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目次

北京城区大气金属元素干湿沉降特征	张国忠, 潘月鹏, 田世丽, 王永宏, 熊秋林, 李广, 顾梦娜, 吕雪梅, 倪雪, 何月欣, 黄威, 刘博文, 王跃思	(2493)
北京城郊 PM _{2.5} 中金属元素的污染特征及潜在生态风险评价	徐静, 李杏茹, 张兰, 陈曦, 杨阳, 刘永桥, 赵清	(2501)
上海城区二次气溶胶的形成: 光化学氧化与液相反应对二次气溶胶形成的影响	高杰, 乔利平, 楼晟荣, 严茹莎, 周敏, 刘瑜存, 冯加良, 黄丹丹	(2510)
天津市 2017 年重污染过程二次无机化学污染特征分析	徐虹, 肖致美, 陈魁, 李立伟, 杨宁, 高璟赞, 李源, 孔君, 毕温凯, 邓小文	(2519)
典型非金属矿物制造工艺过程源成分谱特征	赵雪艳, 于高峰, 王信梧, 张向炎, 殷宝辉, 刘盈盈, 王歆华, 杨文, 赵若杰	(2526)
垃圾焚烧厂排放颗粒物组分粒径分布特征	余卓君, 吴建会, 张裕芬, 张进生, 冯银厂, 李蒲	(2533)
天津市春季样方法道路扬尘碳组分特征及来源分析	马妍, 姬亚芹, 国纪良, 赵静琦, 李越洋, 王士宝, 张蕾	(2540)
霾天气下城市气溶胶吸湿性的观测	杨素英, 田芷洁, 张铁凝, 于兴娜, 李艳伟, 安俊琳, 赵秀勇, 李岩, 王梓航, 吴尚	(2546)
宁东能源化工基地大气 PM _{2.5} 中硝基多环芳烃污染特征及呼吸暴露风险	刘攀亮, 剧媛丽, 毛蒲萱, 黄韬, 高宏, 马建民	(2556)
2014 ~ 2016 年间郑州市控制 PM ₁₀ 和 PM _{2.5} 污染的健康效益评估	韩士杰, 王佳, 燕启社, 杨留明, 赵庆炎, 王申博, 李晨, 张轶舜, 张瑞芹	(2565)
1990 ~ 2017 年中国地区气溶胶光学厚度的时空分布及其主要影响类型	刘莹, 林爱文, 覃文敏, 何利杰, 李霄	(2572)
北京春季一次霾-沙天气污染特性与成因分析	王耀庭, 李青春, 郑祚芳, 窦有俊	(2582)
江苏省内河船舶大气污染物排放清单及特征	徐文文, 殷承启, 许雪记, 张玮	(2595)
亚热带稻区大气 NO ₂ 、HNO ₃ 及硝态氮污染特征及干湿沉降	欧阳秀琴, 王波, 沈健林, 朱潇, 王杰飞, 李勇, 吴金水	(2607)
长江源区降水氢氧稳定同位素特征及水汽来源	汪少勇, 王巧丽, 吴锦奎, 何晓波, 丁永建, 王利辉, 胡召富	(2615)
基于高山湖泊沉积记录的贵州北部大气重金属污染历史重建	梁梦瑶, 刘恩峰, 张恩楼, 纪明, 李小林	(2624)
青岛 4 个海水浴场微塑料的分布特征	罗雅丹, 林千惠, 贾芳丽, 徐功娣, 李锋民	(2631)
鄱阳湖湿地候鸟栖息地微塑料污染特征	刘淑丽, 简敏菲, 周隆胤, 李文华, 吴希恩, 饶丹	(2639)
三峡库区消落带土壤溶解性有机质溯源: 基于氮/碳比值的线性双端元源负荷分析	江韬, Joeri Kaal, 梁俭, 刘江, 张耀玲, 王定勇, 魏世强, 赵铮	(2647)
水生生物光合作用对雪玉洞岩溶水体中 CDOM 的影响	范佳鑫, 蒋勇军, 贺秋芳, 王家楠, 何瑞亮, 张彩云, 马丽娜, 汪啟容	(2657)
水化学和环境同位素对济南东源饮用水源地地下水演化过程的指示	张雅, 苏春利, 马燕华, 刘伟江	(2667)
崇左响水地区地下水水质分析及健康风险评价	周巾枚, 蒋忠诚, 徐光黎, 覃小群, 黄奇波, 张连凯	(2675)
云南荞麦地流域地下水水化学特征及物质来源分析	张勇, 郭纯青, 朱彦光, 于爽	(2686)
鄱阳湖流域多尺度 C、N 输送通量及其水质参数变化特征	陆瑶, 高扬, 贾珺杰, 宋贤威, 陈世博, 马明真, 郝卓	(2696)
合流制面源污染传输过程与污染源解析	房金秀, 谢文霞, 朱玉玺, 沈雷, 马玉坤, 李佳, 姜智绘, 李叙勇, 赵洪涛	(2705)
强化浅基质层干植草沟对道路径流的脱氮效果	段进凯, 李田, 张佳炜	(2715)
水源水库暴雨径流过程水体锰的迁移及其影响	邓立凡, 黄廷林, 李楠, 李凯, 吕晓龙, 毛雪静	(2722)
巢湖十五里河沉积物磷吸收潜力及对外源碳的响应	李如忠, 鲍琴, 张瑞钢, 陈慧	(2730)
金盆水库沉积物磷的来源及分布特征	毛雪静, 黄廷林, 李楠, 徐金兰, 苏露, 吕晓龙, 邓立凡	(2738)
水库水体热分层的水质及细菌群落分布特征	王禹冰, 王晓燕, 庞树江, 杨晓明, 刘洋	(2745)
不同水温分层水库沉积物间隙水营养盐垂向分布与细菌群落结构的关系	王慎, 张思思, 许尤, 官卓宇, 杨正健, 刘德富, 马骏	(2753)
表面流人工湿地长期运行后的底泥营养盐累积特征与释放规律	朱伊梦, 姜翠玲, 朱立琴, 杜观超, 高旭, 陈红卫, 李峰东, 张海阔, 张雪, 秦文凯, 李一平	(2764)
针铁矿改性生物炭对砷吸附性能	朱司航, 赵晶晶, 尹英杰, 商建英, 陈冲, 瞿婷	(2773)
改性稻壳生物炭对水溶液中甲基橙的吸附效果与机制	史月月, 单锐, 袁浩然	(2783)
厌氧膜生物反应器处理含盐废水运行效能及膜污染特性	闫欢沙, 许振钰, 金春姬, 邵梦雨, 郭亮, 赵阳国	(2793)
污水处理厂出水有机磷污染特征及强化去除	王小东, 王子文, 陈明飞, 王燕, 王硕, 李激	(2800)
碱度对沸石序批式反应器亚硝化的影响	王瑞鑫, 陈婧, 汪晓军, 胡浩林, Karasuta Chayangkun	(2807)
活性污泥物理结构对呼吸过程的影响	郭耀, 李志华, 杨成建, 王慧娟, 李亚明, 姜阳	(2813)
好氧颗粒污泥处理畜禽养殖沼液污染物的特性	廖杰, 叶嘉琦, 曾志超, 刘琳, 徐开钦, 刘超翔	(2821)
快速城市化区河流温室内空气排放的时空特征及驱动因素	刘婷婷, 王晓峰, 袁兴中, 龚小杰, 侯春丽	(2827)
生物炭对潜流人工湿地污染物去除及 NO ₃ 排放影响	邓朝仁, 梁银坤, 黄磊, 方丹丹, 陈玉成, 杜刚	(2840)
曾用抗生素磺胺二甲嘧啶对稻田 N ₂ O 排放的影响及其微生物机制	吴杰, 李志琳, 徐佳迎, 王珏, 蒋静艳	(2847)
不同施氮水平对紫花苜蓿草地土壤呼吸和土壤生化性质的影响	胡伟, 张亚红, 李鹏, 张鹏, 李满友, 尤璟涛, 田水泉	(2858)
典型城市化区域土壤重金属污染的空间特征与风险评价	何博, 赵慧, 王铁宇, 孟晶, 肖荣波, 刘胜然, 周云桥, 史斌	(2869)
西南高锡地质背景区农田土壤与农作物的重金属富集特征	刘意章, 肖唐付, 熊燕, 宁增平, 双燕, 李航, 马良, 陈海燕	(2877)
基于 GIS 和受体模型的枸杞地土壤重金属空间分布特征及来源解析	白一茹, 张兴, 赵云鹏, 王幼奇, 钟艳霞	(2885)
喀斯特地区小尺度农业土壤砷的空间分布及污染评价	汪花, 刘秀明, 刘方, 唐启琳, 王世杰	(2895)
黄土高原不同植被带人工刺槐林土壤团聚体稳定性及其化学计量特征	瞿晴, 徐红伟, 吴旋, 孟敏, 王国梁, 薛蓑	(2904)
秸秆还田配施氮肥对喀斯特农田微生物群落及有机碳矿化的影响	徐学池, 苏以荣, 王桂红, 刘坤平, 胡亚军, 陈香碧, 郑小东, 何寻阳	(2912)
生物炭施用及老化对紫色土中抗生素吸附特征的影响	阴文敏, 关卓, 刘琛, 何杨, 杨飞, 唐翔宇	(2920)
稳定性同位素 DNA-SIP 示踪中性紫色土的氨氧化过程	刘天琳, 王智慧, 闫小娟, 赵永鹏, 贾仲君, 蒋先军	(2930)
纳米银和银离子对土壤中硝化微生物及其氨氧化速率的影响	伍玲丽, 张旭, 舒慧慧, 张丽, 司友斌	(2939)
农田沟道土壤中锰氨氧化 (Mn-ANAMMOX) 过程的探究	陈湜, 李正魁, 覃云斌, 丁帮环, 陈志浩	(2948)
污泥四环素含量对蚯蚓堆肥中氨氧化菌群的影响	吴颖, 黄魁, 夏慧, 陈景阳	(2954)
《环境科学》征订启事 (2929)	《环境科学》征稿简则 (2947)	信息 (2646, 2656, 2857)

天津市 2017 年重污染过程二次无机化学污染特征分析

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摘要: 基于 2017 年天津市超级观测站数据, 筛选出 7 次典型重污染过程, 从污染物浓度、二次转化方面分析重污染过程二次无机化学污染特征。结果表明, 重污染期间 NO_3^- 和 SO_4^{2-} 浓度较清洁天气增长幅度最高, 显著高于 $\text{PM}_{2.5}$ 的增长程度, 说明二次无机转化是导致重污染期间 $\text{PM}_{2.5}$ 污染加重的重要原因; 下半年 $\text{PM}_{2.5}$ 和 SO_2 污染程度较上半年减轻, 与秋冬季采取燃煤治理等活动有关; 重污染期间 NO_2/SO_2 比值为 1.5 ~ 19.6, 其中下半年 NO_2/SO_2 比值显著高于上半年, 说明在各项污染源管控下移动源的影响比例相对增加; 大部分重污染期间 NO_3^- 浓度大于 SO_4^{2-} 浓度, SOR 值高于 NOR 值, 说明重污染期间硫酸盐和硝酸盐转化均较重要; 在 SO_2 浓度显著降低的情况下, 重污染期间 SO_4^{2-} 浓度并未明显降低, 说明除二次无机转化外, 硫酸盐生成还受其他因素影响。

关键词: 天津; 重污染; 2017 年; 二次转化; 污染特征

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Secondary Inorganic Pollution Characteristics During Heavy Pollution Episodes of 2017 in Tianjin

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Abstract: Based on monitoring data collected at the supersite of Tianjin in 2017, seven typical heavy pollution episodes were investigated. The concentrations of air pollutants and secondary inorganic transformation products were analyzed to study the secondary inorganic pollution characteristics during the heavy pollution episodes. Compared to clean weather, concentrations of NO_3^- and SO_4^{2-} during the heavy pollution episodes increased at rapid growth rates. These rates were obviously higher than the rate for $\text{PM}_{2.5}$ increases, which indicates that the secondary inorganic reactions had an important influence on $\text{PM}_{2.5}$ pollution during the episodes. The concentrations of $\text{PM}_{2.5}$ and SO_2 during the episodes in the latter half of the year were lower than those in the first half of the year probably because a substantial amount of coal use had been controlled. During the heavy pollution episodes, the NO_2/SO_2 values were 1.5 to 19.6, with higher values in the latter half of the year than the first half of the year suggestive of a greater influence from mobile sources. During most episodes, NO_3^- concentrations were higher than SO_4^{2-} concentrations, and SOR values were higher than NOR values, which shows that the secondary transformation of nitrate and sulfate both played important roles during the heavy pollution episodes. When SO_2 concentrations decreased significantly, SO_4^{2-} concentrations did not decrease obviously, thus indicating that besides the secondary inorganic reactions, other factors also had a large impact on the generation of sulfate.

Key words: Tianjin; heavy pollution episode; 2017; secondary inorganic; pollution characteristics

近年来重污染天气在我国一些地区和城市频频发生^[1~3], 污染程度高, 对人体健康^[4~6]、大气能见度^[7~9]、农作物^[10, 11]等均产生较大的影响。目前国内外不少学者已开展有关大气重污染方面的研究, 从气象条件、污染物浓度、化学机制等方面研究大气重污染的污染特征及其成因^[12~16]。有研究发现大气环流形势稳定、持续性的逆温、风速较小、湿度较大等因素是导致重污染形成的重要外部原因^[17~19], 污染物排放量是重污染形成的重要内部原因^[20, 21]。重污染期间静稳、高湿的环境下容易引发污染物的非均相化学反应, 进而加剧大气污染的程度^[22, 23]。

已有研究得出重污染期间二次无机化学反应对重污染的影响较大, 其中, 西南地区 2015 年重污染期间二次无机组分 (SO_4^{2-} 、 NO_3^- 和 NH_4^+) 浓度在

$\text{PM}_{2.5}$ 中的占比高达 42.4%^[24]; 北京地区 2013 年冬季二次无机离子在 $\text{PM}_{2.5}$ 中占比为 40.29%^[25], 2015 年北京红色预警期间观测到明显的二次无机盐转化过程, 二次无机离子组分约占 $\text{PM}_{2.5}$ 浓度的 68%^[26]; 石家庄市 2013 年冬季从优良天到重污染天, 二次无机离子的比例由 44.9% 上升至 77.6%^[27]; 2016 年冬季北京、石家庄和唐山市二次无机离子在 $\text{PM}_{2.5}$ 的占比分别为 45%、39% 和 42%^[28]。

本研究从二次无机化学反应角度出发, 选取天

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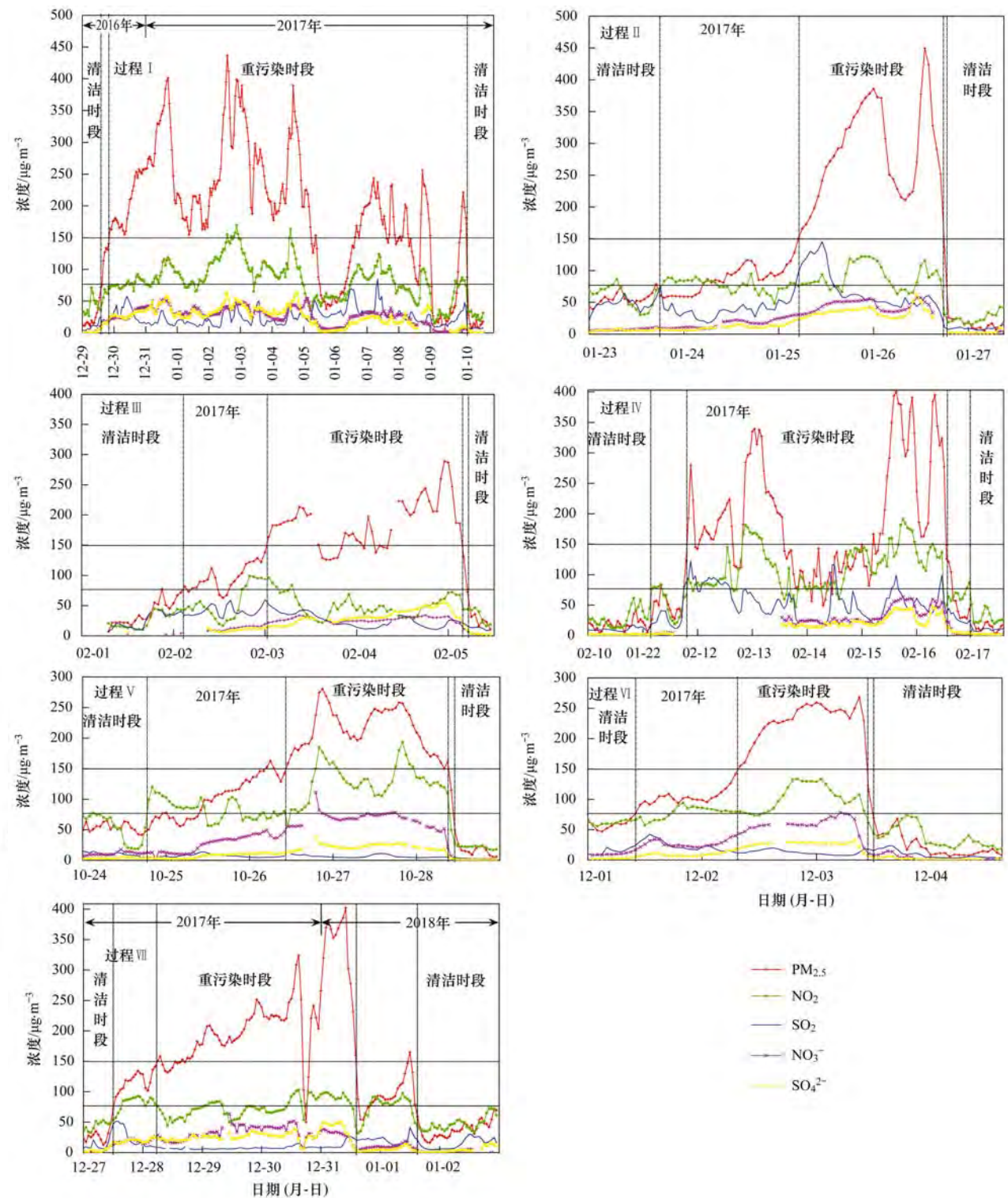


图2 天津市 2017 年典型重污染过程主要污染物及二次化学组分浓度时间变化序列

Fig. 2 Time series of concentrations of $\text{PM}_{2.5}$, NO_2 , SO_2 , NO_3^- , and SO_4^{2-} during seven heavy pollution episodes in 2017

不同重污染过程污染物浓度的变化特征存在差异,如图3所示,上半年的4次重污染过程即2017年1月1~9日(过程Ⅰ)、1月23~27日(过程Ⅱ)、2月1~5日(过程Ⅲ)和2月10~17日(过程Ⅳ)期间, $\text{PM}_{2.5}$ 浓度在重污染时段均值分别是清洁天气时段的5.4倍、4.4倍、3.0倍和6.3倍,发生在2017年下半年的3次重污染过程,即10月24~

28日(过程Ⅴ)、12月1~4日(过程Ⅵ)、12月27~31日(过程Ⅶ)期间, $\text{PM}_{2.5}$ 浓度在重污染时段均值分别是清洁天气时段的3.5倍、4.7倍和3.6倍.此外,上半年4次重污染过程期间 $\text{PM}_{2.5}$ 浓度均值为186~283 $\mu\text{g}\cdot\text{m}^{-3}$,整体高于下半年3次重污染期间的 $\text{PM}_{2.5}$ 浓度均值(179~189 $\mu\text{g}\cdot\text{m}^{-3}$).可见,下半年重污染期间 $\text{PM}_{2.5}$ 浓度较清洁天气的增长程

表 1 典型重污染过程期间重污染时段与清洁天气时段污染物浓度对比¹⁾

Table 1 Average concentrations of various pollutants during the seven pollution episodes and comparisons to the concentrations during the clean weather period

日期(月-日)	时段	PM _{2.5}	NO ₂	SO ₂	NO ₂ /SO ₂	NO ₃ ⁻	SO ₄ ²⁻	NO ₃ ⁻ /SO ₄ ²⁻	NOR	SOR
01-01 ~ 01-09(过程 I)	重污染	196	84	26	4	27	27.3	1.2	0.18	0.38
	清洁	36	40	11	4.3	4.1	3.9	0.9	0.06	0.15
	倍数	5.4	2.1	2.4	0.9	6.6	7.0	1.3	3.2	2.5
01-23 ~ 01-27(过程 II)	重污染	283	89	68	1.5	44.3	34.8	1.3	0.25	0.24
	清洁	64	61	35	2.1	10.8	7.7	1.2	0.10	0.13
	倍数	4.4	1.5	1.9	0.7	4.1	4.5	1.1	2.4	1.9
02-01 ~ 02-05(过程 III)	重污染	186	50	25	2.4	26.9	32.4	0.9	0.29	0.48
	清洁	63	40	31	1.4	8.8	8.1	1.1	0.13	0.15
	倍数	3.0	1.3	0.8	1.7	3.1	4.0	0.8	2.2	3.3
02-10 ~ 02-17(过程 IV)	重污染	194	113	56	2.5	33.3	24.3	1.4	0.18	0.27
	清洁	31	40	17	2.6	2.5	2.8	0.7	0.04	0.11
	倍数	6.3	2.8	3.3	1.0	13.3	8.7	2.0	4.7	2.6
10-24 ~ 10-28(过程 V)	重污染	188	113	6	19.6	57.4	18.3	3.2	0.28	0.65
	清洁	53	59	8	8.5	9.9	3.2	2.7	0.11	0.21
	倍数	3.5	1.9	0.8	2.3	5.8	5.7	1.2	2.5	3.1
12-01 ~ 12-04(过程 VI)	重污染	179	94	16	7.1	44.2	19	2.5	0.25	0.44
	清洁	38	46	12	4.9	6.8	2.7	2	0.08	0.13
	倍数	4.7	2.0	1.3	1.4	6.5	7.0	1.3	3.2	3.5
12-27 ~ 12-31(过程 VII)	重污染	189	75	13	8.2	27.8	24.5	1.3	0.21	0.58
	清洁	53	55	15	3.7	5	5	1	0.04	0.17
	倍数	3.6	1.4	0.9	2.2	5.6	4.9	1.3	5.8	3.4

1) “重污染”代表重污染时段的均值;“清洁”代表清洁天气时段的均值;“倍数”代表重污染平均浓度与清洁时段平均浓度的比值;PM_{2.5}、SO₂、NO₂、NO₃⁻和SO₄²⁻单位为μg·m⁻³,其余参数为比值

度整体低于上半年,且污染程度较上半年减轻,可能与下半年秋冬季期间天津市采取各项减排措施导致颗粒物污染源排放减少有关。

上半年4次重污染过程中,包括过程I、过程II和过程IV在内的3次污染过程期间SO₂浓度在重污染时段的均值均显著高于清洁天气时段,是清洁天气时段的1.9倍~3.3倍。而在下半年3次重污染过程中,有两次重污染过程SO₂浓度较清洁天气时段低,过程V和过程VII期间SO₂浓度均值分别是清洁天气时段的0.8倍和0.9倍。此外,上半年4次重污染过程期间SO₂浓度均值为25~68 μg·m⁻³,而下半年3次重污染期间的PM_{2.5}浓度均值仅6~16 μg·m⁻³。可见下半年重污染期间SO₂浓度较上半年显著降低,且下半年SO₂浓度在重污染期间的均值通常还低于清洁天气时段。这与下半年秋冬季期间天津市采取的燃煤治理等活动导致SO₂排放大幅减少有关^[32]。值得注意的是,下半年天津市大气SO₂浓度已达到较低水平,即使重污染期间SO₂浓度仍未呈现升高趋势,反而可能是因为SO₂作为前体物参与了二次无机化学反应^[33, 34],导致重污染期间SO₂的浓度较清洁天气时段还低,进一步证明重污染过程期间二次无机化学反应加快。

NO₂/SO₂比值通常用于表征移动源和燃煤源对

污染贡献的变化^[35, 36]。其中,NO₂主要来自于燃煤和机动车尾气,SO₂主要来源于燃煤。如图4所示,全年7次重污染过程期间NO₂/SO₂比值为1.5~19.6,其中上半年过程I、过程II、过程III和过程IV期间NO₂/SO₂比值分别为4、1.5、2.4和2.5,下半年过程V、过程VI和过程VII期间NO₂/SO₂比值分别为19.6、7.1和8.2,下半年NO₂/SO₂比值显著高于上半年,进一步说明下半年天津市燃煤源的影响显著降低。与清洁天气时段相比,下半年重污染时段NO₂/SO₂比值是清洁天气时段的1.4~2.3倍,显著高于上半年的0.7~1.7倍,说明在采取各项污染源管控措施后,下半年移动源等排放的NO₂对重污染的影响比例相对增加。

2.3 二次污染物转化特征

NO₃⁻和SO₄²⁻是颗粒物中重要的化学组分,已有研究表明,二者除来自一次污染源排放外,主要由气态前体物NO₂和SO₂经非均相化学反应转化生成^[37, 38]。

2017年天津市7次重污染过程期间,NO₃⁻质量浓度分别是27.0、44.3、26.9、33.3、57.4、44.2和27.8 μg·m⁻³,分别占PM_{2.5}质量的13.8%、15.7%、14.5%、17.2%、30.5%、24.7%和14.7%,7次重污染过程期间NO₃⁻质量浓度是清洁天气时段的6.6、4.1、3.1、13.3、5.8、6.5和5.6

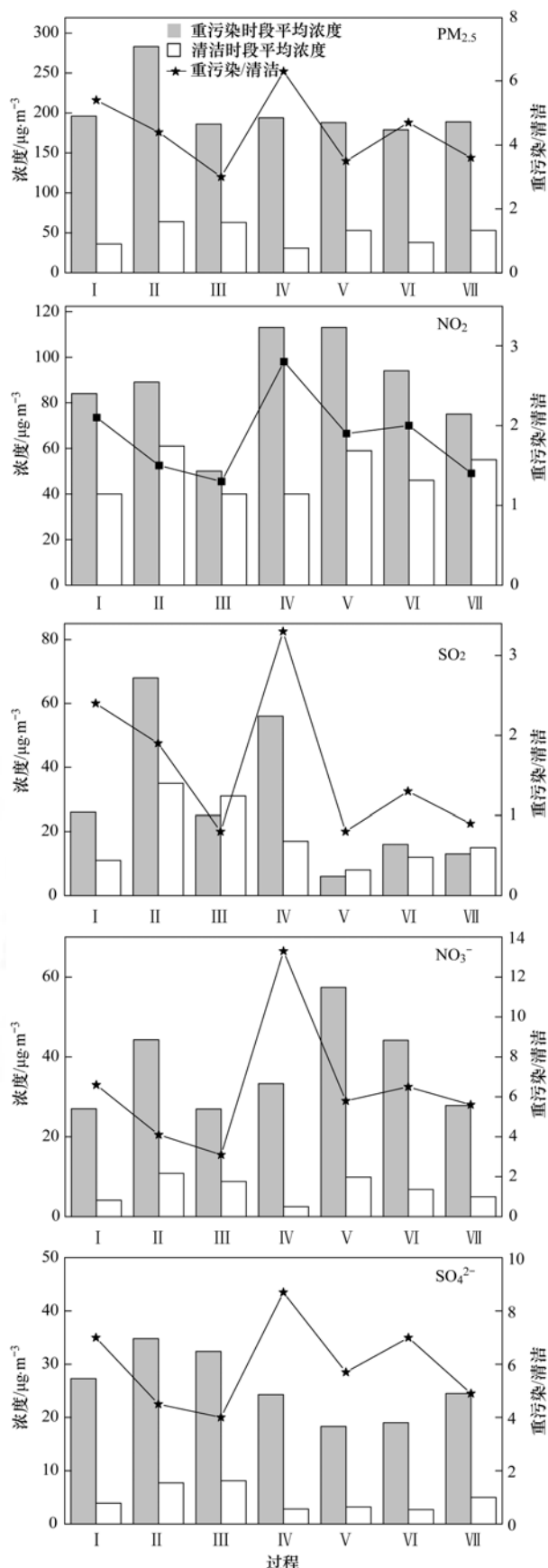


图3 重污染期间 $\text{PM}_{2.5}$ 、 NO_2 、 SO_2 、 NO_3^- 和 SO_4^{2-} 浓度均值及其与清洁时段均值的比较

Fig. 3 Average concentrations of $\text{PM}_{2.5}$, NO_2 , SO_2 , NO_3^- , and SO_4^{2-} during heavy pollution episodes compared with those during clean air weather

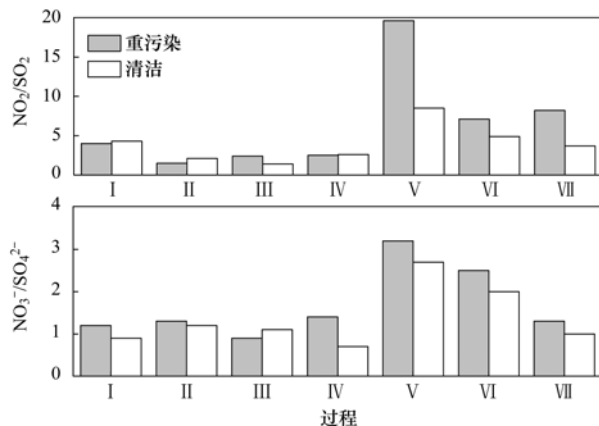


图4 重污染期间 NO_2/SO_2 比值和 $\text{NO}_3^-/\text{SO}_4^{2-}$ 比值平均值及其与清洁时段均值的比较

Fig. 4 Average values of NO_2/SO_2 and $\text{NO}_3^-/\text{SO}_4^{2-}$ during heavy pollution episodes compared with those during clean air weather

倍。7 次重污染过程期间 SO_4^{2-} 质量浓度分别是 27.3、34.8、32.4、24.3、18.3、19.0 和 24.5 $\mu\text{g}\cdot\text{m}^{-3}$, 分别占 $\text{PM}_{2.5}$ 质量的 13.9%、12.3%、17.4%、12.5%、9.7%、10.6% 和 13.0%, 7 次重污染过程期间 SO_4^{2-} 质量浓度分别是清洁天气时段的 7.0、4.5、4.0、8.7、5.7、7.0 和 4.9 倍。可见, 重污染期间 NO_3^- 和 SO_4^{2-} 浓度均显著升高, 分别是清洁天气时段的 3.1~13.3 倍和 4.0~8.7 倍, 显著高于 $\text{PM}_{2.5}$ 浓度的增长倍数(3.0~6.3 倍)。重污染期间二次水溶性离子的增长是导致 $\text{PM}_{2.5}$ 浓度显著升高的重要原因。

$\text{NO}_3^-/\text{SO}_4^{2-}$ 比值用来反映颗粒物中硝酸盐和硫酸盐的相对贡献大小^[35, 36]。7 个重污染过程期间 $\text{NO}_3^-/\text{SO}_4^{2-}$ 比值分别为 1.2、1.3、0.9、1.4、3.2、2.5 和 1.4。可见, 除过程 III 外, 其余 6 个重污染过程期间 NO_3^- 浓度均大于 SO_4^{2-} 浓度, 当前重污染过程硝酸盐转化突出。

气态前体物 NO_2 、 SO_2 向 NO_3^- 、 SO_4^{2-} 的转化过程可以用氮氧化率(NOR)和硫氧化率(SOR)来表示^[35, 36]。计算公式分别为:

$$\text{NOR} = \frac{[\text{NO}_3^-]}{[\text{NO}_3^-] + [\text{NO}_2]}$$

$$\text{SOR} = \frac{[\text{SO}_4^{2-}]}{[\text{SO}_4^{2-}] + [\text{SO}_2]}$$

式中, $[\text{NO}_3^-]$ 、 $[\text{SO}_4^{2-}]$ 、 $[\text{NO}_2]$ 和 $[\text{SO}_2]$ 分别代表对应组分的物质的量浓度($\text{mol}\cdot\text{m}^{-3}$)。

NOR 和 SOR 值较高, 说明大气中存在明显的二次无机化学转化过程。已有研究表明若 SOR 大于 0.1, 此时大气有可能发生 SO_2 的二次转化^[35, 36]。如图 5 所示, 2017 年天津市 7 次重污染过程期间, SOR 值分别为 0.38、0.24、0.48、0.27、

0.65、0.44 和 0.58, 分别是清洁天气时段的 2.5、1.9、3.3、2.6、3.1、3.5 和 3.4 倍, 说明重污染期间存在明显的硫酸盐转化过程. 7 次重污染过程期间, NOR 值分别为 0.18、0.25、0.29、0.18、0.28、0.25 和 0.21, 分别是清洁天气时段的 3.2、2.4、2.2、4.7、2.5、3.2 和 5.8 倍, 除过程 II 外, 其余过程 SOR 值均高于 NOR 值, 说明硫酸盐转化过程同样显著.

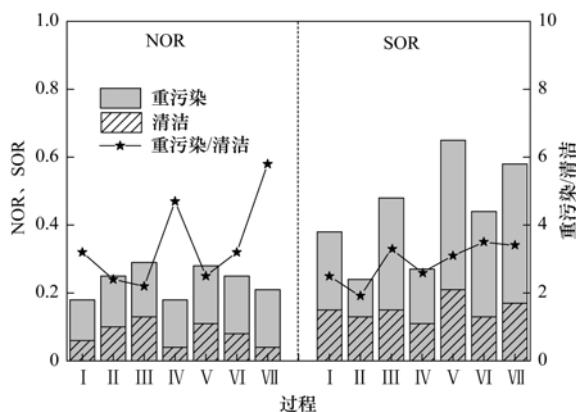


图5 重污染期间 NOR 值和 SOR 值及其与清洁时段均值的比较

Fig. 5 Average values of NOR and SOR during heavy pollution episodes compared with those during clean air weather

比较分析上半年重污染和下半年重污染的二次无机化学反应特征, 发现下半年的 NOR 和 SOR 值分别为 2.4 ~ 3.7 和 2.6 ~ 2.9, 与上半年的 2.2 ~ 4.6 和 1.8 ~ 2.9 并没有明显差别. 值得注意的是, 下半年天津市重污染期间 SO_2 浓度显著降低, 从上半年的 $25 \sim 56 \mu\text{g}\cdot\text{m}^{-3}$ 降低到 $6 \sim 16 \mu\text{g}\cdot\text{m}^{-3}$, 但是 SO_4^{2-} 浓度并未明显降低, 其中上半年重污染期间 SO_4^{2-} 为 $24.3 \sim 34.8 \mu\text{g}\cdot\text{m}^{-3}$, 下半年重污染期间 SO_4^{2-} 浓度为 $18.3 \sim 24.5 \mu\text{g}\cdot\text{m}^{-3}$. 由此可见, 在 SO_2 浓度显著降低的情况下, 重污染期间 SO_4^{2-} 浓度并未明显降低, 说明重污染期间硫酸盐粒子除来自二次无机化学反应外, 还受其他因素影响.

3 结论

(1) 天津市 2017 年重污染过程期间, 二次水溶性离子组分 NO_3^- 和 SO_4^{2-} 浓度的增长幅度最高, 显著高于 $\text{PM}_{2.5}$ 浓度的增长程度, 二次无机化学反应是导致 $\text{PM}_{2.5}$ 污染加重的重要原因.

(2) 下半年重污染期间 $\text{PM}_{2.5}$ 及 SO_2 污染程度较上半年减轻, 较清洁天气的增长程度整体低于上半年, 与下半年秋冬季期间天津市采取各项减排措施尤其是燃煤治理活动有关.

(3) 重污染过程期间 NO_2/SO_2 比值均大于 1,

其中下半年 NO_2/SO_2 比值显著高于上半年, 说明在采取各项污染源管控措施后, 移动源等排放的 NO_2 对重污染的影响比例相对增加.

(4) 天津市 2017 年 7 次重污染过程中, 6 次重污染过程期间 NO_3^- 浓度均大于 SO_4^{2-} 浓度, 6 次重污染期间 SOR 值均高于 NOR 值, 说明当前重污染期间硝酸盐转化和硫酸盐转化均较重要.

(5) 在 SO_2 浓度显著降低的情况下, 重污染期间 SO_4^{2-} 浓度并未明显降低, 说明除二次无机化学反应外, 硫酸盐生成还受其他因素影响.

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CONTENTS

Concurrent Measurement of Wet and Bulk Deposition of Trace Metals in Urban Beijing	ZHANG Guo-zhong, PAN Yue-peng, TIAN Shi-li, <i>et al.</i> (2493)
Concentration and Ecological Risk Assessment of Heavy Metals in PM _{2.5} Collected in Urban and Suburban Areas of Beijing	XU Jing, LI Xing-ru, ZHANG Lan, <i>et al.</i> (2501)
Secondary Aerosol Formation in Urban Shanghai: Insights into the Roles of Photochemical Oxidation and Aqueous-Phase Reaction	GAO Jie, QIAO Li-ping, LOU Sheng-rong, <i>et al.</i> (2510)
Secondary Inorganic Pollution Characteristics During Heavy Pollution Episodes of 2017 in Tianjin	XU Hong, XIAO Zhi-mei, CHEN Kui, <i>et al.</i> (2519)
Characterization of PM ₁₀ and PM _{2.5} Source Profiles for Emissions from Nonmetal Mineral Products Manufacturing Processes	ZHAO Xue-yan, YU Gao-feng, WANG Xin-wu, <i>et al.</i> (2526)
Characteristics of Component Particle Size Distributions of Particulate Matter Emitted from a Waste Incineration Plant	YU Zhuo-jun, WU Jian-hui, ZHANG Yu-fen, <i>et al.</i> (2533)
Characteristics and Source Apportionment of Carbon Components in Road Dust PM _{2.5} and PM ₁₀ During Spring in Tianjin Derived by Using the Quadrat Sampling Method	MA Yan, JI Ya-qin, GUO Ji-liang, <i>et al.</i> (2540)
Urban Aerosol Hygroscopicity During Haze Weather	YANG Su-ying, TIAN Zhi-jie, ZHANG Tie-ning, <i>et al.</i> (2546)
Atmospheric Pollution Characteristics and Inhalation Exposure Risk of Nitrated Polycyclic Aromatic Hydrocarbons in PM _{2.5} at the Ningdong Energy and Chemical Industry Base, Northwest China	LIU Pan-liang, JU Yuan-li, MAO Xiao-xuan, <i>et al.</i> (2556)
Health Benefit Evaluation for PM ₁₀ and PM _{2.5} Pollution Control in Zhengzhou, China, 2014-2016	HAN Shi-jie, WANG Jia, YAN Qi-she, <i>et al.</i> (2565)
Spatial-temporal Distribution of Aerosol Optical Depth and Its Main Influence Types in China During 1990-2017	LIU Ying, LIN Ai-wen, QIN Wen-min, <i>et al.</i> (2572)
Research on the Pollution Characteristics and Causality of Haze-sand Air Pollution in Beijing in Spring	WANG Yao-ting, LI Qing-chun, ZHENG Zuo-fang, <i>et al.</i> (2582)
Vessel Emission Inventories and Emission Characteristics for Inland Rivers in Jiangsu Province	XU Wen-wen, YIN Cheng-qi, XU Xue-ji, <i>et al.</i> (2595)
Atmospheric Nitrogen Dioxide, Nitric Acid, Nitrate Nitrogen Concentrations, and Wet and Dry Deposition Rates in a Double Rice Region in Subtropical China	OUYANG Xiu-qin, WANG Bo, SHEN Jian-lin, <i>et al.</i> (2607)
Characteristics of Stable Isotopes in Precipitation and Moisture Sources in the Headwaters of the Yangtze River	WANG Shao-yong, WANG Qiao-li, WU Jin-kui, <i>et al.</i> (2615)
Historical Trends of Atmospheric Trace Metal Pollution in Northern Guizhou Province as Reconstructed from Alpine Lake Sediments	LIANG Meng-yao, LIU En-feng, ZHANG En-lou, <i>et al.</i> (2624)
Distribution Characteristics of Microplastics in Qingdao Coastal Beaches	LUO Ya-dan, LIN Qian-hui, JIA Fang-li, <i>et al.</i> (2631)
Pollution Characteristics of Microplastics in Migratory Bird Habitats Located Within Poyang Lake Wetlands	LIU Shu-li, JIAN Min-fei, ZHOU Long-yin, <i>et al.</i> (2639)
Use of the Nitrogen/Carbon Ratio (N/C) and Two End-Member Sources Mixing Model to Identify the Origins of Dissolved Organic Matter from Soils in the Water-Level Fluctuation Zones of the Three Gorges Reservoir	JIANG Tao, JOERI Kaal, LIANG Jian, <i>et al.</i> (2647)
Effects of Photosynthesis of Submerged Aquatic Plants on CDOM in a Karst Water System: A Case Study from Xueyu Cave, Chongqing, China	FAN Jia-xin, JIANG Yong-jun, HE Qiu-fang, <i>et al.</i> (2657)
Indicators of Groundwater Evolution Processes Based on Hydrochemistry and Environmental Isotopes: A Case Study of the Dongyuan Drinking Water Source Area in Ji'nan City	ZHANG Ya, SU Chun-li, MA Yan-hua, <i>et al.</i> (2667)
Water Quality Analysis and Health Risk Assessment for Groundwater at Xiangshui, Chongzuo	ZHOU Jin-mei, JIANG Zhong-cheng, XU Guang-li, <i>et al.</i> (2675)
Chemical Characteristics of Groundwater and Material Sources Analysis in Buckwheat Field, Yunnan Province	ZHANG Yong, GUO Chun-qing, ZHU Yan-guang, <i>et al.</i> (2686)
C and N Transport Flux and Associated Changes of Water Quality Parameters from a Multiscale Subtropical Watershed in the Poyang Lake Area	LU Yao, GAO Yang, JIA Jun-jie, <i>et al.</i> (2696)
Pollutant Transport Analysis and Source Apportionment of the Entire Non-point Source Pollution Process in Combined Sewer Systems	FANG Jin-xiu, XIE Wen-xia, ZHU Yu-xi, <i>et al.</i> (2705)
Nitrogen Removal Efficiencies from Road Runoff by Dry Grass Swales with a Shallow Substrate Layer	DUAN Jin-kai, LI Tian, ZHANG Jia-wei (2715)
Migration Characteristics of Manganese During Rainfall Events and Its Impacts on Water Quality in a Drinking Water Source Reservoir	DENG Li-fan, HUANG Ting-lin, LI Nan, <i>et al.</i> (2722)
Potential for Phosphorus Uptake by Bed Sediments and Its Response to Carbon Additions in the Shiwuli River, Chaohu Lake Basin	LI Ru-zhong, BAO Qin, ZHANG Rui-gang, <i>et al.</i> (2730)
Sources and Distribution of Phosphorus in Sediments of the Jinpen Reservoir	MAO Xue-jing, HUANG Ting-lin, LI Nan, <i>et al.</i> (2738)
Water Quality Characteristics and Distribution of Bacterial Communities During Thermal Stratification in the Miyun Reservoir	WANG Yu-bing, WANG Xiao-yan, PANG Shu-jiang, <i>et al.</i> (2745)
Relationship Between the Vertical Distribution of Nutrients and Bacterial Community Structures in Sediment Interstitial Waters of Stratified Reservoirs with Different Water Temperatures	WANG Shen, ZHANG Si-si, XU You, <i>et al.</i> (2753)
Accumulation Characteristics and Release Regularity of Nutrients in Sediments of a Surface-flow Constructed Wetland After Long-term Operation	ZHU Yi-meng, JIANG Cui-ling, ZHU Li-qin, <i>et al.</i> (2764)
Application of Goethite Modified Biochar for Arsenic Removal from Aqueous Solution	ZHU Si-hang, ZHAO Jing-jing, YIN Ying-jie, <i>et al.</i> (2773)
Effects and Mechanisms of Methyl Orange Removal from Aqueous Solutions by Modified Rice Shell Biochar	SHI Yue-yue, SHAN Riu, YUAN Hao-ran (2783)
Performance and Membrane Fouling Properties in an Anaerobic Membrane Bioreactor for Salty Wastewater	YAN Huan-xi, XU Zhen-yu, JIN Chun-ji, <i>et al.</i> (2793)
Pollution Characteristics and Enhanced Removal of Organic Phosphorus in Effluent from a Wastewater Treatment Plant	WANG Xiao-dong, WANG Zi-wen, CHEN Ming-fei, <i>et al.</i> (2800)
Effects of Alkalinity on Partial Nitrification in a Zeolite Sequencing Batch Reactor	WANG Rui-xin, CHEN Jing, WANG Xiao-jun, <i>et al.</i> (2807)
Effects of the Physical Structure of Activated Sludge on Respiration Processes	GUO Yao, LI Zhi-hua, YANG Cheng-jian, <i>et al.</i> (2813)
Performance of an Aerobic Granular Reactor Treating Biogas Slurry from Pig Farm	LIAO Jie, YE Jia-qi, ZENG Zhi-chao, <i>et al.</i> (2821)
Spatial-temporal Characteristics and Driving Factors of Greenhouse Gas Emissions from Rivers in a Rapidly Urbanizing Area	LIU Ting-ting, WANG Xiao-feng, YUAN Xing-zhong, <i>et al.</i> (2827)
Influences of Biochar on Pollutant Removal Efficiencies and Nitrous Oxide Emissions in a Subsurface Flow Constructed Wetland	DENG Chao-ren, LIANG Yin-kun, HUANG Lei, <i>et al.</i> (2840)
Effects of the Veterinary Antibiotic Sulfamethazine on N ₂ O Emissions and the Associated Microbiological Mechanism in a Rice Field	WU Jie, LI Zhi-lin, XU Jia-ying, <i>et al.</i> (2847)
Effects of Different Levels of Nitrogen Fertilization on Soil Respiration Rates and Soil Biochemical Properties in an Alfalfa Grassland	HU Wei, ZHANG Ya-hong, LI Peng, <i>et al.</i> (2858)
Spatial Distribution and Risk Assessment of Heavy Metals in Soils from a Typical Urbanized Area	HE Bo, ZHAO Hui, WANG Tie-yu, <i>et al.</i> (2869)
Accumulation of Heavy Metals in Agricultural Soils and Crops from an Area with a High Geochemical Background of Cadmium, Southwestern China	LIU Yi-zhang, XIAO Tang-fu, XIONG Yan, <i>et al.</i> (2877)
Spatial Distribution Characteristics and Source Apportionment of Soil Heavy Metals in Chinese Wolfberry Land Based on GIS and the Receptor Model	BAI Yi-ru, ZHANG Xing, ZHAO Yun-peng, <i>et al.</i> (2885)
Spatial Distribution and Pollution Assessment of As at a Small Scale in Agricultural Soils of the Karst Region	WANG Hua, LIU Xiu-ming, LIU Fang, <i>et al.</i> (2895)
Soil Aggregate Stability and Its Stoichiometric Characteristics in <i>Robinia pseudoacacia</i> Forest within Different Vegetation Zones on the Loess Plateau, China	QU Qing, XU Hong-wei, WU Xuan, <i>et al.</i> (2904)
Straw Returning Plus Nitrogen Fertilizer Affects the Soil Microbial Community and Organic Carbon Mineralization in Karst Farmland	XU Xue-chi, SU Yi-rong, WANG Gui-hong, <i>et al.</i> (2912)
Effects of Biochar Application and Ageing on the Adsorption of Antibiotics in Purple Soil	YIN Wen-min, GUAN Zhuo, LIU Chen, <i>et al.</i> (2920)
Ammonia Oxidation in a Neutral Purple Soil Measured by the ¹³ C-DNA-SIP Method	LIU Tian-lin, WANG Zhi-hui, YAN Xiao-juan, <i>et al.</i> (2930)
Effects of Silver Nanoparticles and Silver Ions on Soil Nitrification Microorganisms and Ammonoxidation	WU Ling-li, ZHANG Xu, SHU Kun-hui, <i>et al.</i> (2939)
Insight into the Process of Mn-ANAMMOX in Soils of Agricultural Drainage Ditches	CHEN Shi, LI Zheng-kui, QIN Yun-bin, <i>et al.</i> (2948)
Effects of Different Concentrations of Tetracycline in Sludge on Ammonia Oxidizers During Vermicomposting	WU Ying, HUANG Kui, XIA Hui, <i>et al.</i> (2954)