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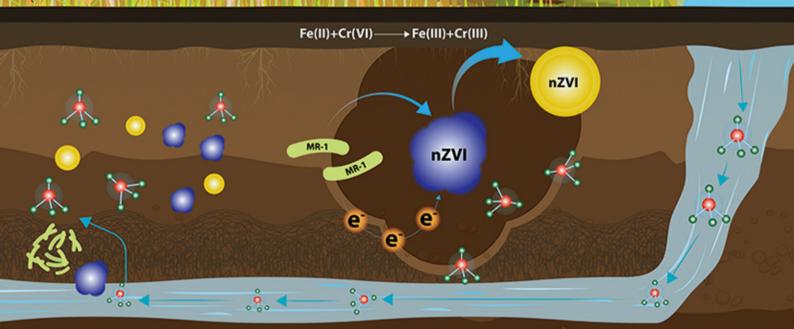
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ENVIRONMENTAL SCIENCE

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电活性微生物激活生物质炭/零价铁协同钝化Cr(VI)及机制

廖聪坚,赵晓蕾,刘凯,钟松雄,李芳柏,方利平,叶挺进,石虎砚



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乌海市夏季臭氧污染特征及基于过程分析的成因探究

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摘要:乌海市地形复杂,周边工业园区分布密集,近年来夏季 O_3 污染问题突出,且污染特征与形成机制尚不明确,分析乌海市 O_3 变化特征,探究 O_3 污染形成机制对该区域大气污染防治具有重要意义.本文在分析乌海市 2018 年 $6\sim8$ 月 3 次持续 O_3 污染过程特征的基础上,利用 WRF-CMAQ 模式系统进行模拟并根据过程分析输出结果对污染的成因进行了深入分析,探讨了区域输送和局地光化学反应对乌海市 O_3 的影响.结果表明,乌海市夏季 O_3 呈现"单峰"的日变化特征,近地面 O_3 与向下短波辐射和气温显著呈正相关,与相对湿度呈负相关;空间分布上,乌海市 3 个工业园区白天和夜间均为 0_3 低值区,乌海西南部宁夏石嘴山地区、乌海城区和西北部乌兰布和沙漠地区白天为 O_3 高值区;过程分析结果表明,输送和化学过程及其相对大小对乌海市 O_3 有决定性影响,6 月和 7 月的污染过程中局地光化学反应和输送共同导致 O_3 显著升高,且化学过程的影响是输送的两倍左右,8 月 O_3 的升高主要为输送作用的贡献;进一步对输送作用进行分解可知偏南和西北方向的输送对 O_3 的升高有较大贡献,结合前体物的排放,可能的传输来源为宁夏银川、石嘴山及巴彦淖尔等区域,因此,乌海市应在控制本地排放的基础上,加强区域联防联控,减少区域传输对 O_3 的影响.

关键词: 臭氧 (O_3) ; 社区多尺度空气质量模型(CMAQ); 污染特征; 过程分析; 乌海中图分类号: X515 文献标识码: A 文章编号: 0250-3301(2021)09-4180-11 **DOI**: 10.13227/j. hjkx. 202101094

Exploring Characteristics and Causes of Summer Ozone Pollution Based on Process Analysis in Wuhai

ZHANG Rui-xin¹, CHEN Qiang¹*, XIA Jia-qi¹, LIU Xiao¹, GUO Wen-kai¹, LI Guang-yao¹, CHEN Mei² (1. Key Laboratory of Semi-Arid Climate Change, Ministry of Education, College of Atmospheric Sciences, Lanzhou University, Lanzhou 730000, China; 2. School of Electronic and Information Engineering, Lanzhou Jiaotong University, Lanzhou 730070, China) Abstract: In recent years, summer O3 pollution has become more severe in Wuhai, where the terrain is complex and industrial parks are densely distributed. However, the characteristics and formation mechanisms of this pollution have not yet been investigated and remain unclear. Analyzing the variation and formation mechanisms of O3 is crucial to the prevention and control of air pollution in this region. By analyzing characteristics and using a WRF-CMAQ model to simulate three O₃ pollution periods in Wuhai from June to August 2018, this study explored the causes of O₃ pollution based on in-depth process analysis, and the effects of regional transportation and local photochemical reaction on O3 were also discussed. The diurnal variation of ozone exhibited a single-peak distribution, and near-surface O3 was positively correlated with short-wave radiation and temperature, and negatively correlated with relative humidity. The areas of Shizuishan in Ningxia and the Ulanbuhe desert exhibited high O₃ values during the day, while the three industrial parks in Wuhai exhibited low values during both the day and night. Process analysis showed that transportation, chemical processes, and their relative magnitudes had a significant impact on O₂. Local photochemical reactions and transport during the pollution period in June and July led to an obvious increase in O₂, while the impact of chemical processes was about twice as large as that of transport. The increase of O3 in August was mainly caused by transport. Further decomposition of the transportation effect showed that transportation in the south and northwest directions had a remarkable effects on the increase of O₃. Together with the emission of O₃ precursors, the main sources of transportation were the Yinchuan, Shizuishan, and Bayannaoer regions. Therefore, Wuhai and neighboring cities should strengthen regional joint prevention and control by jointly formulating and implementing control measures for air pollution to reduce the impact of regional transmission on O3.

 $\textbf{Key words}: ozone(O_3); \ community \ multiscale \ air \ quality \ model(CMAQ); \ pollution \ characteristics; \ processes \ analysis; \ Wuhai \ and \ analysis; \ whai \ analysis; \ what \ analysis;$

近地面臭氧(ozone, O₃)主要由人为排放的挥发性有机物(VOCs)和氮氧化物(NO_x)等前体物在太阳光照射下经复杂的光化学反应产生. O₃ 污染不仅会导致农作物减产,树木生长缓慢,同时对气候变化和人体健康也会产生不利影响^[1-4]. 基于 O₃ 污染的危害性,国内外学者已从区域和城市尺度上对 O₃ 污染的时空变化规律、主要影响因素和成因分析等开展了大量研究^[5-8].

从 O₃ 的日变化特征来看,我国香港、杭州、青岛和烟台等地城市区域可观察到 O₃ 的双峰变化,白天和夜间 O₃ 峰值相差不大,分别出现在 14:00 和 04:00左右^[9~11],京津冀和珠三角等地区 O₃ 多呈现

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单峰型特征,但不同区域夜间 0,浓度水平存在差 异[12~14]. 在 0, 污染成因探究分析方面,后向轨迹、 潜在源区分析,臭氧生成潜势评估,经验动力学模拟 方法(EKMA)和数值模拟等方法得到了广泛应 用[15~19],其中社区多尺度空气质量模型(community multiscale air quality model, CMAQ) 过程分析(PA) 模块输出的 IPR(integrated process rate)可以定量区 分大气中不同的物理和化学过程对 O, 形成的相对 贡献,因此在 O, 污染成因分析中发挥了重要的作 用^[20,21]. 利用 IPR 探究我国 O, 污染成因的研究主 要集中在京津冀、珠三角和长三角地区,也有部分学 者将过程分析的结果应用于成都、青岛等地 区[22~24]. Wang 等[25] 对 2004 年珠江三角洲空气质 量综合观测实验(PRIDE-PRD2004)期间的 O。污染 进行了模拟与过程分析,发现光化学反应是珠三角 地区 09:00~15:00 O。浓度增大的主导因素; Shu 等[26]利用 IPR 分析了副热带高压和台风系统对长 三角地区一次区域性 0, 污染事件的综合影响,表明 垂直扩散和气相化学过程是 03 维持高值的主要原 因;李莉[27]对长三角地区 0,污染开展过程分析结 果显示:03最大浓度出现时段内,污染输送对地面 0, 贡献较大, 气相化学过程主要发生在 300~ 1500 m,通过向地面传输使得近地层 O, 升高; 赵 文龙^[28]利用 IPR 定量分析了广州地区海陆风条件 下各物理化学过程对 0, 的贡献, 发现垂直输送为城 市和郊区站点 0, 主要源过程. 综上, 受源排放特征、 自然地理环境和气象条件等影响,不同区域和城市 O, 污染特征和形成机制存在显著差异.

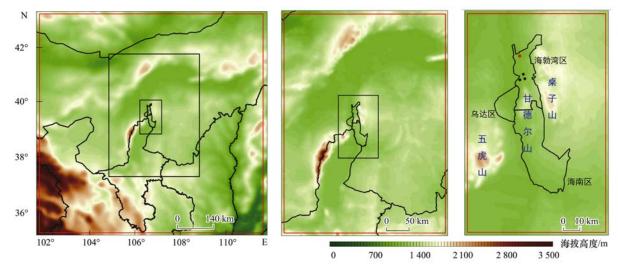
乌海市处于青藏、黄土和蒙古三大高原结合部的干旱地区,受"三山两谷一条河"地形的影响,局

地气象条件复杂,且由于区域丰富的煤炭资源,形成 了以煤炭开采加工和化工行业为主的园区"围城" 的工业分布特征,使得乌海 0、污染出现明显的逐年 上升趋势, 2019 年乌海市 O, 共超标 28 d, 占总超标 天数的 40.6%, O₃ 日最大 8 h 滑动平均值第 90 百 分位数浓度较 2018 年上升 1.3%, 当地夏季 0, 污染 问题日益突出,但 0、污染特征及不利的扩散条件、 集中的排放分布等对 0, 污染的影响尚不明确. 基于 此本文选取乌海市夏季 2018 年 6 月 10~14 日 (P1)、7月16~21日(P2)和8月18~24日(P3)的 3次 O, 污染过程, 在利用地面和气象观测数据分析 乌海市 0, 污染特征及其与气象因子关系的基础上, 结合 WRF-CMAQ 模式系统,进一步探究了3次污染 过程中乌海及周边区域 O, 的时空变化规律,并根据 IPR 输出结果量化了各个物理化学过程对当地 0, 生成的贡献,探讨了区域输送和局地光化学反应对 3次 O, 污染过程的影响,深入分析了污染过程持续 的原因,以期为乌海市及处于干旱地区地形复杂城 市的 0, 污染防治工作提供科学指导.

1 材料与方法

1.1 研究区域及监测点位

乌海市地处内蒙古自治区西南部,由东到西依次为桌子山、甘德尔山和五虎山,中间形成两条狭长的谷地,地势东高西低,地形复杂.乌海及周边分布着海勃湾千里山、乌达、海南西来峰、蒙西、乌斯太、棋盘井和石嘴山河滨等共7个煤化工为主的工业园区,形成了园区"围城"的工业分布特征.乌海市共有3个国控站点(图1),分别为聚英学校、林业局和中海勃湾学校站点,1个气象站点(图1),均位于海



黑框为 WRF 模拟区域, 红框为 CMAQ 模拟区域, 黑点为国控站点, 红点为气象站点

图 1 研究区域及监测站点分布示意

Fig. 1 Modeling domains and location of monitoring stations

勃湾区.

1.2 数据来源及模式设置

本研究采用的气象数据为乌海市气象局公布的逐小时气象观测资料. 空气污染物数据来自全国城市空气质量实时发布平台(http://106.37.208.233:20035/)公布的乌海市国控站点小时浓度数据. 利用 WRF v3.6.1 模拟气象场, CMAQ v5.0.1 对乌海市 O₃ 及其前体物进行模拟. 模拟区域如图 1 所示, CMAQ 采用水平分辨率为 9、3 和 1 km 的三层嵌套. WRF 模式的气象资料来源于 NCEP 发布的FNL 全球再分析资料,空间分辨率为 1°×1°,时间间隔为 6 h. 模式源排放清单采用清华大学开发的2016 年 MEIC 排放清单(http://meicmodel.org),空间分辨率为 0.25°×0.25°[29~32],其中乌海市排放清单采用本课题组自建的2018 年高分辨率源排放清单(HEI-WH18),涵盖化石燃料燃烧源、工艺过程

源、移动源、溶剂使用源、扬尘源和天然源等 11 大类源,包括 SO_2 、 NO_x 、CO、VOCs、 NH_3 、 PM_{10} 、 $PM_{2.5}$ 、 BC 和 OC 等 9 种污染物的排放.

2 结果与讨论

2.1 模拟结果评估

采用平均偏差(MB)、平均误差(ME)、均方根误差(RMSE)、皮尔逊相关系数(R)和一致性指数(IOA)对 WRF和 CMAQ的模拟效果进行定量评价(表1).相关系数体现了模拟结果与观测数据的相关程度,即趋势是否一致, MB 体现模拟对观测的相对偏离; ME 反映模拟对观测的绝对偏差; RMSE 是模拟值误差大小的量度,均方根误差越小表明预报的准确度越高; IOA 指数代表模拟值和观测值的吻合程度,越接近于1 代表模拟效果越好.

表 1 WRF 和 CMAQ 模拟结果与监测数据对比

Table 1	Statistical	comparison	between t	he observed a	and simulated	meteorological	variables
	MB	C: 1	ME		RMSE	1	R

要素	4,70	0-	MB	0	14	ME			RMSE	1	1	R		1 '	IOA
女系	1	P1	P2	P3	P1	P2	P3	P1	P2	P3	P1	P2	Р3	P1	P2 P3
辐射/W·m ⁻²	/	- 36. 27	80.04	- 22. 07	148. 89	161. 59	153. 27	225.06	276.60	250. 98	0.89	0.69	0.77	0. 93	0. 81 0. 87
风速/m·s ⁻¹	111	0.68	1.44	0. 44	1.47	2. 13	1. 23	1.83	2. 75	1.59	0.28	0. 16	0.39	0.55	0.44 0.59
温度/℃	511	0. 48	1.68	1.69	1. 85	2. 50	2. 50	2. 22	3. 10	3. 27	0.95	0. 90	0.83	0. 97	0. 93 0. 87
相对湿度/%	6 L	-1.98	-7.26	-2.39	10.02	11.04	14. 96	13. 59	13. 45	19.32	0.59	0.88	0.60	0.77	0. 91 0. 78
聚英学校 O_3/μ	lg•m ⁻³	-9.16	- 24. 72	-8.68	31. 52	44. 19	36. 65	38. 51	53. 52	43.06	0.63	0.49	0.62	0.76	0. 61 0. 74
林业局 O ₃ /μg·	m ⁻³	-2.55	- 25. 82	-7.58	44. 38	44. 75	37. 15	56. 22	55.00	42. 83	0.67	0.54	0.62	0.72	0.64 0.73
中海勃湾学校(O ₃ /μg·m ⁻³	-12.86	2. 96	-16.32	39. 81	50.06	41. 85	46. 60	62. 73	49. 96	0.60	0.55	0.62	0.72	0.60 0.69

对于向下短波辐射(SR)和气温(T),3个时 段内模拟值与观测值均有很好地一致性: P3 时段 相对湿度(RH)的模拟结果与观测的差异最大, ME 和 RMSE 分别为 14.96% 和 19.32%; 对于风 速(WS), P3 时段内模拟值与实测值的 IOA 最大 为 0.59,与其他研究相近[33~35]. 从 0,的模拟上 看, P1 时段, 林业局站点模拟值与监测值的差异 最大, ME 和 RMSE 分别为 44.38 µg·m⁻³和 56.22 $\mu g \cdot m^{-3}$,站点模拟值与监测值的 R 在 0.60 ~ 0.67 之间; P2 时段,中海勃湾学校站点 RMSE 最大为 62. 73 μg·m⁻³, 站点模拟值与监测值的 IOA 在 0.60~0.64 之间; P3 时段,中海勃湾学校站点 MB、ME 和 RMSE 最大,分别为 - 16.32、41.85 和 49.96 μg·m⁻³,除8月21日、22日外,其他时段 内模式未模拟出日间 O, 浓度的高值, 站点模拟值 与监测值的 R 均为 0.62; O, 模拟结果与监测结果 的相关性与目前光化学传输模式模拟 O, 的相关 性一致[36~39],整体来看, CMAQ 的模拟结果在 3 个站点均能再现 O, 的变化.

2.2 0、污染特征及与气象因子的关系

2.2.1 0, 时间变化特征及与气象因子的关系

由于气象因素在 O_3 的形成、扩散稀释、传输和沉降等过程起着非常重要的作用,为了更好地了解 3 次污染过程 O_3 变化特征及其形成机制的异同情况,对 3 次污染过程中 O_3 与 SR、T 和 RH 的日变化特征进行分析(图 2).

 O_3 浓度与 T 的日变化趋势吻合较好,均呈单峰型分布,峰值出现在 $12:00 \sim 18:00$ 左右, 3 个时段内 O_3 -1h 和 O_3 -8h 最大超标倍数分别在 $1.23 \sim 1.64$ 和 $0.08 \sim 0.29$ 之间;随后 T 逐渐下降, O_3 浓度也出现下降趋势,至次日 06:00 左右 T 达到 1 d 中的最低值, O_3 浓度也降至最低,在 $3 \sim 18$ $\mu g \cdot m^{-3}$ 内变化;之后 T 上升, O_3 浓度开始不断升高,至中午或午后达到 1 d 中的最大值.相关性分析显示,3 个站点 O_3 浓度与 T 在 0.05 置信水平(双侧检验)下显著相关,P1 和 P3 时段 Pearson 相关系数均大于 0.88. SR 和 T 的变化趋势基本一致,随着 SR 达到最大, O_3 浓度也逐渐达到 1 d 中的最大值,受日照的

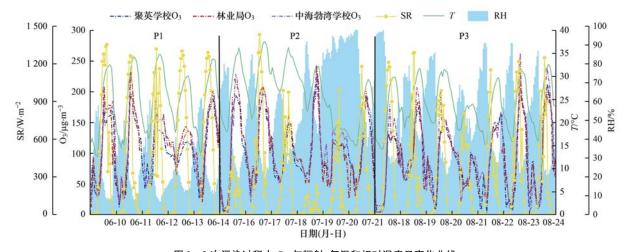


图 2 3 次污染过程中 O_3 与辐射、气温和相对湿度日变化曲线

Fig. 2 Daily variation of O₃ and meteorological factors during three pollution processes

影响, SR 在 18:00 左右之后显著下降, O_3 浓度的高值可在 18:00 后维持一段时间, 再逐渐下降, 夏季 SR 和 T 是影响近地面 O_3 浓度的关键因素. 大气中的水汽通过影响太阳辐射、前体物浓度从而对 O_3 产生影响, 剔除降水影响后, 3 个时段内 Pearson 相关系数大于 -0.81(0.05 置信水平, 双侧检验), RH 与 O_3 显著负相关. 综合 SR、T 和 RH 对 O_3 浓度的影响, 强光照、高温和低湿的条件,有利于乌海市 O_3 的生成, 这与国内外大多数研究结果相一致 O_3 不良的。

2.2.2 0, 空间分布特征

根据 O_3 的"单峰"日变化特征,得到 3 次污染过程中夜间($O0:O0\sim O6:O0$)和白天($I2:O0\sim I8:O0$)O₃的分布情况(图 3).

夜间,宁夏北部和乌海东部为 0,高值区,乌海 市工业园区、海勃湾区(电厂周边)和宁夏石嘴山地 区由于 NO 等物质对 O, 的消耗而呈现低值区, 低于 40 μg·m⁻³;同时乌海市城区夜间受东南风控制,3 次污染过程均存在显著从海南工业园区向乌达工业 园区及其北部的前体物输送,在此路径上的区域均 为 O, 浓度低值区. 白天, 乌海市 3 个工业园区(图 3,蓝色圈)及国控站点附近两个电厂位置(图3,紫 色圈)为 O,浓度低值区, O,浓度较低的原因可能 是由于工业园区和电厂排放的 VOCs 及 NO, 在高温 光照的条件下发生光化学反应, O, 的消耗大于生 成导致的;乌海西南部宁夏石嘴山地区、西北部乌 兰布和沙漠地区和乌海城区 O。浓度较高,但不同污 染过程 O, 的高值中心和浓度水平存在差异, P1 时 段从石嘴山地区-海南区-乌达区整体 O, 浓度较高, 达到 140 μg·m⁻³, P2 时段石嘴山地区 O₃ 高值仍维 持在 140 μg·m⁻³左右,但乌海城区 O₃ 浓度在 110~ 120 μg·m⁻³之间, P3 时段区域整体 O₃ 浓度相差不 大. 从 NO_x 和 VOCs 的排放分布来看(图 4),乌海市西南部宁夏银川、石嘴山地区和北部巴彦淖尔地区均存在前体物的排放,为可能的 O₃ 源区,但区域传输和本地光化学反应对 3 次污染过程的影响仍需进一步量化.

2.3 模拟时段内 O, 污染过程分析

CMAQ 过程分析模块 IPR 输出结果具体包括 7 个过程:垂直扩散(VDIF)、垂直平流(ZADV)、水平扩散(HDIF)、水 平 平流(HADV)、化学 过程(CHEM)、干沉降(DDEP)和云过程(CLDS). 从表 2 和图 5 可知, HTRA 代表水平输送,为水平扩散和水平平流的总和; VTRA 代表垂直输送,是垂直扩散和垂直平流的总和; TRAN 代表输送过程,为水平输送和垂直输送过程的总效应. 利用 IPR 量化对不同物理化学过程在 3 次 O₃ 污染过程中的贡献情况(表 2 和图 5).

根据 IPR 输出结果, DDEP 在 3 次污染过程中 对 O, 均为负贡献, 起到降低 O, 的作用, 3 个站点 DDEP 最大负贡献可达 - 11 µg·m⁻³左右,平均负贡 献率在 18.69%~45.77%之间,08:00~18:00 DDEP对 O, 的消耗较大,尤其在中午 CHEM 和 TRAN 的贡献量级变小,此时 DDEP 将成为 O, 的主 要汇之一. CLDS 在大部分时刻均对 O, 有正贡献, 3 次污染过程平均正贡献率 1.41%~51.82% 之间. TRAN 和 CHEM 对乌海市 O, 贡献的变化幅度较大, 且输送和化学过程的相对大小对 0、浓度有决定性 影响. P1 和 P2 时段, 00:00~10:00 左右 TRAN 对 0,有较大的正贡献,站点平均正贡献在75.89%~ 95.06%之间,同时00:00~06:00左右, CHEM与 TRAN 的贡献相反,主要起到消耗 O, 的作用, P1 和 P2 时段站点平均负贡献分别为 - 83.95% 和 -88.96%,整体来看 00:00~06:00 O,浓度的净变

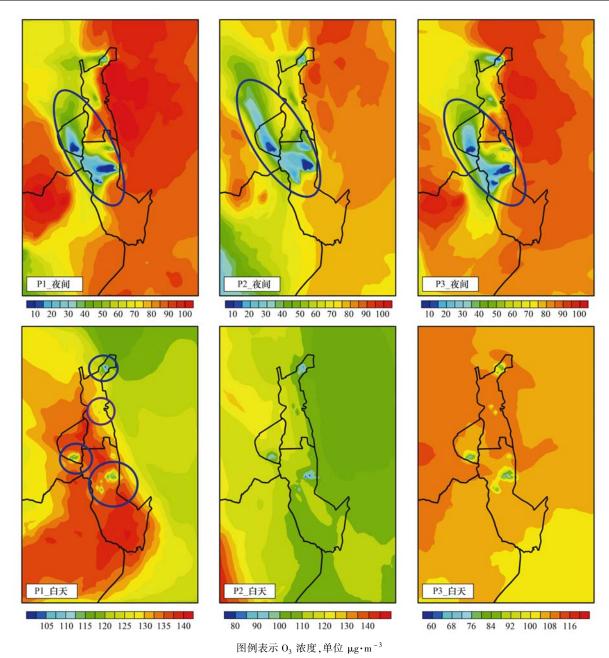


图 3 3 次污染过程中 O_3 空间分布特征

Fig. 3 Spatial distribution of O_3 during different pollution processes

表 2 不同污染时段内国控站点物理化学过程平均贡献率/%

Table 2 Average contribution rate of different physical and chemical processes at air quality monitoring stations during different pollution periods/%

n-l F/L	*F-E	DDEP		Cl	LDS	CH	IEM	TRAN		
时段	站点	源	汇	源	汇	源	汇	源	汇	
	聚英学校	_	- 30. 63	29. 39	_	47. 95	- 87. 31	77. 82	- 58. 88	
P1	林业局	_	-40.59	5.71	-2.59	72. 99	-91.77	86. 19	-31.89	
LI	中海勃湾学校	_	-45.77	51.82	-9.36	68. 58	- 72. 78	75. 89	- 29. 91	
	平均	_	- 39. 00	28. 97	-5.98	63. 17	- 83. 95	79. 97	-40.23	
	聚英学校	_	- 18. 69	8. 98	_	78. 90	- 92. 23	86. 73	-81.16	
P2	林业局	_	- 19. 01	0.86	-1.37	85.78	- 95. 81	95.06	- 70. 42	
ГΔ	中海勃湾学校	_	-42.98	_	-6.25	68. 29	- 78. 83	86.04	- 32. 28	
	平均	_	- 26. 89	4. 92	-3.81	77.66	- 88. 96	89. 28	-61.29	
	聚英学校	_	- 32. 81	39. 37	_	18. 86	- 82. 16	67. 09	- 33. 43	
Р3	林业局	_	- 28. 67	1.41	-1.19	8.75	- 80. 78	98. 26	_	
13	中海勃湾学校	_	- 32. 88	_	- 12. 67	18. 94	- 72. 00	95.06	- 10. 72	
	平均	_	- 31. 45	20. 39	-6.93	15. 52	- 78. 31	86. 80	- 22. 08	

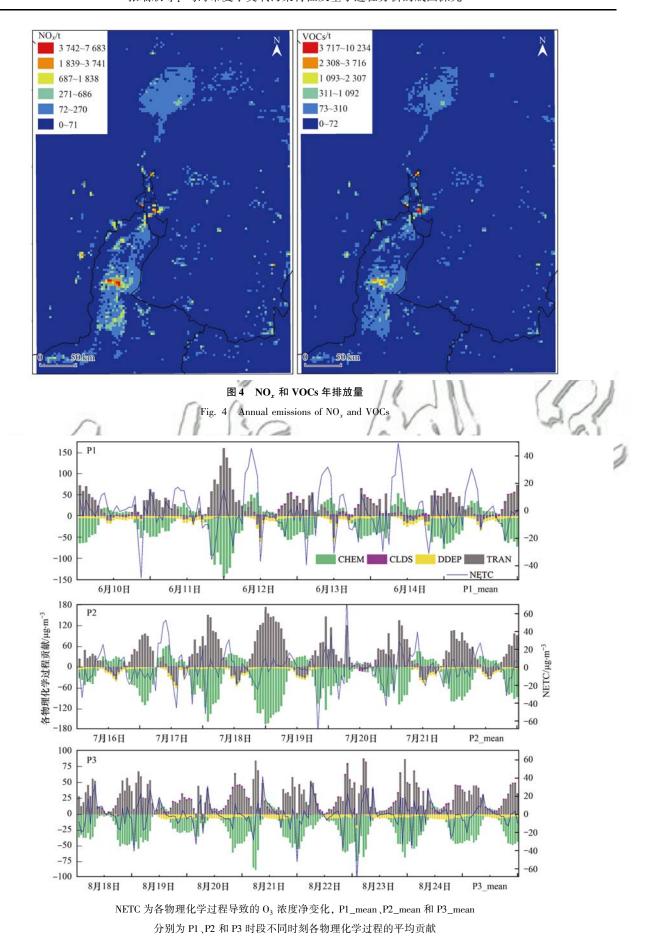


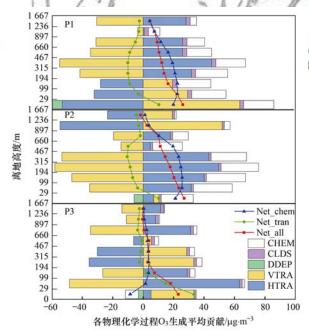
图 5 不同污染时段内各物理化学过程对 O_3 小时浓度的贡献 Fig. 5 Contribution of various physical and chemical processes to the hourly O_3 concentration

化在 ± 25 μ g·m⁻³以内,不会出现 O₃ 显著升高情况; 07:00~10:00 左右 TRAN 和 CHEM 对 O₃ 浓度贡献的量级减小,但由于光照促进了光化学反应的发生, CHEM 对 O₃ 的影响由夜间的负贡献转变为正贡献,此时 TRAN 和 CHEM 共同作用导致 O₃ 显著上升, O₃ 净变化出现大于 30 μ g·m⁻³的情况; 11:00~17:00左右, CHEM 保持正贡献,但 TRAN转变为负贡献,消耗 O₃; 17:00~00:00 由于光照逐渐减弱消失, CHEM 成为 O₃ 的主要汇, TRAN 是 O₃ 的源之一. P3 时段,除个别时刻外, TRAN 对 O₃ 均为正贡献; 10:00~15:00 光照较强时刻的 CHEM 在 8 月 19~21 日和 8 月 24 日对 O₃ 有正贡献, 8 月 18 日和 8 月 23 日有较小负贡献,从平均来看 CHEM 在 10:00~14:00 是 O₃ 的源之一,但贡献不大,平均

由于输送过程与气象条件密切相关,且输送的污染物会对局地光化学过程产生影响,因此,3次污染过程中近地层O3主要来源于局地光化学生成,还是周边地区污染输送需要进一步讨论.对3次过程中站点O3净增量最大时刻前后站点垂直方向上各物理化学过程的贡献进一步分析(图6).

正贡献率为 15.52%. 说明 P3 时段输送是导致 O。

浓度升高并维持的主要原因.



Net_all 为所有物理化学过程影响下 O_3 的净变化,Net_tran 为输送过程导致的 O_3 变化,Net_chem 为化学过程导致的 O_3 变化

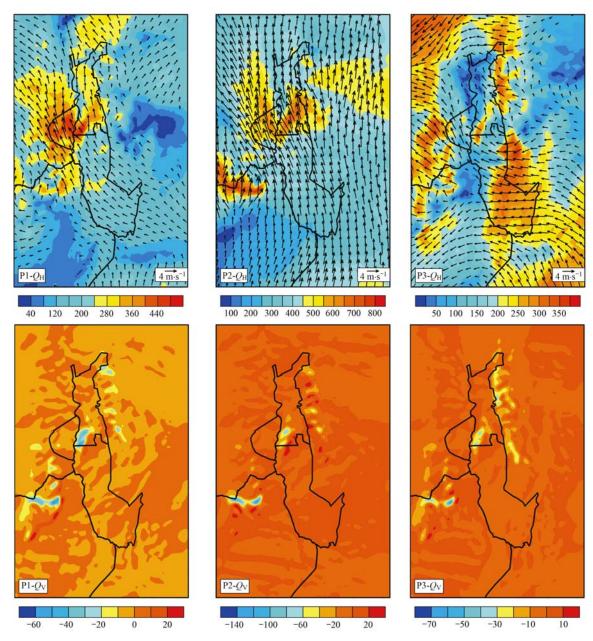
图 6 各物理化学过程对 O3 生成的平均贡献垂直廓线

Fig. 6 Vertical profile of averaged contribution of various physical and chemical processes to O_3 generation

P1 和 P2 时段最低层(0~29 m)输送和化学过程对 O₃ 的贡献均为正,其余高度输送过程的贡献均为负,说明输送和化学过程共同导致了低层 O₃ 的快

速升高; 这与 IPR 站点小时贡献的结果一致. 进一 步将输送作用分解到水平和垂直方向上, P1 时段 低层(0~194 m), VTAR 的贡献为正, HTAR 为负, 在中高层(>194 m)则相反,表示中高层 O, 水平传 输至乌海上空,通过局地下沉运动,造成乌海市 O。 浓度的积累,并在低层通过水平运动扩散出去,但扩 散的量级较小,因此0~29 m 由输送过程导致 O,浓 度的增量为 10.34 μg·m⁻³; P2 时段,最低层(0~29 m), HTAR 和 VTAR 均为正贡献,在 29~897 m, VTAR 的贡献为负, HTAR 的贡献为正,说明在 P2 时段 0~897 m 水平输送造成乌海 O, 的积累,进一 步经垂直输送向低层和高空扩散,最终导致低层 O。 平均升高 10.35 μg·m⁻³. P1 和 P2 时段, CHEM 在 所有高度对 O, 的贡献均为正, 且随着高度的增加由 气相化学反应导致的 0, 浓度增量略有增大后逐渐 减小,最低层由 CHEM 导致的 O, 变化量分别为 21.11 μg·m⁻³ 和 25.47 μg·m⁻³, 最低层 CHEM 对 O, 的贡献是输送过程的一倍左右,说明局地化学反 应在P1 和P2 时段主导了O, 的升高,并且在整个边 界层内均是 O, 的重要来源之一. P3 时段, 最低层 CHEM 和 DDEP 的贡献为负, HTAR、VTAR 的贡献 为正,随着高度的增加 CHEM 虽有正贡献,但贡献 量均小于 3.68 $\mu g \cdot m^{-3}$,且低层输送过程对 O_3 的贡 献平均达到 33. 27 μg·m⁻³,持续的输送过程正贡献 是 O3 暴发式升高的主要原因, CHEM 则起到消耗 O, 的作用. 若白天输送过程正贡献使得 O, 小时净 增量大于 40 μg·m⁻³为暴发机制,具有暴发机制的 污染为输送型污染,不具备该暴发机制且日间输送 贡献明显为负,化学贡献显著高于平均值的污染型 为局地生成型污染[22,24],考虑地域差异, P3 时段输 送的平均正贡献接近 40 µg·m⁻³,满足输送区域输 送型污染特征; P1 和 P2 时段,化学贡献是输送的 两倍左右,但二者均为正贡献,无显著局地生成型污 染特征,因此, P1 和 P2 时段的污染过程是由输送 和局地生成共同导致的.

根据 O_3 净增量较大时刻前后平均输送通量分布(图 7),进一步分解 3 次污染过程中的输送作用, P1 和 P2 时段海勃湾区 O_3 水平输送通量分布情况相似,乌海市整体被偏南风控制,海勃湾区甘德尔山附近和乌达区 O_3 向南输送显著, P1 时段海勃湾区国控站点附近输入和输出通量相差不大在 300~450 μ g·(s·m²)⁻¹变化, P2 时段,甘德尔山附近向南的输送通量大于 700 μ g·(s·m²)⁻¹,显著高于海勃湾市区的向南输送,市区南边界输入通量大于北边界输出通量,导致 O_3 的积累,站点附近垂直输送



 $Q_{\rm H}$ 为 O_3 水平输送通量, $Q_{\rm V}$ 为 O_3 垂直输送通量;图例表示输送通量,单位 μg·(s·m²) $^{-1}$

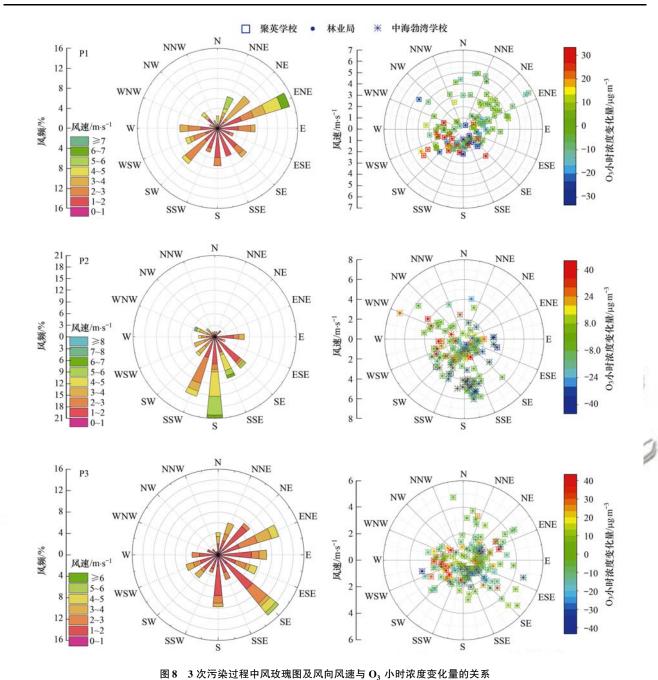
图 7 不同污染过程近地层($0 \sim 29 \text{ m}$) O_3 输送通量小时平均

Fig. 7 Hourly average of O_3 transport flux in the near-surface layer (0-29 m)

通量在 $-20 \sim 20 \ \mu g \cdot (s \cdot m^2)^{-1}$ 左右,因此 P1 和 P2 时段的输送主要以偏南方向的输送导致 O_3 净累积为主. P3 时段,海勃湾区向西北方向的输送小于 100 $\mu g \cdot (s \cdot m^2)^{-1}$,但甘德尔山附近和市区东北部向海勃湾的输送通量在 $350 \ \mu g \cdot (s \cdot m^2)^{-1}$ 左右,水平方向上输入大于输出,且这两个区域在垂向上向下输送通量在 $-50 \sim -30 \ \mu g \cdot (s \cdot m^2)^{-1}$,向下输送扩散将进一步导致海勃湾区 O_3 的积累,因此 P3 时段偏南和东北方向的水平输送以及垂向输送共同导致乌海 O_3 的积累. 综上, O_3 的输送方向与 O_3 前体物排放的分布具有很好的一致性.

结合监测数据,分析3次0,污染过程中的风玫

瑰图及对应国控站点近地面风向、风速与 O₃ 小时净变化量的关系可知(图 8), P1、P2 和 P3 时段内的主导风向分别为东北风、南风和东南风, 1~3 m·s⁻¹风速出现频率分别为 72%、61% 和 82%.由风向、风速和 O₃ 小时净变化量的关系,污染时段内主导风向上均未出现 O₃ 小时浓度的显著升高; P1 时段风向分布在 E~W 扇区, P2 时段在 SSE~WNW扇区, P3 时段在 S~W 和 NE~E 扇区,风速为 1~3 m·s⁻¹时(P2 时段存在风速大于 6 m·s⁻¹的情况),国控站点 O₃ 小时净变化量较大,与模式输出结果中各污染过程 O₃ 水平输送通量方向基本一致,进一步证明了当存在偏南或者偏东北风,风速在 1~3



g. 8 Wind rose diagram and the relationship between wind direction, wind speed, and hourly variation of O₃ concentration

 $m \cdot s^{-1}$ 时,乌海市易出现 O_3 快速升高现象.

3 结论

(1)根据 IPR 输出结果,干沉降对 O₃ 的贡献均为负,为 O₃ 的汇;云过程对 O₃ 的影响较小,在大部分时刻均有正贡献,是 O₃ 的源之一;输送和化学过程及其相对大小对乌海市 O₃ 有决定性影响,其中P1 和 P2 时段,07:00~10:00,输送和化学过程共同导致 O₃ 浓度的显著上升,化学过程的作用是输送的两倍左右;P3 时段水平和垂直输送是 O₃ 的浓度升高的主要原因,化学过程的正贡献不大,满足区域输送型污染特征.

- (2)区域 O₃ 输送通量的分布进一步证明偏南和东北方向上的水平输送对乌海的 O₃ 的升高有显著贡献,乌海市应在控制乌海市本地排放的基础上,加强区域性 O₃ 的联防联控与协同治理工作.
- (3)夏季 3 次污染过程中,乌海市 0₃ 均呈现 "单峰"的日变化特征,峰值出现在 12:00~18:00. 近地面 0₃ 浓度与向下短波辐射、气温呈正相关,与相对湿度呈负相关.乌海市 3 个工业园区及电厂位置白天和夜间均为 0₃ 低值区;白天乌海西南部宁夏石嘴山、西北部乌兰布和沙漠地区及乌海城区为 0₃ 浓度高值区;结合 NO_x和 VOCs 排放的分布情况,乌海市 0₃ 可能的传输源区为宁夏银川、石嘴山和巴彦淖尔等区域,夜间乌海市本地存在海南工业

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