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成都市冬季相对湿度对颗粒物浓度和大气能见度的影响

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摘要: 利用成都市城区 2015 年 12 月的连续在线观测数据, 如相对湿度(RH)、能见度、颗粒物(PM₁₀、PM_{2.5}和PM₁)浓度、气态污染物(SO₂和NO₂)浓度以及PM_{2.5}中SO₄²⁻和NO₃⁻浓度, 探讨RH对颗粒物浓度和大气能见度的影响。结果表明, 高颗粒物浓度和高RH协同作用导致低能见度事件。观测阶段, PM_{2.5}在PM₁₀中的平均比重为64%, 表明成都市冬季细颗粒物污染严重; 随着RH增加, PM_{2.5}/PM₁₀显著增加, 表明高RH会加重细颗粒物污染。随着PM_{2.5}浓度增加, 能见度呈幂指数下降; 在相同PM_{2.5}浓度下, RH越高, 能见度越低。当颗粒物浓度较低时, RH对能见度的影响作用较强; 当颗粒物浓度较高时, 大气消光主要由PM_{2.5}浓度控制, RH对能见度的影响减弱。当RH大于70%时, 硫酸化率(SOR)和氮氧化率(NOR)的均值分别从0.27和0.11(RH小于40%)增长至0.40和0.19, 表明较高RH对二次硫酸盐和硝酸盐的生成有显著的促进作用, 二次硫酸盐和硝酸盐单独或协同影响空气质量。

关键词: 相对湿度; 能见度; 颗粒物; 二次无机盐; 成都

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Effect of Relative Humidity on Particulate Matter Concentration and Visibility During Winter in Chengdu

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Abstract: The effect of relative humidity (RH) on particulate matter concentrations and atmosphere visibility were investigated using the continuous on-line observed data of Chengdu city during December 2015, including RH, visibility, the concentrations of particulate matters (PM₁₀, PM_{2.5} and PM₁) and gaseous pollutants (SO₂ and NO₂), and the concentrations of SO₄²⁻ and NO₃⁻ in PM_{2.5}. The results showed that the haze process occurred because of the synergistic effects of higher particulate matter concentrations and RH, leading to the reduction of visibility. The average ratio of PM_{2.5} to PM₁₀ was 64% and it significantly increased with the increase of RH during observation period, which indicated that the pollution of fine particles during winter in Chengdu was serious, and high RH aggravated the pollution caused by fine particles. Visibility decreased exponentially with the increase of particulate matter concentrations. When RH was higher, visibility was lower at the same concentrations of particulate matter. RH had a strong effect on visibility at lower particle concentrations, while the effect of RH on the visibility decreased, and atmospheric extinction was controlled by PM_{2.5} concentrations at higher particle concentrations. With RH increasing from less than 40% to more than 70%, the average sulfur oxidation ratio (SOR) and nitrogen oxidation ratio (NOR) increased from 0.27 and 0.11 to 0.40 and 0.19, respectively, indicating that higher RH significantly promoted the formation of secondary sulfate and nitrate. Secondary sulfate and nitrate separately or coordinatively influenced the air quality.

Key words: relative humidity(RH); visibility; particulate matter; secondary inorganic salt; Chengdu

近年来的秋冬季节, 空气重污染事件在全国范围内频繁出现, 其中京津冀、长江三角洲、珠江三角洲和四川盆地地区为霾高发区, 以高颗粒物浓度和低能见度为特征的重污染事件已经成为科研工作者和民生关注的焦点^[1~3]。大气能见度是反映大气透明度的指标, 也是反映环境空气质量的视觉指标, 相对湿度(RH)和颗粒物浓度是影响大气能见度的主要影响因素。于兴娜等^[4]分析了南京RH和气溶胶组分对大气能见度下降的影响, 研究结果显示高气溶胶浓度和RH是造成南京市低能见度事件的两个重要因素, 当RH>80%时, 颗粒物吸湿增

长对大气能见度下降有更显著的作用。

RH是颗粒物理化和光学特性的重要影响因素, 会影响颗粒物的吸湿增长、粒径分布、气态前体物的气粒转化等特性^[5~7]。当环境中RH较高时, 颗粒物中硫酸盐、硝酸盐和铵盐等具有吸湿特性的成分会吸湿增长, 粒径变大, 消光能力显著增强, 导致能见度降低^[8, 9]。此外, RH通过影响颗粒

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物表面液相非均相反应影响无机气溶胶二次生成, RH 升高能促进颗粒物表面二次无机组分(硫酸盐、硝酸盐和铵盐)的积累,导致颗粒物浓度增加^[10]. Sun 等^[11]对北京市霾期间颗粒物特征进行分析,发现 RH 与硫酸化率(SOR)和氮氧化率(NOR)显著相关,相关性系数分别为 0.96 和 0.88,表明较高的 RH 可以促进硫酸盐和硝酸盐二次转化过程. Wang 等^[12]对四川盆地气溶胶消光性质进行研究,发现成都市 2015 年冬季硫酸铵和硝酸铵对消光的贡献超过 60%. 可见, RH 对颗粒物浓度升高、二次无机盐生成和能见度下降有重要影响.

成都作为西南地区最大的以高湿度为特征的城市,属于亚热带湿润季风气候区,雨量充沛,典型的气象特征是高湿度低风速,近几年,成都平原城市地区发生多次大面积较长时间区域性霾污染事件^[13, 14]. 然而,成都市的高 RH 对颗粒物浓度和二次无机盐生成以及大气能见度的影响尚未见报道. 鉴于此,本研究拟利用成都市城区 2015 年 12 月的连续在线观测数据,如 RH、大气能见度、颗粒物(PM₁₀、PM_{2.5}和 PM₁)浓度、气态污染物(SO₂和 NO₂)浓度以及 PM_{2.5}中 SO₄²⁻和 NO₃⁻浓度等,探讨 RH 对颗粒物浓度和二次无机盐生成以及大气能见度的影响,以期为进一步改善成都地区大气能见度提供理论支撑.

1 材料与方法

1.1 观测地点及时间

冬季霾天气过程频发,因此,本次观测时间定为 2015 年 12 月. 观测点位于成都市青羊区成都市环境保护科学研究院楼顶(7 楼,距离地面 21 m, 30°39'N, 104°02'E),靠近锦江,四周 2 km 内无高大建筑物,视野开阔、气流稳定,附近 5 km 范围内无明显工业大气污染源,属于典型的居住区. 研究表明,该观测点在一定程度上可代表成都市城区的大气污染水平^[15].

1.2 观测仪器

PM₁₀、PM_{2.5}和 PM₁均采用微量振荡天平法

(Thermo TEOM Series 1405, USA)在线、实时分析. 气态污染物 SO₂和 NO₂的体积浓度分别由脉冲荧光 SO₂分析仪(Thermo 43i, USA)和化学发光 NO-NO₂-NO_x分析仪(Thermo 42i, USA)进行监测. 采用基于旋转湿式扩散管气体吸收技术和气溶胶蒸汽捕集技术的在线气体与气溶胶成分监测仪(Fortelice, IGAC S-611EG, 中国台湾)与离子色谱仪(DIONEX ICS-2100 & ICS-1100)联用来测定 PM_{2.5}中水溶性离子组分 SO₄²⁻和 NO₃⁻,离子交换色谱柱型号分别为 IonPac AS18 和 IonPac CS12A. 气象要素(大气能见度、RH、温度和风速)采用德国 LUFFT WS600 一体式气象站进行监测. 所有监测数据经过异常值筛选和剔除后统一处理为小时均值.

2 结果与讨论

2.1 RH 与颗粒物浓度及大气能见度的总体特征

整个观测期间,基本无降雨事件,风速偏低,平均风速为(0.95 ± 0.37) m·s⁻¹,平均气温为(9.87 ± 2.35) °C. 观测期间,成都市的 RH 和大气能见度、各粒径段颗粒物质量浓度及其与 PM₁₀比值的逐时变化如图 1 所示,其中,PM_{1~2.5}和 PM_{2.5~10}分别指的是空气动力学当量直径在 1 ~ 2.5 μm 和 2.5 ~ 10 μm 的颗粒物. 可以看出,当 RH 较低时,大气能见度较高, RH 和大气能见度的负相关关系明显,尤其是观测后期,随 RH 增大,颗粒物浓度持续升高,大气能见度骤降.

表 1 列出了观测期间 RH、大气能见度、颗粒物质量浓度及其与 PM₁₀质量浓度比值的统计结果. 平均 RH 为(66.53 ± 13.12)%,平均大气能见度为(5.89 ± 3.47) km, PM_{2.5}平均浓度为(103.33 ± 75.46) μg·m⁻³, PM_{2.5~10}、PM_{1~2.5}和 PM₁在 PM₁₀中的比值分别为 0.36、0.24 和 0.40. 总体来说,成都市冬季颗粒物浓度较高,其中亚微米颗粒物(即 PM₁)对 PM₁₀质量浓度贡献最大,说明成都市冬季气溶胶中细颗粒物污染较为严重.

表 1 观测期间 RH、能见度和颗粒物质量浓度及与 PM₁₀质量浓度比值的统计特征
Table 1 Statistical characteristics of RH, visibility, particulate mass concentrations, and their ratio to PM₁₀ concentration during the observation period

项目	能见度/km	RH /%	PM ₁₀ /μg·m ⁻³	PM _{2.5} /μg·m ⁻³	PM ₁ /μg·m ⁻³	PM _{2.5~10} /PM ₁₀	PM _{1~2.5} /PM ₁₀	PM ₁ /PM ₁₀
平均值	5.89	66.53	159.81	103.33	61.82	0.36	0.24	0.40
标准差	3.47	13.12	114.30	75.46	40.45	0.07	0.07	0.06
最大小时值	30.47	86.90	655.00	434.00	249.20	0.60	0.45	0.61
最小小时值	0.72	23.80	28.00	13.00	9.20	0.21	0.05	0.25

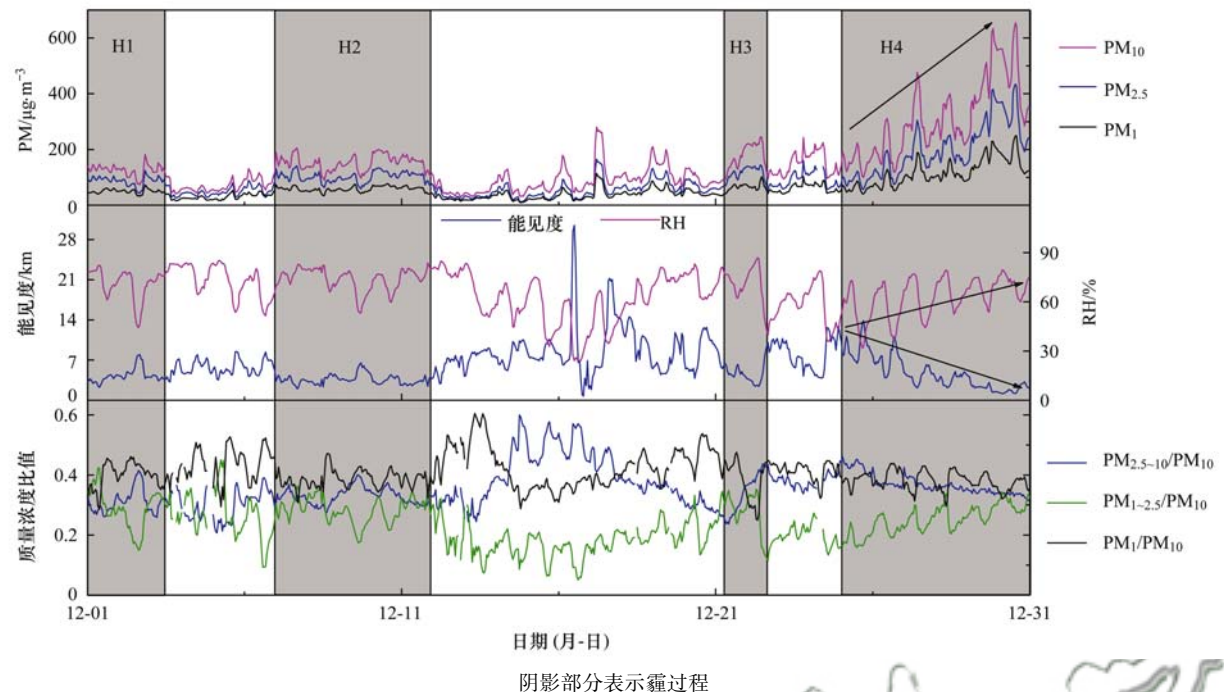


图1 观测期间 RH、能见度和颗粒物质量浓度及其与 PM₁₀ 质量浓度比值的逐时变化

Fig. 1 Hourly variations of RH, visibility, particulate mass concentrations, and their ratio to PM₁₀ concentration during the observation period

结合中国气象局对霾的定义(大气能见度 < 10.0 km, RH < 90%)和《环境空气质量标准》,观测期间发生4次时间较长且 PM_{2.5} 浓度超过环境空气质量二级浓度限值(75 μg·m⁻³)的霾天气过程,如图1中阴影部分所示,4次霾过程的持续时间、大气能见度、RH、颗粒物质量浓度及其与 PM₁₀ 质量浓度比值的均值如表2所示.霾过程持续时间最短为29 h(过程3),最长为168 h(过程4).

霾过程与非霾过程相比,有以下4个特征:①颗粒物浓度较高;②大气能见度较低;③PM_{1-2.5}/PM₁₀显著上升;④RH增大.如图1可知,

在霾过程4中,随RH从35%持续增大至75%,PM_{2.5}浓度从79 μg·m⁻³暴发式增长到观测期间的峰值434 μg·m⁻³,大气能见度也由10 km降至1.5 km左右.由表2可知,在4个霾过程中,霾过程2的平均RH最高,为73.92%,PM_{2.5}平均浓度为101.50 μg·m⁻³,平均能见度最低,仅为3.60 km;霾过程4的平均RH最低,为64.76%,PM_{2.5}平均浓度为204.51 μg·m⁻³,平均能见度为4.34 km.可以看出,RH的变化对霾的形成过程有重要影响,与其他霾过程研究结果一致,高RH和高颗粒物浓度协同对大气能见度降低以及霾过程形成起作用^[16,17].

表2 霾天气与非霾天气下 RH、能见度、颗粒物质量浓度及其与 PM₁₀ 质量浓度比值的均值
Table 2 Average data of RH, visibility, particulate mass concentrations, and their ratio to PM₁₀ mass concentration under the haze and non-haze episodes

项目	发生时间	能见度 /km	RH /%	PM ₁₀ /μg·m ⁻³	PM _{2.5} /μg·m ⁻³	PM ₁ /μg·m ⁻³	PM _{2.5-10} /PM ₁₀	PM _{1-2.5} /PM ₁₀	PM ₁ /PM ₁₀
1	12-01T00:00 ~ 12-03T11:00	4.28	71.03	125.75	87.03	49.91	0.31	0.29	0.40
2	12-06T23:00 ~ 12-11T23:00	3.60	73.95	151.68	101.50	58.26	0.33	0.28	0.39
霾过程	3	12-21T09:00 ~ 12-22T14:00	4.27	72.12	186.97	120.73	0.34	0.28	0.37
4	12-25T00:00 ~ 12-31T23:00	4.34	64.76	316.72	204.51	116.82	0.36	0.26	0.37
非霾		7.76	64.02	93.66	58.11	39.13	0.37	0.20	0.43

2.2 RH 对颗粒物浓度和能见度的影响

已有研究表明,能见度与气溶胶质量浓度及 RH 呈负相关关系^[18,19].观测期间,RH 变化幅度较大,RH 与能见度及各粒径段颗粒物与 PM₁₀ 质量

浓度比值的 关系如图2所示.随 RH 升高,PM₁/PM₁₀ 变化不大;RH 从 < 40% 增长至 80% ~ 90%,PM_{1-2.5}/PM₁₀ 增长了 146%,PM_{2.5-10}/PM₁₀ 降低了 36.2%.表明成都市 RH 越高,细颗粒物污染特征

越明显. 随着 RH 从 0 ~ 40% 升高到 80% ~ 90%, 能见度下降了 35.1%. 可以看出, RH 升高对能见度的恶化和细颗粒物污染加重影响很大^[20].

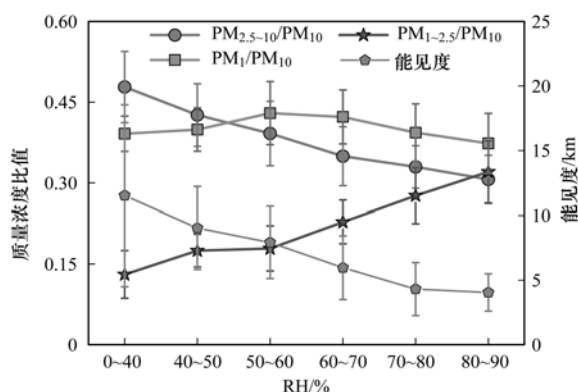


图2 RH 与能见度及不同粒径段颗粒物与 PM_{10} 质量浓度比值的关系

Fig. 2 Relationship between RH, visibility, and mass concentration ratio of particulate matters with different diameters to that of PM_{10}

图3给出了不同 RH 区间内 $PM_{2.5}$ 浓度与能见度的散点图及拟合曲线. 可以看出, 在各个 RH 范围内, 随着 $PM_{2.5}$ 浓度的增加, 能见度都呈幂指数下降, 这种下降趋势在 $PM_{2.5}$ 浓度较低时 ($< 75 \mu g \cdot m^{-3}$) 表现得尤为明显. 比如, RH 在 50% ~ 60% 时, $PM_{2.5}$ 从 $50 \mu g \cdot m^{-3}$ 增长到 $100 \mu g \cdot m^{-3}$, 能见度从 10 km 降低到 7 km, 能见度对于 $PM_{2.5}$ 增长的变化率 ($\Delta \text{能见度} / \Delta PM_{2.5}$) 为 $-0.06 \text{ km} / (\mu g \cdot m^{-3})$; $PM_{2.5}$ 从 $150 \mu g \cdot m^{-3}$ 增长到 $200 \mu g \cdot m^{-3}$, 能见度从 6 km 降低到 5 km, 能见度对于 $PM_{2.5}$ 增长的变化率为 $-0.02 \text{ km} / (\mu g \cdot m^{-3})$.

图3也显示, 在 $PM_{2.5}$ 浓度较低时, 较高的 RH 条件下能见度也会显著降低. 比如在 $PM_{2.5}$ 浓度为 $50 \mu g \cdot m^{-3}$ 的条件下, RH 为 50% ~ 60% 时, 能见度大约 11 km; RH 为 70% ~ 80% 时, 能见度大约 5 km. 这是由于 $PM_{2.5}$ 中的吸湿组分吸湿增长, 粒径增大, 显著增强了对光的散射作用, 导致能见度降低^[21,22]. 可见, 当颗粒物浓度较低时, 不同 RH 区间内能见度差异较大, RH 对能见度的影响较强; 当颗粒物浓度较高时, 不同 RH 区间内能见度差异不大, 说明此时大气消光主要由 $PM_{2.5}$ 浓度控制, RH 对能见度的影响减弱.

2.3 RH 对 $PM_{2.5}$ 中二次无机盐生成的影响

$PM_{2.5}$ 中含量最高的水溶性无机组分是硫酸盐和硝酸盐, 来源于直接排放和二次转化生成. 大气中 SO_2 和 NO_2 被羟基自由基或 O_3 等氧化剂经过一系列复杂的大气化学反应氧化生成硫酸和硝酸, 然

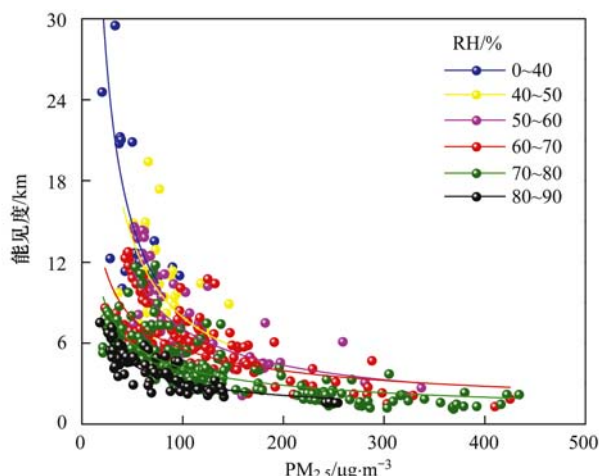


图3 $PM_{2.5}$ 浓度与能见度及 RH 的关系

Fig. 3 Relationship between $PM_{2.5}$ concentration, visibility, and RH

后和气态 NH_3 反应, 生成二次无机组分硫酸铵和硝酸铵^[23]. 通常用 SOR 和 NOR 分别作为 SO_2 和 NO_2 二次转化程度的指示参数^[24, 25]. SOR 和 NOR 定义为:

$$SOR = nSO_4^{2-} / (nSO_4^{2-} + nSO_2)$$

$$NOR = nNO_3^- / (nNO_3^- + nNO_2)$$

式中, n 代表物质的量^[26] (mol).

有研究指出, 当 SOR 的值大于 0.1 时, 就说明环境空气中的 SO_2 发生了一系列的光化学反应生成二次硫酸盐, 且 SOR 和 NOR 的数值越高, 代表 SO_2 和 NO_2 被氧化生成二次硫酸盐和硝酸盐越多^[27].

RH 是 SO_2 和 NO_2 在颗粒物表面进行非均相氧化反应过程的重要影响因素^[28]. 根据上式计算得到的 SOR、NOR 与 RH 的时间序列如图4所示, 总体来看, SOR 和 NOR 与 RH 基本呈现同升同降的变化趋势. 整个观测期间, SOR 和 NOR 的均值分别为 0.37 和 0.17. RH 小于 40% 时, SOR 和 NOR 的均值分别为 0.27 和 0.11; RH 大于 70% 时, SOR 和 NOR 的均值分别增长至 0.40 和 0.19. 这表明成都市冬季 $PM_{2.5}$ 中的硫酸盐和硝酸盐通过二次转化生成的贡献很大, 较高的 RH 对二次硫酸盐和硝酸盐生成促进作用显著.

为了研究不同天气条件下 RH 对二次硫酸盐和硝酸盐生成的影响, 图5给出了整个观测期间、非霾过程和4个霾过程的 RH 与 SOR 和 NOR 的关系. 可以看出, 整个观测期间, 随着 RH 增加 ($> 60\%$), SOR 和 NOR 呈上升趋势, 4个霾过程的 SOR 和 NOR 值基本上均大于非霾期间的 SOR 和 NOR 值. 对于不同霾过程, RH 对二次硫酸盐和硝酸盐生成的作用不同. 霾过程1和4, 随 RH 升高,

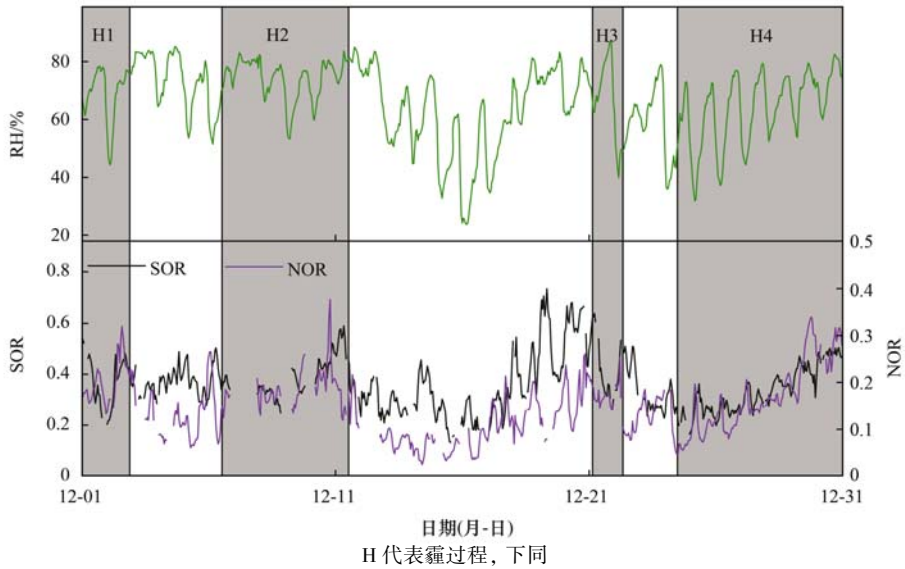
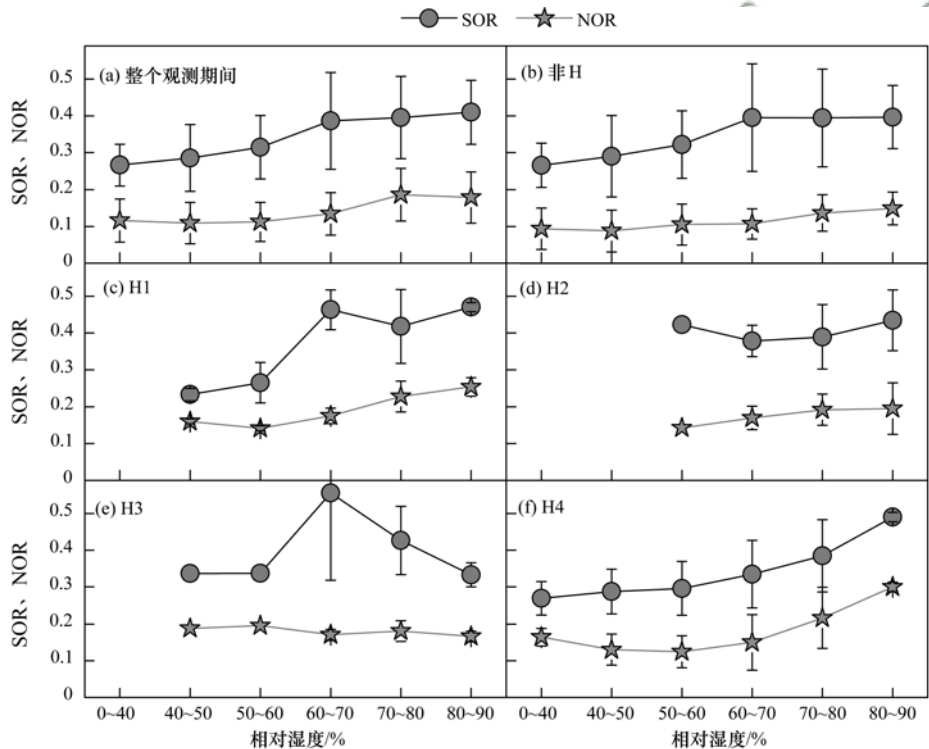


图 4 观测期间 RH 与 SOR 和 NOR 的时间序列

Fig. 4 Hourly variations of RH, SOR, and NOR during the observation period



线上的点分别表示 RH 在 <40%、40%~60%、60%~80% 和 80%~90% 区间的 SOR 与 NOR 的均值

图 5 整个观测期间、非霾期间、4 个霾过程的 RH 与 SOR 和 NOR 的关系

Fig. 5 Relationship between RH and SOR and NOR during the entire observation period, non-haze period, and the four haze episodes

SOR 和 NOR 均有较大的增长, 表明两个过程中通过非均相化学反应生成的二次硫酸盐和硝酸盐协同对大气能见度的降低起作用; 霾过程 2, 随 RH 升高, SOR 变化不明显, 一直保持在高值段, 而 NOR 缓慢增长, 表明此过程主要是二次硫酸盐大量生成导致能见度降低; 霾过程 3, 随 RH 升高, SOR 先升高, 后下降, NOR 变化不明显, 表明还是二次硫酸

盐对空气质量恶化起重要作用. 通过以上分析可知, 不同霾过程, RH 对二次硫酸盐和硝酸盐生成的促进作用不同, 影响空气质量的主要二次无机盐有所区别.

3 结论

(1) 观测结果表明, 成都市冬季气溶胶中细颗

颗粒物污染较为严重. 随 RH 升高, $PM_{1\sim2.5}/PM_{10}$ 增长了 146%, $PM_{2.5\sim10}/PM_{10}$ 降低了 36.2%, 表明成都市 RH 越高, 细颗粒物污染特征越明显, 能见度的恶化和细颗粒物污染加重与 RH 升高关系较大.

(2) 在不同 RH 区间内, 随着 $PM_{2.5}$ 浓度的增加, 能见度都呈幂指数下降. 当颗粒物浓度较低时, RH 对能见度的影响作用较强; 当颗粒物浓度较高时, 大气消光主要由 $PM_{2.5}$ 浓度控制, RH 对能见度的影响减弱.

(3) 整个观测期间, 随 RH 升高, SOR 和 NOR 显著增大, 表明 RH 对二次硫酸盐和硝酸盐生成促进作用显著. 不同霾过程, RH 对二次硫酸盐和硝酸盐生成的促进作用不同, 二次硫酸盐和硝酸盐单独或协同影响空气质量.

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