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## 2022年8月成渝两地臭氧污染差异影响因素分析

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摘要: 2022年8月成都和重庆呈现显著的臭氧 $(O_3)$ 污染差异,成都 $O_3$ 污染天高达20 d,重庆无 $O_3$ 污染天,本文从前体物排放水平和气象条件分析此差异的影响因素.结果表明:①成都52种挥发性有机物(VOCs)(包含 26种烷烃、16种芳香烃和10种烯烃)的总体积分数 $(18.8 \times 10^9)$ 是重庆 $(6.6 \times 10^9)$ 的2.8倍,总 $O_3$ 生成潜势 $(OFP=51.2 \times 10^9)$ 是重庆 $(25.0 \times 10^9)$ 的2.0倍,总·OH损耗速率 $(L_{OH}=3.9 \, \mathrm{s}^{-1})$ 是重庆 $(2.3 \, \mathrm{s}^{-1})$ 的1.7倍.成都OFP前3是乙烯、间/对-二甲苯和异戊二烯;重庆OFP前3是异戊二烯、乙烯和丙烯.重庆仅烯烃对 $O_3$ 的贡献率是60.7%,而成都烯烃和芳香烃的OFP分别是重庆的1.6倍和2.9倍.综上,成都VOCs总体积分数、大气光化学活性和 $O_3$ 生成潜势均较重庆高。②成渝两地 $L_{OH}$ 排名第1均为异戊二烯,表明8月 $O_3$ 污染生物源贡献显著,但生物源排放活性与温度存在响应关系,8月14~24日重庆高温 $(38.3^\circ C)$ 使得生物源排放活性下降,而成都温度 $(34.9^\circ C)$ 加剧了生物源排放活性。③重庆水平、垂直大气扩散条件整体优于成都,且成都受到了区域性污染传输的影响。

关键词: O3污染; 影响因素; 传输轨迹; 前体物排放水平; O3生成潜势

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# Analysis of Influencing Factors of Ozone Pollution Difference Between Chengdu and Chongqing in August 2022

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Abstract: In August 2022, Chengdu and Chongqing showed significant differences in ozone  $(O_3)$  pollution. Chengdu had  $O_4$  pollution days for 20 days, whereas Chongqing had no  $O_3$  pollution days. In this study, we analyzed the influencing factors of this difference from the emission level of precursors and meteorological conditions. The results showed that: ① the total mixing ratio of 52 VOCs (volatile organic compounds) (including 26 alkanes, 16 aromatics, and 10 alkenes) in Chengdu  $(18.8 \times 10^{-9})$  was 2.8 times that of Chongqing  $(6.6 \times 10^{-9})$ , and the total  $O_3$  formation potential (OFP)  $(51.2 \times 10^{-9})$  was 2.0 times that of Chongqing  $(25.0 \times 10^{-9})$ . The  $\cdot OH$  radical loss rate  $(L_{\cdot OH})$   $(3.9 \text{ s}^{-1})$  was 1.7 times that of Chongqing  $(2.3 \text{ s}^{-1})$ . The top three OFP in Chengdu were ethylene, mtp-xylene, and isoprene, and those in Chongqing were isoprene, ethylene, and propylene. The contribution rate of alkenes to  $O_3$  in Chongqing was 60.7%, whereas the OFP of alkenes and aromatics in Chengdu were 1.6 times and 2.9 times that in Chongqing. In conclusion, the total mixing ratio of VOCs, atmospheric photochemical activity, and  $O_3$  formation potential of Chengdu were higher than those of Chongqing. ② Isoprene was ranked first place in  $L_{\cdot OH}$  in both Chengdu and Chongqing, indicating that the contribution of biogenic sources to  $O_3$  pollution in August was significant. However, the biogenic source emission activity was in response to temperature. From August 14 to 24, the high temperature in Chongqing (38.3%) decreased biogenic source emission activity, whereas the temperature in Chengdu (34.9%) increased the biogenic sources emission activity. (3) The horizontal and vertical atmospheric diffusion conditions of Chongqing were better than those of Chengdu, and Chengdu was affected by regional pollution transmission.

**Key words:** O<sub>2</sub> pollution; influencing factors; transmission track; precursor emission level; O<sub>3</sub> formation potential

近年来,我国大气  $PM_{2.5}$ (空气动力学直径 < 2.5  $\mu$ m 的颗粒物)浓度水平显著下降,夏季  $O_3$ 污染在城市环境空气质量中受到了广泛的关注 $[1^{-3}]$ . 对流层  $O_3$  作为对人体健康有害的空气污染物[4],在大气中由其关键前体物氮氧化物 $(NO_x)$ 和 VOCs 通过复杂的和非线性的光化学反应产生[5]. 成渝城市群位于四川盆地,四面环山,夏秋季发生高温干旱、降雨少、静风及弱风的天气概率较高[6],大气边界层稳定度高于同纬度地区,污染物不易扩散 $[7^{-9}]$ ,也是成渝地区  $O_3$ 污染高发季节, $O_3$ 呈现区域性污染的态势 $[10^{-12}]$ ,因此成都和重庆是我国大气污染备受关注的地区之一.

影响 0,浓度既有内因本地排放源,也有外因不

利的气象条件,常利用物理-化学模型和监测数据研究 O<sub>3</sub>污染成因. Li等<sup>[13]</sup>利用观测数据结合全球气候-化学传输模型(GEOS-Chem)模拟发现甲醛光解产生的自由基是驱动华北冬季 2月 O<sub>3</sub>快速上升的主要机制; Han等<sup>[14]</sup>利用气相-色谱质谱(GC-MS)和质子转移反应质谱仪(PTR-MS)监测发现珠三角地区含氧挥发性有机物(OVOCs)是 VOCs 中体积分数较高的物

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种,对本地O3生成贡献最大,甲醇为OVOCs中的主要 成分; Huang等[15]利用后向轨迹模型(Hysplit)计算高 空气团传输轨迹,当存在高空干净气团且长距离传 输时,利于污染物的扩散,当以短距离传输为主时, 加重 O,污染程度;Tan等[16]为了解成都 VOCs的来源, 利用苏玛罐采样和GC-MS离线分析28个不同类型的 站点,结果表明成都 VOCs 主要来源于机动车尾气和 溶剂源,而夏季生物源排放对O,的贡献也相对较高; 徐锟等[17]利用长时间序列的气象监测数据发现,成 都夏季高温、低湿、强辐射气象条件下利于 0,的生 成;曹庭伟等[11]利用国控点监测数据对比分析川渝 15个城市,春末和夏季0,污染较严重,呈区域性污 染,且边界层较低的区域极易产生0,污染;李陵等[18] 发现在重庆 O<sub>3</sub>污染期间 OVOCs 占比最高,体积分数 靠前物种是甲醛、乙烯和丙酮,三者之和占比超过 30%.

2022年8月成都和重庆呈现显著的O<sub>3</sub>污染差异,8月成都超标20d,重庆未一天超标(https://air.cnemc.cn:18007/).本文利用O<sub>3</sub>污染时空特征、前体物浓度水平、大气化学活性、O<sub>3</sub>生成潜势、近地面气象条件和高空气团传输轨迹及污染源潜在源区,分析成都和重庆O<sub>3</sub>污染差异的特征及其关键驱动因素,以期为O<sub>3</sub>污染科学防控提供决策参考.

### 1 材料与方法

#### 1.1 采样地点

重庆采样地点位于重庆渝北区龙山小学教学楼楼顶科研观测站(29.59°N,104.05°E),采样口离地面约25 m;成都采样地点位于成都青羊区成都环科院楼顶超级监测站(30.65°N,104.05°E),采样口离地面约25 m.两个站点周围无明显建筑遮挡物,无规模以上企业等,均位于集交通干道、商业和居民混合区,是一个典型的城市站点.采样时间为2022年8月1日至8月31日.

#### 1.2 仪器设备信息及数据分析

在线 VOCs 数据获取:重庆利用杭州谱育公司生产的 EXPEC 2000 获得 116 种物质(本文仅使用其中52 种物种和成都进行对比分析)[19.20],成都利用荷兰 SYNSPRC 公司生产的 GC955-611/811 获得 52 种物种[21,22],包含 26 种烷烃、16 种芳香烃和 10 种烯烃.重庆仪器的检测原理为 GC-FID/MS(气相色谱-火焰离子化检测器/质谱检测器),测量原理是样品气通过预处理浓缩富集,进入 GC 分离后通过 MS或 FID 进行测量,低碳( $C_2 \sim C_5$ ) VOCs 样品使用 FID 检测,其余组分,比如高碳( $C_6 \sim C_{12}$ )和含氧 VOCs 使用 MS 检测.成都 GC955-611/811 仪器采用了双通道检测系统,低碳烃

 $(C_2 \sim C_5)$ 使用 FID 检测,高碳烃 $(C_6 \sim C_{12})$ 使用光电离检测器(PID)进行检测.

重庆和成都的空气质量数据包括 NO<sub>2</sub>(二氧化氮)和 O<sub>3</sub>分别由美国赛默飞热电公司 i 系列气体监测仪 42i和 49i测得. 气象数据包括温度、湿度、风向和风速,重庆由 Lufft WS502-WTB100气象仪获得,成都由 WS600-UMB气象仪获得. 边界层高度数据重庆由安徽蓝盾光电子公司生产的 LGO-01 获得<sup>[23,24]</sup>,该仪器基于差分吸收原理,根据气溶胶粒子浓度变化的快慢,反映在激光雷达回波信号上,即回波信号的快速衰减,通过查找衰减最快的高度,得出大气边界层高度;成都边界层数据由北京恰孚和融公司生产的EV-LIDAR-CAM 获得.

#### 1.3 质量控制与质量保证(QA/QC)

为保证数据准确性、有效性,观测期间采取严格的质量保证和质量控制措施,参考 HJ1010-2018(环境空气挥发性有机物气相色谱连续监测系统技术要求及检测方法)中质控单元执行,利用 Spectra/Linde标准气体进行 PAMS标样每周一次的空白和单点校准,80%以上的物质偏差在 20%以内,经 AQMS-200动态气体校准仪,利用高纯氮气将  $1 \times 10^6$ 标气稀释成摩尔分数依次为 0.5、2.4、6.8 和 10 nmol·mol·的标准气体进样,每个浓度分析 3 次来校准仪器,校准相关系数均大于 0.99. 热电仪器通过零气和标准气体校准,采样期间零点漂移和跨度漂移测试结果范围均为 $\pm 2\%$ 之间.  $0_3$  激光雷达也是进行零点校准,调整底噪和背景噪声平整度,使拟合斜率  $\leq 10^{-5}$ ,计算饱和度或线性度,使得  $R^2 \geq 95\%$ .

#### 1.4 ·OH损耗速率和O3生成潜势

利用·OH损耗速率(·OH loss rate,  $L_{.OH}$ )量化污染物在大气中的化学反应活性[25.26]:

$$L_{\text{OH},i} = k_{\text{OH},i} \times [\text{VOC}]_i$$

式中, $L_{\text{OH.i}}$ 表示物种i的·OH损耗速率, $s^{-1}$ ; $k_{\text{OH.i}}$ 表示物种i与·OH的反应速率常数<sup>[27]</sup>, $cm^{3}$ ·(molecule·s)<sup>-1</sup>;[VOC],表示 VOC 物种i的环境浓度,molecule· $cm^{-3}$ .

利用 VOCs 最大增量反应活性(MIR)量化 VOCs 的 O<sub>3</sub>生成潜势(OFP)<sup>[25,26]</sup>:

$$OFP_i = MIR_i \times [VOC]_i$$

式中,OFP<sub>i</sub>表示物种i的 O<sub>3</sub>生成最大潜势,MIR 表示物种i的最大增量反应活性<sup>[28]</sup>, $g \cdot g^{-1}$ (以 O<sub>3</sub>/VOC 计), [VOCs],单位为×10<sup>-9</sup>.

1.5 后向气团轨迹模型(Hysplit)和潜在来源贡献 因子法(PSCF)

利用 Hysplit<sup>[29]</sup>官 网上(https://www. ready. noaa. gov/HYSPLIT. php)公布的气团轨迹数据,基于 Wang 等<sup>[30,31]</sup>开发 MeteoinfoMap 软件中的 TrajStat 工具,采用

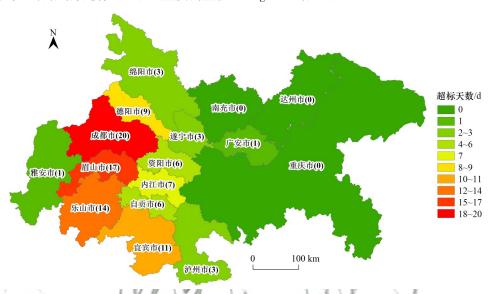
角距离轨迹聚类方法,模拟计算采样期间典型时刻 (00:00、03:00、06:00、09:00、12:00、15:00、18:00 和 21:00)实时混合边界层中间高度的 48 h 气团后向轨迹.再利用 PSCF 模型<sup>[30,32]</sup>计算 O<sub>3</sub>潜在源贡献分析,该模型已广泛应用于识别监测点位大气污染物高浓度时所对应的潜在来源区域<sup>[33-36]</sup>; PSCF 值越大,表明该网格区域对点位污染物浓度的影响越大.

#### 2 结果与讨论

#### 2.1 0,污染时空特征

从空间分布上看,成都超标20d(1d重度),重庆

未一天超标.四川省  $O_3$ 污染以成都为辐射中心,其中川南  $O_3$ 污染较重,如眉山超标 17 d(3 d + pe)、乐山超标 14 d(1 d + pe),而紧挨重庆的川东北和川东南区县,除达州和南充未一天超标,其余区县均超标 17 d(图 1).从时间上看,观测期间成都 17 d(图 1).



括号中的数字表示超标天数 1 2022年8月成渝城市群超标天数空间分布

Fig. 1 Spatial distribution of days exceeding standard in Chengdu-Chongqing urban agglomeration (CCUA) in August 2022

H 73 -6-4															20	22年8	月														
成市区县	1日	2日	3日	4日	5日	6日	7日	8日	9日	10日	11日	12日	13日	14日	15日	16日	17日	18日	19日	20日	21日	22日	23日	24日	25日	26日	27日	28日	29日	30日	31
重庆	140	147	141	136	142	119	113	139	119	127	112	106	116	129	143	128	106	107	131	121	100	132	114	107	133	114	100	101	102	64	64
成都	121	138	160	133	170	132	174	188	196	189	184	198	192	186	204	201	208	195	164	182	171	189	216	161	101	175	138	77	69	73	106
自贡	168	140	159	148	131	120	132	150	163	137	112	116	139	154	184	184	147	153	205	155	114	142	158	112	156	130	129	164	60	75	96
沙州	167	152	143	130	132	125	120	147	135	142	97	107	122	141	189	146	127	125	172	133	104	140	135	108	117	128	108	144	83	76	100
德阳	103	155	129	95	122	125	142	166	147	202	223	166	176	159	145	149	165	171	138	201	174	142	156	126	84	134	114	75	68	99	94
绵阳	130	147	123	80	91	128	131	153	150	174	186	145	142	142	124	148	152	155	126	170	154	134	147	155	82	109	122	63	61	91	86
遂宁	142	152	149	135	146	129	130	136	140	142	106	131	134	133	166	185	139	143	148	169	97	135	153	118	129	131	120	106	65	74	79
内江	172	157	170	138	141	140	146	158	170	148	114	128	155	142	207	185	142	144	210	165	120	140	156	119	152	134	120	143	60	83	90
乐山	103	140	140	130	122	119	144	159	170	147	148	154	166	174	170	175	205	184	155	170	176	174	201	246	197	142	161	91	65	57	74
南充	115	139	136	132	128	104	134	137	128	143	119	127	127	120	137	148	107	134	134	150	112	104	110	99	120	119	117	100	68	77	75
眉山	102	135	132	129	153	138	152	165	175	143	158	163	173	190	196	190	233	221	205	173	191	197	200	220	173	157	184	75	70	67	91
宜宾	166	131	142	107	132	115	137	157	169	141	131	134	153	173	168	176	176	179	162	160	125	167	176	131	176	149	154	156	69	77	92
广安	136	176	139	138	140	126	131	148	124	126	125	114	138	114	142	128	118	124	125	129	116	116	106	115	147	147	117	120	70	60	67
达州	107	104	106	114	151	124	106	106	101	100	106	92	96	88	159	98	94	88	111	101	97	98	80	104	119	118	86	79	53	52	52
雅安	77	100	133	98	132	140	144	152	121	137	141	142	152	158	152	160	142	144	94	139	136	173	144	157	115	141	103	63	63	41	46
资阳	172	130	150	144	142	140	143	153	152	168	138	142	158	138	146	182	160	158	175	174	133	135	168	142	135	143	155	109	68	84	107

数值单位为μg·m<sup>-3</sup>

#### 图 2 2022年8月成渝城市群 O<sub>3</sub>-8h污染特征

Fig. 2 Pollution characteristics of O<sub>3</sub>-8h in CCUA in August 2022

#### 2.2 VOCs 变化特征

#### 2.2.1 VOCs体积分数

表1总结了观测期间重庆和成都污染物的浓度

水平,成都 52 种 VOCs 的总体积分数  $(18.8 \times 10^{-9})$  明显高于重庆  $(6.6 \times 10^{-9})$ . 烯烃和芳香烃是大气化学活性较高的物种 [26], 而成都烯烃  $(2.6 \times 10^{-9})$ 和芳香

MF

烃(3.0×10°)分别是重庆的1.9倍和2.7倍;烷烃来源复杂且大部分活性不高[37],成都烷烃(13.2×10°)是重庆的3.2倍.成都VOCs各组分从高到低排序为烷烃(70.3%)、芳香烃(15.9%)和烯烃(13.8%),体积分数排名前3为乙烷(4.4×10°)、丙烷(2.8×10°)和正丁烷(1.6×10°),累计占比为39.4%;重庆和成都组分占比相似,占比最大均为烷烃(61.8%),其次烯烃(21.2%)和芳香烃(17.0%),排名前3为乙烷(0.9×10°)、异戊烷(0.7×10°)和正丁烷(0.5×10°),累计占比为49.3%。由此可见,成都和重庆高

体积分数组分主要以低碳数的 VOCs 为主,  $C_2 \sim C_5$ 的 烷烃主要来源于机动车尾气和汽油蒸发 [37.38],且观测期间成渝总 VOCs 的时序趋势和峰值时间与 NO<sub>2</sub>相似 (图 3),机动车尾气影响显著.异戊二烯通常作为生物源示踪物 [39],8月1~13日重庆异戊二烯  $(0.6 \times 10^{-9})$  较成都高  $(0.4 \times 10^{-9})$ ,但8月14~24日重庆异戊二烯浓度较8月14日前显著下降,约70.9%,而成都异戊二烯浓度上升约31.5%,8月14~24日成都异戊二烯是重庆的1.4倍(图3);生物源活性差异可能也是成渝两地 $O_3$ 污染差异成因之一.

#### 表 1 观测期间重庆和成都 O<sub>3</sub>和 NO<sub>2</sub>浓度水平以及 VOCs 体积分数

Table 1 Concentration levels of O<sub>3</sub> and NO<sub>2</sub> and the mixing ratio of VOCs in Chongqing and Chengdu during the observation period

位置	ρ(O <sub>3</sub> ) /μg·m <sup>3</sup>	$\rho(\mathrm{NO_2})$ / $\mu \mathrm{g \cdot m}^3$	φ(烷烃)×10 <sup>-9</sup>	φ(烯烃)×10 <sup>-9</sup>	φ(芳香烃)×10 <sup>-9</sup>	φ(异戊二烯) ×10 <sup>-9</sup>
重庆	81.1±24.7	16.9±10.3	4.1±3.1	1.4±0.8	1.1±0.6	0.5±0.2
成都	111.4±42.0	23.9±9.2	13.2±5.0	2.6±0.7	$3.0 \pm 1.4$	$0.4 \pm 0.2$

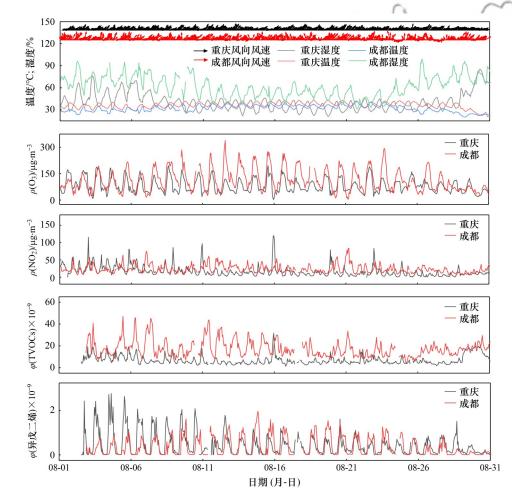


图 3 观测期间重庆和成都的温湿度、风速风向、 $O_3$ 、 $NO_2$ 、TVOCs和异戊二烯时间序列变化

Fig. 3 Temperature and humidity, wind speed and direction,  $\rm O_3$ ,  $\rm NO_2$ , TVOCs and isoprene time series in Chongqing and Chengdu during the observation period

#### 2.2.2 日变化规律

图 4 比较了重庆和成都 O<sub>3</sub>、NO<sub>2</sub>和 VOCs 各组分的日变化趋势,成渝两地 O<sub>3</sub>均呈现单峰态势,NO<sub>2</sub>呈

双峰态势.从VOCs各组分上看,成都呈现夜间高、 日间低和双峰的特征,表明夜间存在一次源,而日间 低是受到大气氧化反应的影响.而重庆VOCs在 08:00~10:00有明显的峰值,与NO<sub>2</sub>日变化时间同步,是交通源所致;重庆夜间NO<sub>2</sub>浓度峰值较成都高,也是日最大值,这可能与夜间大货车运行有关. 芳香烃作为主要的人为源非甲烷挥发性有机物(NMHCs),成都芳香烃在04:00~12:00维持在较高水平,除早高峰交通源贡献外,还受到了溶剂使用源的影响,这与Tan等[16]的研究结果相似,而在重庆主要是来源于机动车尾气[18]. 异戊二烯主要由生物源在日间排放,在10:00~11:00排放速率达到最高,迅速升高接近日最高值,与O<sub>3</sub>的日变化趋势相似.

#### 2.2.3 L.OH和 OFP

成都总 OFP 为  $51.2 \times 10^{\circ}$ ,其中烷烃、烯烃和芳香烃的 OFP 分别为  $10.9 \times 10^{\circ}$ 、 $24.6 \times 10^{\circ}$  和  $15.7 \times 10^{\circ}$ ;总  $L_{.oH}$  为  $3.9 \, \mathrm{s}^{-1}$ ,烷烃、烯烃和芳香烃的  $L_{.oH}$  分别为  $0.5 \times 2.5$  和  $0.9 \, \mathrm{s}^{-1}$ . 重庆总 OFP 为  $25.0 \times 10^{\circ}$ ,烷烃、烯烃和芳香烃的 OFP 分别为  $4.4 \times 10^{\circ}$ 、 $15.2 \times 10^{\circ}$  和  $5.4 \times 10^{\circ}$ ;总  $L_{.OH}$  为  $2.3 \, \mathrm{s}^{-1}$ ,烷烃、烯烃和芳香烃的  $L_{.OH}$  分别为  $0.2 \times 1.8$  和  $0.3 \, \mathrm{s}^{-1}$ . 对比结果显示成都的大气光化学活性更强, $0_3$ 生成潜势也更高;OFP显示成都烯烃和苯系物的占比分别达 48.1% 和 30.1%,而重庆仅烯烃就占比 60.7%.

从图 5 可知,成都 $L_{.OH}$ 、OFP 和 VOCs 体积分数排放前 10 的物种都包括乙烯、甲苯、间/对-二甲苯和1-己烯,而重庆都包括异戊二烯、乙烯、甲苯和间/对-二甲苯.成都和重庆 $L_{.OH}$ 排名第 1 均为异戊二烯,成都异戊二烯(1.0 s<sup>-1</sup>)的 $L_{.OH}$ 占比为 26.0%,而重庆仅异戊二烯(1.2 s<sup>-1</sup>)的 $L_{.OH}$ 就占比 50.9%.成都 $L_{.OH}$ 排名前 10 中,7种烯烃累计占比为 59.3%,3种芳香烃

累计占比为 15.6%. 重庆  $L_{.OH}$  排名前 10 中,6种烯烃 累计占比为 74.5%,3种芳香烃累计占比为 8.3%和1种烷烃占比为 1.4%. 成都 OFP前 3(乙烯、间/对-二甲苯和异戊二烯)累计占比为 37.8%,重庆 OFP前 3(异戊二烯、乙烯和丙烯)累计占比为 44.5%. 与  $L_{.OH}$  形成对比的是,成都 OFP前 10 中,6 中烯烃累计占比为 42.9%,3 种芳香烃累计占比为 24.2%和 1 种烷烃占比为 3.8%;重庆 OFP前 10 种,5 种烯烃累计占比为 56.0%,3 种芳香烃累计占比为 16.1%和 2 种烷烃累计占比为 6.5%,表明烯烃在成渝两地  $L_{.OH}$ 和 OFP占主导地位。综上,成渝应重点管控烯烃排放源,但芳香烃对成都  $O_3$ 生成的影响也不可忽略,芳香烃不仅对  $O_3$ 生成潜势大,也对二次气溶胶的生成有着较强的优势 [40].

#### 2.2.4 特征比值

利用特征比值法初步分析成渝大气 VOCs来源差异.成都 C<sub>3</sub> ~ C<sub>6</sub>烷烃之间的相关性较好(r²为0.6~0.9),丙烷和丁烷是液化石油气(LPG)的主要成分,也是成都体积分数较高的物种,成都丙烷/异丁烷(2.9)和丙烷/正丁烷(1.9)与 LPG 排放比(分别为3~7和2~4)相似[44],而重庆分别为1.4和0.7,表明成都液化石油气使用率高于重庆.当甲苯/苯(T/B)小于1时,燃煤或生物质影响更大[41];T/B小于2,机动车尾气为主要贡献源;T/B大于5,被认为受工业、溶剂源等影响明显.异戊烷/正戊烷(I/N)在0.56~0.80之间时,燃煤贡献占主导[42];I/N在2.2~3.8之间时机动车尾气占主导;I/N在1.8~4.6之间时溶剂源占主导[43,44].重庆和成都T/B分别为1.7和2.7,这表明重

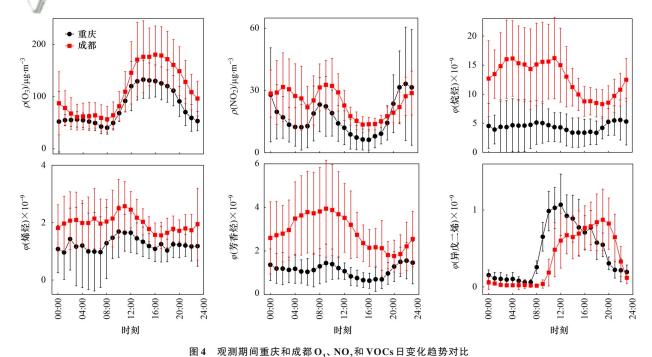


Fig. 4 Comparison of diurnal variation trends of O<sub>3</sub>, NO<sub>2</sub> and VOCs between Chongqing and Chengdu during the observation period

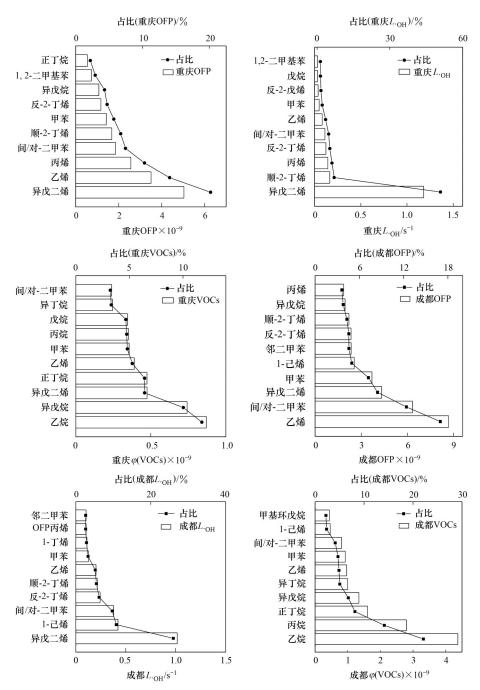


图 5 观测期间重庆和成都 OFP、 $L_{.OH}$ 和 VOCs 体积分数排名前 10 位及其占比的物种

Fig 5  $\,$  Top 10 OFP,  $L_{\rm oH}$  and mixing ratios of VOCs and their percentages in Chongqing and Chengdu during the observation period

庆主要受机动车尾气的影响大;重庆I/N为2.5,成都I/N为5.3,表明成都受机动车尾气和溶剂源的共同影响.

#### 2.3 近地面气象条件和高空气团传输

表 2 是观测期间成都和重庆的气象参数,重庆温度高于成都,湿度较成都低,可能会影响生物源 VOCs的排放  $^{[45]}$ .  $O_3$ 与温度呈非线性关系,温度是影响对流层瞬时  $O_3$ 生成速率多重因素中的主要天气变量  $^{[46-48]}$ . 因  $O_3$ 仅在白天光化学反应生成,选取 07:00~19:00的数据  $^{[15]}$ ,利用非线性拟合发现随温度上升、湿度下降(重庆  $^2$  = 0.88,成都  $^2$  = 0.75,图 6), $O_3$ 

浓度明显呈先升后降的趋势. 当重庆( $r^2 = 0.49$ )温度超过 39.1°C和湿度低于 39.3%时  $O_3$ 浓度开始下降; 当成都 ( $r^2 = 0.70$ )温度超过 37.3°C和湿度低于 48.4%时  $O_3$ 浓度开始下降. 8月 14~24日成渝两地异戊二烯浓度存在差异,期间成都异戊二烯呈上升趋势,而重庆异戊二烯呈下降趋势(图 3),生物源排放活性与温湿度存在响应关系 [45]. 对比此期间成渝两地的温湿度发现,期间成都温度 34.9°C,湿度 51.8%,未达到成都  $O_3$ 浓度下降的温湿度环境;重庆温度 38.3°C,湿度 33.1%,接近重庆  $O_3$ 浓度下降的温湿度环境. 因此,8月成渝生物源活性存在差异可能

是由不同的温湿度环境所造成,温湿度也是影响成渝 O<sub>3</sub>污染存在差异的成因之一.

重庆边界层高度(2.2 km)是成都(0.7 km)的3.1倍,这表明重庆的垂直大气扩散条件较成都好(表2).利用 HYSPLIT和 PSCF 计算高空气团传输轨迹和

#### 表 2 观测期间成都和重庆的气象数据

Table 2 Meteorological data of Chengdu and Chongqing during the observation period

位置	温度/℃	湿度/%	风速/m·s <sup>-1</sup>	边界层/km
重庆	35.9±4.0	41.1±11.7	$0.7 \pm 0.2$	2.2±0.7
成都	31.7±4.3	61.8±12.6	1.2±0.8	$0.7\pm0.3$

O<sub>3</sub>潜在源贡献水平(图 7),总体来看,重庆水平大气扩散条件较成都好.观测期间重庆以中长距离传输为主,来自贵州和云南的气团占比约 57.0%,上风向大气污染源分布较少,大气扩散条件较好;PSCF显示O<sub>3</sub>高值点出现在綦江和巴南方向的轨迹上(24.4%).成都以短距离传输为主(58.1%),轨迹较短,且"回旋"轨迹占比 8.9%,PSCF显示O<sub>3</sub>高值点主要出现在川南气团(42.0%)和德阳(16.7%)回旋气团上,成都气团传输轨迹与川渝O<sub>3</sub>空间分布的结果相似(图 1),成都上风向的川南城市群和成都平原城市群大气污染源

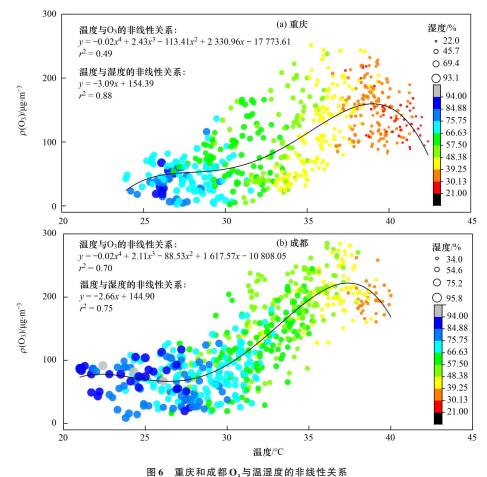


Fig. 6 Nonlinear relationship between O<sub>3</sub> and temperature and humidity in Chongqing and Chengdu

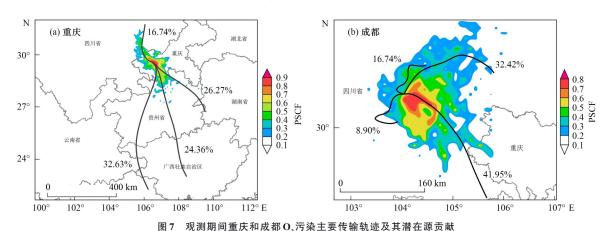


Fig. 7 Main transmission trajectories and potential contributions of O3 pollution over Chongqing and Chengdu during the observation period

分布比较密集<sup>[33]</sup>,致使成都周边存在 O<sub>3</sub>区域性污染的态势.因此,重庆较成都垂直和水平大气扩散条件好,是导致夏季成渝两地 O<sub>3</sub>污染差异的成因之一.

#### 3 结论

- (1)观测期间成都 VOCs 的总体积分数(18.8×10°)明显高于重庆(6.6×10°),成都和重庆高体积分数物种均以低碳数 VOCs 为主,优势组分也均为烷烃;成都体积分数前 3 为乙烷、丙烷和正丁烷,累计占比为 39.4%;重庆体积分数排名前 3 为乙烷、异戊烷和正丁烷,累计占比为 49.3%.
- (2)成都总 OFP(51.2 × 10°)是重庆(25.0 × 10°)的2.0倍,  $L_{.OH}(3.9\,\mathrm{s}^{-1})$ 是重庆(2.3 s<sup>-1</sup>)的1.7倍,表明成都的大气光化学活性更强, $O_3$ 生成潜势也更高.成渝两地 $L_{.OH}$ 排名第1均为异戊二烯,成都OFP前3是乙烯、间/对-二甲苯和异戊二烯,重庆前3是异戊二烯、乙烯和丙烯,表明夏季 $O_3$ 污染生物源贡献显著,烯烃是两地 $O_3$ 污染主要贡献源,但芳香烃对成都的影响也不可忽略.此外,成都 $C_3 \sim C_6$ 烷烃之间的相关性较好,丙烷/丁烷的比值与LPG排放比相似,且烷烃日变化峰值与 $NO_2$ 同步,表明成都VOCs受机动车尾气的影响显著.T/B和I/N显示重庆主要受机动车的影响,而成都受机动车和溶剂源的共同影响。
- (3)气象条件的差异导致成渝两地生物源活性不同,8月14~24日重庆平均温度38.3℃使得生物源排放活性有所下降.重庆边界层高度(2.2 km)是成都(0.7 km)的3.1倍;重庆高空气团以中长距离传输为主,成都以短距离传输为主,轨迹较短且有"回旋",总体上看,重庆水平和垂直大气扩散条件均较成都好.

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