text{m}^{-3}$)^[40]

相当,远低于石家庄的($31.4 \text{ ng}\cdot\text{m}^{-3}$)^[45].

从时间上看,菏泽学院、华润制药和污水处理厂这3个采样点的PM_{2.5}中金属元素均表现出类似的逐日变化规律,且与PM_{2.5}浓度时间变化大体一致(图3和图4).空间变化表现为菏泽学院的地壳元素浓度整体高于华润制药和污水处理厂,这可能是由于菏泽学院采样点位于近郊,周围的裸露土壤面积较大且存在一定的建筑施工.此外,菏泽学院和华润制药中Ti和Zn浓度均高于污水厂,且菏泽学院的Fe浓度最高,说明周边可能存在更多的交通源;污水处理厂中Cr、Mn和Ni浓度较高,可能是因为污水厂靠近钢铁、燃料等工业设施,并有更多的重质油燃烧排放等^[46];其余微量元素浓度值在3个采样点差异较小.

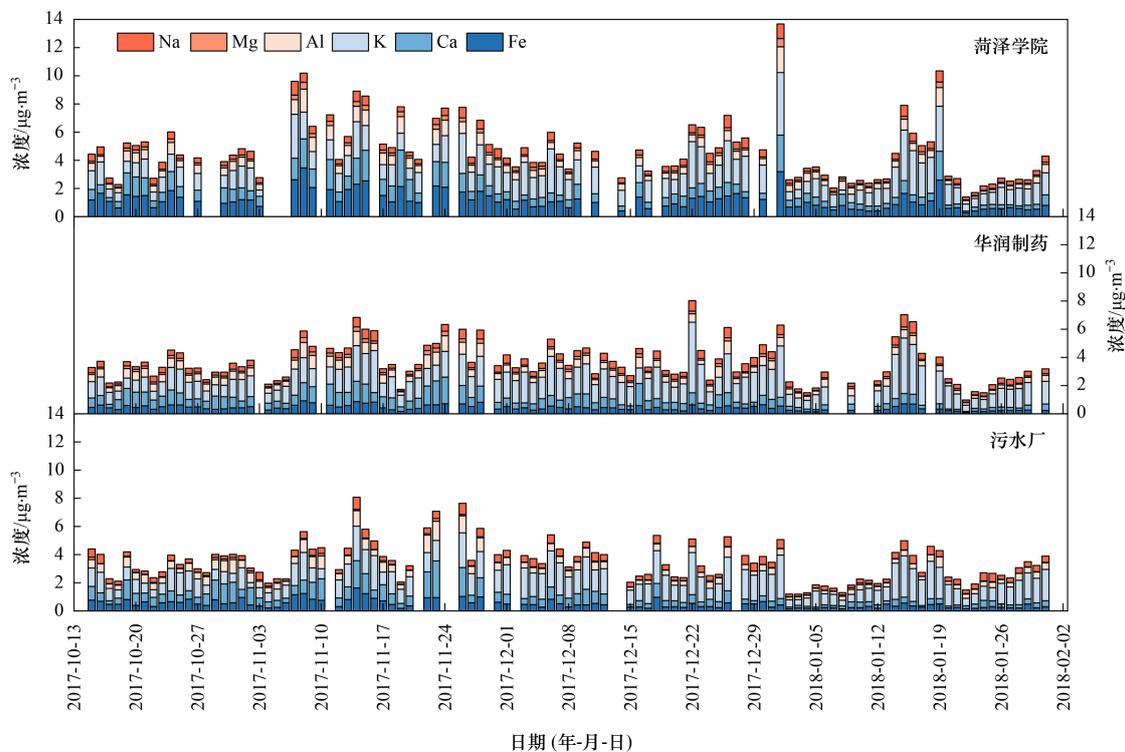


图3 菏泽学院、华润制药和污水处理厂地壳元素浓度

Fig. 3 Concentration of crustal elements in Heze College, Huarun Pharmacy, and Wastewater Treatment Plant

2.2 富集因子

图5显示了以Al为参考元素的重金属富集因子值.菏泽学院、华润制药和污水处理厂中Mg、Ti、Ba、V、Na、Fe、Mn、Ca和Co(污水处理厂)的富集因子值均值小于10,表明这些元素主要源于自然源;K、Ni、Cu、As和Cr(华润制药除外)的富集因子值在10~100之间,表明主要来自人为源.3个采样点中,Zn、Pb、Sn、Sb、Cd和Se的富集因子值均高于100,其中Cd和Se的富集因子分别高于2000和4000,为高度富集,受人为影响严重.其中,Cd可能与工业排放的废气有关,Se主要源于燃煤和机动车源等,Sn和Sb主要来源于金属冶炼,Pb可能主要来源于燃煤^[47-49],因

此造成极重污染的金属元素可能主要与工业生产、金属冶炼、道路源和燃煤排放等人为活动有关.污水处理厂的Mn、Cr、Co和Ni的富集因子大于其他两个采样点,这可能是由于污水处理厂及其周边存在较多的燃煤、重质油燃烧和钢铁等冶金工业并有一定的交通排放^[16].

2.3 健康风险评估

根据US EPA推荐的风险评估模型,分别探究了菏泽市菏泽学院、华润制药和污水处理厂大气PM_{2.5}载带的As、Cd、Cr、Ni、V、Cu、Pb和Zn共8种金属元素产生的致癌和非致癌健康风险,表5给出了各金属元素具体非致癌风险值.3个采样点非致癌风险的主

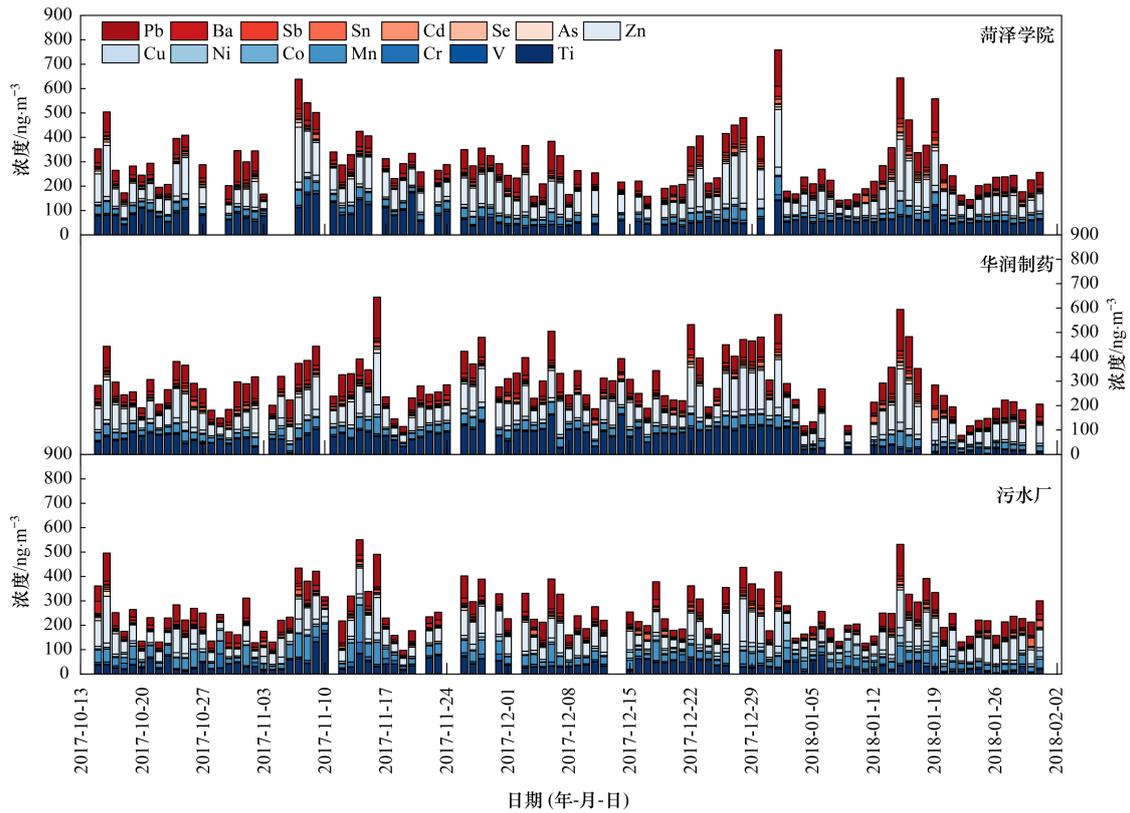


图4 菏泽学院、华润制药和污水处理厂微量元素浓度

Fig. 4 Trace element concentrations in Heze College, Huarun Pharmacy, and Wastewater Treatment Plant

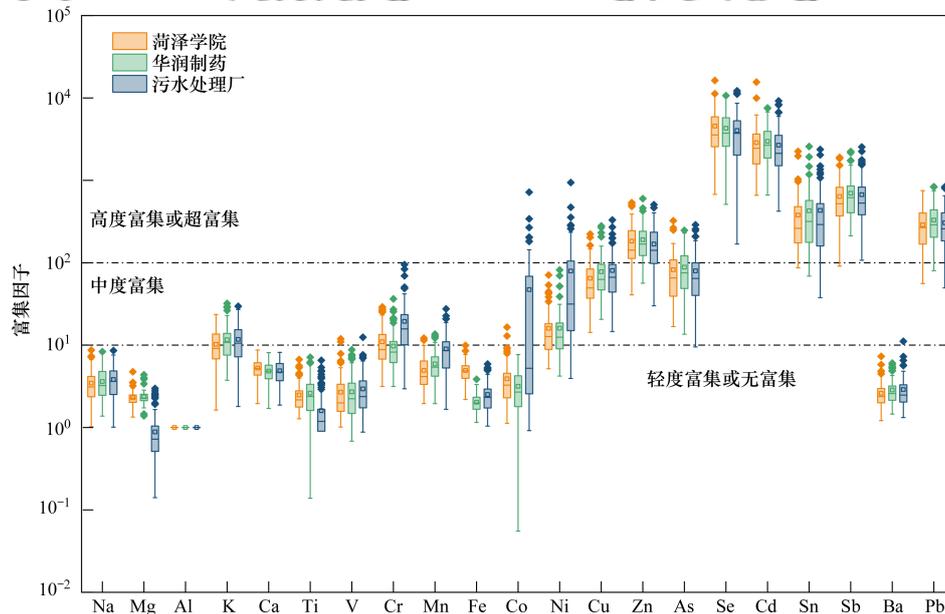


图5 菏泽学院、华润制药和污水处理厂金属元素富集因子

Fig. 5 Metal element enrichment factors of Heze College, Huarun Pharmacy, and Wastewater Treatment Plant

要来源是 As(儿童和成人的 HQ>0.1),其对儿童和成人存在潜在非致癌风险;其余元素的 HQ 均小于 0.1,且 3 个采样点对儿童和成人均存在综合潜在非致癌风险(HI>0.1).

表 6 给出了各金属元素具体致癌风险值,其中 Ni、Cd 和 Pb 在 3 个采样点对成人和儿童均不存在致

癌风险($CR < 1 \times 10^{-6}$). 对于成人,As 在 3 个采样点均存在潜在致癌风险($1 \times 10^{-6} < CR < 1 \times 10^{-4}$),且 As 的致癌风险强度表现为华润制药>菏泽学院>污水处理厂. Cr 在菏泽学院和华润制药存在潜在致癌风险($1 \times 10^{-6} < CR < 1 \times 10^{-4}$);在污水处理厂存在显著的致癌风险($CR > 1 \times 10^{-4}$). 此外,成人在菏泽学院、华润制药和污水处

理厂的综合潜在致癌风险值(CRT)分别为 8.52×10^{-5} 、 6.84×10^{-5} 和 1.26×10^{-4} ,表明菏泽学院和华润制药对成人存在综合潜在致癌风险、污水处理厂对成人存在显著的综合致癌风险. 对于儿童,As在菏泽学院和华润制药存在潜在致癌风险($1 \times 10^{-6} < CR < 1 \times 10^{-4}$)、在污水处理厂不存在致癌风险($CR < 1 \times 10^{-6}$); Cr在3个采样点均存在潜在致癌风险($1 \times 10^{-6} < CR < 1 \times 10^{-4}$),且Cr的致癌风险强度表现为污水处理厂>菏泽学院>华润制药. 此外,重金属元素在菏泽学院、华

润制药和污水处理厂的综合致癌风险值(CRT)分别为 2.13×10^{-5} 、 1.71×10^{-5} 和 3.14×10^{-5} ,即3个采样点对儿童均存在综合潜在致癌风险.

总体而言,需要重点关注Cr引起的健康风险并重视相关排放源的管控. 此外,通过不同人群对比研究发现,菏泽地区对于成人的致癌风险略高于儿童,这可能与人群室外活动的时间长短有关,通常成年人室外活动时间较长,PM_{2.5}的暴露量要高于儿童^[15].

表5 儿童和成人呼吸途径非致癌风险(HQ)和综合非致癌风险(HI)

Table 5 Respiratory route non-carcinogenic risk (HQ) and combined non-carcinogenic risk (HI) in children and adults

类型	采样点	V	Cr	Ni	Cu	Zn	As	Cd	Pb	HI
成人	菏泽学院	0.008	0.004	0.017	1.48×10^{-4}	1.47×10^{-4}	0.187	0.059	0.008	0.284
	华润制药	0.008	0.003	0.015	1.62×10^{-4}	1.38×10^{-4}	0.188	0.058	0.008	0.281
	污水处理厂	0.008	0.006	0.059	1.50×10^{-4}	1.11×10^{-4}	0.153	0.047	0.007	0.279
儿童	菏泽学院	0.008	0.004	0.017	1.48×10^{-4}	1.47×10^{-4}	0.187	0.059	0.008	0.284
	华润制药	0.008	0.003	0.015	1.62×10^{-4}	1.38×10^{-4}	0.188	0.058	0.008	0.281
	污水处理厂	0.008	0.006	0.059	1.50×10^{-4}	1.11×10^{-4}	0.153	0.047	0.007	0.279

表6 儿童和成人呼吸途径致癌风险(CR)和综合致癌风险(CRT)

Table 6 Respiratory route carcinogenic risk (CR) and combined carcinogenic risk (CRT) in children and adults

类型	采样点	Cr	Ni	As	Cd	Pb	CRT
成人	菏泽学院	8.02×10^{-5}	1.38×10^{-7}	4.14×10^{-6}	5.49×10^{-7}	1.15×10^{-7}	8.52×10^{-5}
	华润制药	6.35×10^{-5}	1.22×10^{-7}	4.16×10^{-6}	5.38×10^{-7}	1.20×10^{-7}	6.84×10^{-5}
	污水处理厂	1.21×10^{-4}	4.74×10^{-7}	3.38×10^{-6}	4.31×10^{-7}	1.01×10^{-7}	1.26×10^{-4}
儿童	菏泽学院	2.01×10^{-5}	3.46×10^{-8}	1.03×10^{-6}	1.37×10^{-7}	2.87×10^{-8}	2.13×10^{-5}
	华润制药	1.59×10^{-5}	3.05×10^{-8}	1.04×10^{-6}	1.35×10^{-7}	3.01×10^{-8}	1.71×10^{-5}
	污水处理厂	3.03×10^{-5}	1.19×10^{-7}	8.46×10^{-7}	1.08×10^{-7}	2.51×10^{-8}	3.14×10^{-5}

2.4 潜在生态风险评估

PM_{2.5}中含有大量有毒重金属,本研究对V、Ni、Cr、As、Cu、Zn、Pb和Cd共8种重金属元素的潜在生态风险进行了评估,结果见表7. 其中,Cd表现为极高的潜在生态风险,其指数值 E_r 在菏泽学院、华润制药和污水处理厂分别为6 990.7、6 562.4和5 164.8,对综合潜在生态风险的贡献分别为96.8%、96.6%和95.9%,与邯郸、东莞及厦门类似^[50],可能是Cd的毒性系数和生物可吸收性使其在生态系统中更容易形

成高富集状态^[32]. 此外,Pb在华润制药和污水处理厂表现为较高的潜在生态风险($80 < E_r < 160$); As和Pb(菏泽学院)表现为中等潜在生态风险($40 < E_r < 80$);其余元素表现为低潜在生态风险($E_r < 40$),造成的环境风险影响较小. 同时,3个采样点的综合潜在生态风险值(RI)分别为7 220.0、6 793.2和5 386.3,存在极高的综合潜在生态风险且强度表现为菏泽学院>华润制药>污水处理厂,单个元素的影响主要体现在Cd和Pb上,应当引起重视.

表7 重金属潜在生态风险指数(E_r)和综合潜在生态风险(RI)

Table 7 Heavy metal potential ecological risk index (E_r) and total potential ecological risk (RI)

项目	E_r								RI
	V	Ni	Cr	As	Cu	Zn	Pb	Cd	
菏泽学院	0.5	7.1	2.0	63.0	26.5	15.0	56.1	6 990.7	7 220.0
风险级别	低	低	低	中等	低	低	中等	极高	极高
华润制药	0.4	6.3	1.5	61.1	29.4	13.6	118.3	6 562.4	6 793.2
风险级别	低	低	低	中等	低	低	较高	极高	极高
污水处理厂	0.4	29.8	3.4	50.0	28.1	11.1	98.8	5 164.8	5 386.3
风险级别	低	低	低	中等	低	低	较高	极高	极高

3 结论

(1) 2017~2018年冬季, 菏泽地区3个采样点中 $\rho(\text{PM}_{2.5})$ 范围为26.7~284.1 $\mu\text{g}\cdot\text{m}^{-3}$, 大气 $\text{PM}_{2.5}$ 污染水平较高, 其中华润制药和污水处理厂大气中 $\text{PM}_{2.5}$ 含量整体略高于菏泽学院, 且逐日变化趋势呈现出高度的一致性。

(2) 菏泽学院、华润制药和污水处理厂 $\text{PM}_{2.5}$ 中21种金属元素累计浓度分别为7.30、6.01和5.47 $\mu\text{g}\cdot\text{m}^{-3}$, 占 $\text{PM}_{2.5}$ 总量的7.17%、5.77%和5.20%; 地壳元素占金属元素总量的64.05%、61.52%和63.23%。金属元素中K浓度最高, 分别占总量的31.03%、39.47%和38.43%; 微量元素中 $\rho(\text{Zn})$ 最高, 分别为89.70、84.21和67.68 $\text{ng}\cdot\text{m}^{-3}$ 。

(3) 3个采样点 $\text{PM}_{2.5}$ 中Zn、Pb、Sn、Sb、Cd和Se的富集因子值均高于100, 其中Cd和Se分别高于2000和4000, 为高度富集, 受人为影响严重; Mg、Ti、Ba、V、Na、Fe、Mn和Ca的富集因子值均小于10, 为轻度富集, 主要来源为自然源。

(4) 3个采样点非致癌风险的主要来源是As(HQ>0.1), 其对儿童和成人存在潜在非致癌风险; 且3个采样点对儿童和成人均存在综合潜在非致癌风险(HI>0.1)。菏泽学院、华润制药和污水处理厂的儿童致癌风险值分别为 2.13×10^{-5} 、 1.71×10^{-5} 和 3.14×10^{-5} , 存在综合潜在致癌风险; 成人致癌风险值分别为 8.52×10^{-5} 、 6.84×10^{-5} 和 1.26×10^{-4} , 其中菏泽学院和华润制药存在综合潜在致癌风险、污水处理厂存在显著的综合致癌风险。

(5) 3个采样点中, Cd表现为极高的潜在生态风险, Pb在华润制药和污水处理厂中表现为较高的潜在生态风险, As和Pb(菏泽学院)表现为中等潜在风险, 其余元素表现为低潜在风险。同时, 3个采样点均存在极高的综合潜在生态风险, 强度表现为: 菏泽学院>华润制药>污水处理厂。

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CONTENTS

Research on the Evaluation Method and Application of Provincial Differentiated Carbon Peaking in China	LIU Run-pu, PENG Shuan, CHEN Yu-shuo, <i>et al.</i> (1233)
Evolution and Influencing Factors of Spatial Correlation Network of Construction Carbon Emission in China from the Perspective of Whole Life Cycle	REN Xiao-song, LI Zhao-ru (1243)
Efficiency Characteristics and Evolution Patterns of Urban Carbon Metabolism of Production-Living-Ecological Space in Beijing-Tianjin-Hebei Region	TIAN Chao, CHENG Lin-lin, SHAO Ying-chao (1254)
Assessment of CO ₂ Co-benefits of Air Pollution Control Policies in Taiyuan's 14th Five-Year Plan	XIAO Ting-yu, SHU Yun, LI hui, <i>et al.</i> (1265)
Coordinated Control of Carbon Emission Reduction and Air Quality Improvement in the Industrial Sector in Hunan Province	LI Nan, LIU Wan-wan, ZHU Shu-han, <i>et al.</i> (1274)
Coal Control and Carbon Reduction Path in Henan Province's Power Industry Under the Carbon Peak and Neutralization Target: A Medium- and Long-term Study	ZHANG Jing, YANG Meng, ZHANG Wei, <i>et al.</i> (1285)
Environmental Benefits of Pollution and Carbon Reduction by Bus Fleet Electrification in Zhengzhou	ZOU Chao, WANG Ya-nan, WU Lin, <i>et al.</i> (1293)
Multi-scale Driving Mechanism of Urbanization on PM _{2.5} Concentration in Urban Agglomeration in the Middle Reaches of the Yangtze River	ZHANG Zheng, ZHOU Ting-gang, ZHOU Zhi-heng, <i>et al.</i> (1304)
Analysis of Spatio-temporal Distribution Characteristics and Influencing Factors of PM _{2.5} Concentration in Urban Agglomerations on the Northern Slope of Tianshan Mountains	WANG Xiang-nan, ZHANG Zhe, LIU Fang-qing (1315)
Spatial Variability and Source Apportionment of PM _{2.5} Carbon Components in Tianjin	WU Fu-liang, WU Jian-hui, DAI Qi-li, <i>et al.</i> (1328)
Characteristics and Source Analysis of Carbonaceous Aerosols in PM _{2.5} in Huaxi District, Guiyang	GUI Jia-qun, YANG Yuan, WANG Xian-qin, <i>et al.</i> (1337)
Pollution Characteristics, Source Apportionment, and Meteorological Response of Water-soluble Ions in PM _{2.5} in Xinxiang, North China	LIU Huan-jia, LI Lan-qing, LI Huan-li, <i>et al.</i> (1349)
Characterization of Metal Elements in Atmospheric PM _{2.5} and Health Risk Assessment in Heze in Winter from 2017 to 2018	DU Hong-xuan, REN Li-hong, ZHAO Ming-sheng, <i>et al.</i> (1361)
Analysis of the Jumping Characteristics and Influencing Factors of Ozone Pollution in Beijing	PAN Jin-xin, AN Xin-xin, LIU Bao-xian, <i>et al.</i> (1371)
Analysis of Photochemical Characteristics and Sensitivity of Atmospheric Ozone in Nanjing in Summer	LUO Li-tong, ZHANG Yan-lin, LIN Yu-qi, <i>et al.</i> (1382)
Ozone Pollution in Suzhou During Early Summertime: Formation Mechanism and Interannual Variation	WU Ye-zheng, ZHANG Xin, GU Jun, <i>et al.</i> (1392)
Distribution Characteristics, Ecological Risk Assessment, and Source Tracing of Heavy Metals in the Sediments of Typical Lakes in the Middle Reaches of the Yangtze River	LU Hong-bin, LU Shao-yong, LI Xiang, <i>et al.</i> (1402)
Spatiotemporal Distribution and Source Analysis of Heavy Metals in Surface Sediments in Lake Ulansuhai Based on PCA-APCS-MLR Model	CUI Zhi-mou, SHI Xiao-hong, ZHAO Sheng-nan, <i>et al.</i> (1415)
Spatial and Temporal Distribution and Risk Assessment of Heavy Metals in Surface Water of Changshou Lake Reservoir, Chongqing	ZHANG Rui-xi, LIU Ya-jun, LUO Yong-nan, <i>et al.</i> (1428)
Pollution Characteristics and Ecological Risk Assessment of Microplastics in the Yangtze River Basin	LI Si-qiong, WANG Hua, CHU Lin-you, <i>et al.</i> (1439)
Assessment of Microplastic Pollution and Estimation of Annual Emission Volume in the Dongshan Canal of Yichang City	DING Shuang, LI Wei-ming, ZHANG Xu-tong, <i>et al.</i> (1448)
Occurrence Characteristic and Risk Assessment of Microplastics in Sishui River (Xingyang Section)	ZHAO Chang-min, HE Bing, LI He-tong, <i>et al.</i> (1457)
Contamination Characteristics and Ecological Risk Assessment of Pharmaceuticals and Personal Care Products in Drains Flowing into the Yellow River of Ningxia	GAO Li, LI Ling-yun, ZHENG Lan-xiang, <i>et al.</i> (1468)
Characteristics and Risk Assessment of Antibiotic Contamination in Oujiang River Basin in Southern Zhejiang Province	ZHONG Yi-xin, LI Li-xiang, WU Xin, <i>et al.</i> (1480)
Spatial-temporal Distribution and Source Analysis of Polycyclic Aromatic Hydrocarbons in the Sediments of Poyang Lake	MA Yan, SUN Chen, BI Jia-le, <i>et al.</i> (1492)
Change and Prediction of Water Purification Function in the South Bank of Hangzhou Bay in the Past 20 Years	WANG Shan-shan, CAO Gong-ping, XU Ming-wei, <i>et al.</i> (1502)
Effects of Land Use Types on Water Quality at Different Buffer Scales: Tianjin Section of the Haihe River Basin as an Example	DAI Meng-jun, ZHANG Bing, DU Qian-qian, <i>et al.</i> (1512)
Hydrochemical Characteristics and Control Factors of Shallow Groundwater in Anqing Section of the Yangtze River Basin	LIU Hai, SONG Yang, LI Ying-chun, <i>et al.</i> (1525)
Dynamic Changes of Dissolved Organic Matter Derived from Algal Decomposition and the Environmental Effects in Eutrophic Lakes	ZHANG Jin, CHEN Ming-ying, HAO Zhi-neng, <i>et al.</i> (1539)
Degradation of Carbamazepine in Water by UV-activated Sulfite Process	LIN Tao, YUAN Yu-jie (1553)
Detection, Generation, and Control of Disinfection By-products of Reclaimed Water	LIAO Yu-feng, WANG Zheng, PAN Yang, <i>et al.</i> (1561)
Absorption of Ammonium by Three Substrates Materials in Constructed Wetland System	HE Qiang, CHEN Bo-wen, HAN Ling, LIU Ming, <i>et al.</i> (1577)
Ecological Environment Assessment and Driving Mechanism Analysis of Nagqu and Amdo Sections of Qinghai-Xizang Highway Based on Improved Remote Sensing Ecological Index	FU Kai-xiang, JIA Guo-dong, YU Xin-xiao, <i>et al.</i> (1586)
Spatial-temporal Evolution and Driving Factors Analysis of Ecological Environment Quality in Daihai Basin based on AWRSEI	ZHAO Jia-li, LI Xing, SUN Bing (1598)
Quantitative Assessment of the Impact of Climate Change on the Growing Season of Vegetation Gross Primary Productivity in the Middle and Lower Reaches of the Yangtze River	XU Yong, PAN Yu-chun, ZOU Bin, <i>et al.</i> (1615)
Effect of Vegetation Restoration on Soil Organic Carbon Storage in Coal Mining Areas Based on Meta-analysis	LI Jian-ming, KANG Yu-xin, JIANG Fu-zhen, <i>et al.</i> (1629)
Effects of Continuous Annual Crop Rotation and Fallow on Soil Aggregate Stability and Organic Carbon	LU Ze-rang, LI Yong-mei, YANG Chun-huai, <i>et al.</i> (1644)
Effects of Four Amendments on Fertility and Labile Organic Carbon Fractions of Acid Purple Soil	DING Xin-ru, YAN Ning-zhen, WANG Zi-fang, <i>et al.</i> (1655)
Changes in Soil Nitrogen Components and Their Relationship with Environmental Factors with Different Tea Plantation Ages	SHAO Qi, WU Tao, XIE Xue-feng, <i>et al.</i> (1665)
Nutrients and Ecological Stoichiometry Characteristics of Typical Wetland Soils in the Lower Yellow River	WANG Chuan-ying, WANG Kai-yue, WANG Hao-ran, <i>et al.</i> (1674)
Effect of Film Mulching Age and Organic Fertilizer Application on the Distribution Characteristics of Microplastics in the Soil of a Peanut Field	SONG Ning-ning, LI Meng-jia, WANG Xue-xia, <i>et al.</i> (1684)
Effects of Straw Returning and Biochar Addition on Greenhouse Gas Emissions from High Nitrate Nitrogen Soil After Flooding in Rice-vegetable Rotation System in Tropical China	HU Tian-yi, CHE Jia-yue, HU Yu-jie, <i>et al.</i> (1692)
Tillage Depth Regulation and the Effect of Straw Return on Soil Respiration in Farmland	CHEN Xi, ZHANG Yan-jun, ZOU Jun-liang, <i>et al.</i> (1702)
Distribution Prediction of Soil Heavy Metals Based on Remote Sensing Temporal-Spatial-Spectral Features and Random Forest Model	WANG Ze-qiang, ZHANG Dong-you, XU Xi-bo, <i>et al.</i> (1713)
Characteristics and Identification Priority Source of Heavy Metals Pollution in Farmland Soils in the Yellow River Basin	LI Jun, LI Xu, LI Kai-ming, <i>et al.</i> (1724)
Characteristics of Cd Flux in Topsoil Around Typical Mining Area in Hezhou, Guangxi	YANG Ye-yu, LI Cheng, YANG Zhong-fang, <i>et al.</i> (1739)
Potential Ecological Risk Assessment of Soil Heavy Metals in Fengdong New District Based on Information Diffusion Model	YANG Nan-nan, HAN Ling, LIU Ming (1749)
Traceability Analysis and Environmental Quality Assessment of Soil Heavy Metal Pollution in West Hunan Province	XIAO Kai-qi, XU Hong-gen, GAN Jie, <i>et al.</i> (1760)
Distribution Characteristics, Source Analysis and Ecological Risk Assessment of Heavy Metals in the Typical Industry Reclaimed Soil	SHEN Cheng, WANG Wen-juan, SHA Chen-yan, <i>et al.</i> (1769)
Provincial-scale Soil As Migration and Transformation and Rice Safe Planting Zoning: A Case Study of Guizhou Province	DONG Xin-yue, WU Yong, ZHOU Zi-han, <i>et al.</i> (1781)
Effect of Silica Fertilizer (Husk Ash) to Improve Soil Quality and Reduce Cd and As Accumulation in Rice	YI Xuan-tao, OUYANG Kun, GU Jiao-feng, <i>et al.</i> (1793)
Effect of EDDS Application on Soil Cu/Cd Availability and Uptake/transport by Castor	LIU Wen-ying, WU Gang, HU Hong-qing (1803)
Blocking Effects of Foliar Conditioners on Cadmium, Arsenic, and Lead Accumulation in Wheat Grain in Compound-contaminated Farmland	XIAO Bing, WANG Qiu-shi, GAO Pei-pei, <i>et al.</i> (1812)
Soil Microbial Community Structure and Functional Diversity Character of Abandoned Farmland in Minqin Oasis	LI Chang-le, ZHANG Fu, WANG Li-de, <i>et al.</i> (1821)
Microbial Mechanisms of Removal of Phthalic Acid Esters in Purple Soils Revealed Using Metagenomic Analysis	LI Yu-tong, YU Hai, LIU Kun, <i>et al.</i> (1830)
Air Microbial Contamination and Risk of Respiratory Exposure of Workers in Chicken Farms	BAI Yu-qiao, SUN Xing-bin, QIU Tian-lei, <i>et al.</i> (1840)
Occurrence Characteristics of Microplastics in Multi-environmental Media and <i>Bellamyia aeruginosa</i> of Manao River	GAO Ya-kun, LI Wei-ming, ZHANG Xu-tong, <i>et al.</i> (1849)
Biological Effect of Microplastics with Different Functional Groups on the Bacterial Communities and Metabolic Functions of Zebrafish (<i>Danio rerio</i>) Embryos	YAN Zhen-hua, ZHANG Yan, BAO Xu-hui, <i>et al.</i> (1859)