影响我国霾天气的多尺度过程

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摘要 频发的霾天气是我国现阶段面临的最主要大气环境问题之一。霾期间高浓度大气细颗粒物(PM_{2.5})是多种物理化学过程综合影响的结果, 包括排放、气-粒转化、大气边界层、局地环流、天气与气候等过程。上述过程的时空尺度跨越了几个数量级, 在空间尺度上涵盖了纳米尺度至上千千米尺度。多尺度过程本身的复杂性以及不同过程之间的相互影响是目前大气环境领域面临的最严峻挑战, 直接影响到对于霾天气形成机制的科学认识、预报技术与数值模式研发, 以及相应的大气污染治理。文章综述了在影响我国霾天气的多尺度过程及其与气溶胶的相互作用领域取得的研究进展。研究表明: 二次气溶胶已经成为我国大气气溶胶的主要部分, 在霾过程后期, 液相非均相过程对气-粒转化有重要贡献; PM_{2.5}呈现多时间尺度周期性振荡, 包括1, 4~7以及40~60 d等, 边界层、天气和气候等多尺度过程是造成上述周期性变化的主因; 已有证据表明, 我国高气溶胶已经影响到该区域大气光化学、大气边界层, 甚至天气和气候过程。气溶胶与上述过程的相互作用进一步影响了气溶胶浓度及其空间分布, 但是此问题极为复杂, 尚存在很大不确定性。为此, 今后需重点加强以下研究: 加强包含气溶胶理化性质、大气光化学、气象要素在内的多要素协同观测, 重点开展对流层内多要素协同垂直探测; 增强跨学科领域研究, 尤其是大气物理-大气化学-天气/气候等多学科间的交叉性研究; 加强气溶胶与大气化学、边界层、天气气候等过程相互作用的数值模拟研究。

关键词 霾, 气溶胶, 多尺度过程, 天气/气候过程

频发的霾天气是现阶段我国面临的最主要大气环境问题之一^[1~8], 尤其是在京津冀及周边、汾渭平原和长三角等重点区域。霾是大量极细微的干尘粒等均匀地浮游在空中, 使水平能见度小于10 km的空气出现普遍混浊现象。研究显示, 高浓度大气气溶胶是造成我国

霾天气频发的主要因素^[9~22], 气溶胶增加后消光系数增加, 能见度相应降低, 在霾天气中PM_{2.5}质量浓度可高达几百微克/立方厘米^[9,12,18]。气溶胶由直接排放的一次气溶胶(例如土壤粒子、海盐粒子、燃烧烟尘等)和气态前体物转化形成的二次气溶胶组成(例如硫酸盐、硝

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酸盐、铵盐、有机物等)。近期研究显示,二次气溶胶已成为我国大气气溶胶的主要部分,在重霾污染期间占比可高达80%左右^[20,23~29]。二次气溶胶的形成机理十分复杂,是多种化学过程综合作用的结果,包括光化学反应、成核过程、液相非均相反应等^[7,10,13,19,30~33]。源解析结果显示,交通源、燃煤、生物质燃烧、餐饮和矿物尘是气溶胶主要来源^[23~29]。

不利的气象条件也是推高霾天气过程中PM_{2.5}浓度的重要因素^[5,11,16,34,35],导致霾过程加重。风速小、大气静稳、近地层逆温、湿度高是产生霾天气的典型气象特征。气象因素一方面影响大气污染物的扩散与输送,大气扩散能力的降低导致气溶胶在近地面累积;另一方面气象因素也直接影响二次气溶胶的形成,例如霾天气过程中相对湿度的增加可促进液相非均相反应,加速气态前体物向颗粒物转化^[13]。因此,霾天气的形成与局地环流、天气和气候等气象过程密切相关^[14,35]。另外,地形特征也可通过影响流场,进而影响霾天气的形成。例如,徐祥德等人^[17]研究发现,中国区域霾日空间分布显著地受到青藏高原大地形的影响。在西风带背景下,青藏高原大地形东侧背风坡可构成“避风港”效应,这是中国东部区域霾日高频区域性分布的重要影响因素之一。此外,京津冀及周边区域西面、北面环山的地理特征也是导致该区域霾天气频发的重要因素之一^[36]。

霾天气的形成是多尺度物理化学过程综合影响的结果,在空间尺度上涵盖了纳米尺度至上千米尺度。多尺度过程本身的复杂性以及不同过程之间潜在的相互影响是目前科学认识我国霾天气成因面临的最严峻挑战。为此,我们综述了影响我国霾天气的多尺度过程及其与气溶胶的相互作用。此外,还针对未来需要开展的研究给出了部分参考建议。

1 影响霾天气的多尺度过程

1.1 大气气溶胶的来源

二次气溶胶已经成为我国大气气溶胶的主要组分。大气中气态前体物向颗粒物的转化主要包括光化学与成核过程、非均相反应过程和云水化学过程^[7,13,19,37~39],前两类过程是近地面二次气溶胶形成的主要过程,而后者主要发生在一定高度的对流层^[40~43]。在光化学与成核过程中,气态污染物氧化生成的低挥发性物质(例如硫酸和有机物氧化产物等)在大气中冷凝为分子簇,

继而通过冷凝碰并形成颗粒物^[7,44~46]。气态硫酸是参与成核的关键物种,二元成核机制($\text{H}_2\text{SO}_4+\text{H}_2\text{O}$)和三元成核机制($\text{H}_2\text{SO}_4+\text{NH}_3+\text{H}_2\text{O}$)被认为是经典的成核机制^[44~46]。除此之外,挥发性有机物在大气中的浓度很高,其氧化产物的挥发性较低,也被认为可能参与到新粒子生成过程^[22,47,48],包括有机酸、有机胺在内的多种有机物都被外场观测和实验室研究证明在新粒子生成过程中具有重要的作用^[49~56]。与光化学均相反应过程不同,非均相反应是气态分子在颗粒物表面发生的两相或多相反应。高相对湿度条件下,在亲水性颗粒物表面形成水膜。气态分子,如二氧化硫、O₃、过氧化氢(H₂O₂)、有机过氧化物、NO₂和OH等,可分配到液相,并发生液相反应^[7],加速气态前体物向颗粒物转化,导致颗粒物浓度快速增加^[13,37,38]。近期实验室和外场的观测研究表明,NO₂和SO₂的液相反应可能是我国大气中硫酸盐形成的关键机制之一^[10,21]。但也有研究认为,该反应机制取决于颗粒物的pH,一般发生在pH>5的情况,而华北地区大气颗粒物在雾霾期间pH一般在4~5之间^[57],且存在很高过氧化氢浓度^[58]。因此,H₂O₂与SO₂的经典液相反应被认为是硫酸盐形成的主要途径。所以,尽管取得了一些突破性认识,但对于气-粒转化过程的认识仍然不足,尤其是挥发性有机物(VOCs)在成核过程中的作用、VOCs的液相非均相反应机制等,有待继续开展更加深入的研究工作。光化学与成核过程生成的颗粒物的粒径小(纳米级)、浓度高,而液相非均相反应由于发生在颗粒物表面,反应的结果是增加气溶胶的粒径,而不是数浓度。因此,可根据气溶胶的数谱变化分析影响二次气溶胶形成的主要过程。图1显示了北京市大气气溶胶数谱与组分变化特征。结果显示,霾天气过程后期PM_{2.5}质量浓度增加主要由粒径增加造成,而不是数浓度,并且二次组分(硫酸盐、铵盐、硝酸盐)所占比例大幅增加。上述结果意味着在霾天气过程的不同阶段影响二次气溶胶形成的核心机制发生了很大变化。过程的初期以光化学过程为主,而后期伴随着相对湿度增加则转为以液相非均相反应为主。近期基于霾天气过程中气态前体物向颗粒态转化速率的分析结果也支持上述分析^[13,25]。以上分析显示,二次气溶胶的形成与气象要素密切相关,包括辐射、相对湿度、温度等,而气象要素则受局地、天气和气候等气象过程影响。同时,后者还会影响气溶胶的输送、扩散和累积等,下面将针对不同气象过程对气溶胶浓度变化以及相应霾天气的形成进行分析。

图 1 $PM_{2.5}$ 质量浓度(a)、13~736 nm粒径段粒子均值粒径(b)与数浓度(c)、气溶胶化学组分(d)以及 $PM_{2.5}$ 浓度为0~35, 35~75, 75~150, $>150 \mu g cm^{-3}$ 区间内平均化学组分(e). 观测时间为2012年9月27日~11月27日, 观测地点: 北京市海淀区宝联公园, 观测设备信息详见文献[12]

Figure 1 Time series of $PM_{2.5}$ mass concentration (a), mean diameter (b) and number concentration (c) of submicron aerosols (13–736 nm) measured by SMPS, and the aerosol composition (d) of PM_{10} measured by AMS from Sep. 27 to Nov. 27, 2012, at the Baolian meteorological station, with a time resolution of 1 h. Aerosol compositions (pie charts) with $PM_{2.5}$ concentrations ranging 0–35, 35–75, 75–150, and $>150 \mu g cm^{-3}$ are also shown (e). Detailed instruments' information are listed in Ref. [12]

1.2 大气边界层

大气边界层是靠近地球表面、受地面摩擦阻力影响的大气层区域, 也是地面与大气发生物质、能量交换的通道, 大气中绝大部分污染物都是通过大气边界层从地面输送进入大气. 受局地热力、动力过程以及天气形势的影响, 边界层存在很大的时空变化^[59–66], 垂直尺度通常为几百米至一两千米, 边界层可达到的最

大高度与热通量和湍能(TKE)强度密切相关, Quan等人^[11]对于城市三类典型天气(晴天、多云、霾)下边界层结构特征的分析结果显示, 云和气溶胶减弱了到达地面的辐射通量, 导致边界层内热通量大幅下降, 相应的边界层发展受到抑制, 高度降低. 而在沙漠地区, 超强的垂直热通量可导致最大边界层高度达到5 km. 边界层具有典型的日变化特征, 这将直接影响到大气污染物的日变化, 导致气溶胶浓度呈现夜间高白天低的

日变化特征。在边界层内部,垂直混合作用通常较强,污染物能够很快扩散到边界层顶部,导致在边界层内部大气污染物均匀分布,而边界层顶的存在会阻碍气溶胶向自由大气的输送^[67-69],因此大气边界层高度变化将直接影响地面大气气溶胶浓度^[11,70,71]。Zhang等人^[69]分析的飞机观测结果证实了上述观点,在边界层顶部存在较大气溶胶浓度梯度变化,并且大气稳定度越大,边界层内外气溶胶浓度差异越大。在霾天气过程中,边界层高度大幅下降,致使垂直扩散能力降低,造成气溶胶在近地面累积,浓度增加。除此之外,边界层高度的日变化特征也是边界层内气溶胶溢出边界层的重要途径,白天扩散至边界层上部的气溶胶并不能随着边界层高度的下降返回到近地面,而是遗留在残留层内,由此从边界层溢出。霾天气过程中,边界层高度昼夜差逐步减小,不利于边界层内的气溶胶溢出,进一步促进气溶胶在近地面内累积。除此之外,边界层结构还关系到气溶胶的区域输送,边界层内风速小、气溶胶浓度高;而自由对流层内风速大、气溶胶浓度低。因此,对于气溶胶是以边界层内输送为主,还是以自由对流层内输送为主目前尚存在很大争议。近期对于大气污染关注度的提高同时也推进了边界层领域的研究,目前边界层的研究主要是关注边界层高度,今后还需要开展边界层内辐射与湍流、感热/潜热通量、多要素垂直廓线协同观测,重点认识边界层过程对大气气溶胶垂直和水平方向输送的影响机制,以及气溶胶在边界层发展过程中的作用。

1.3 局地环流

局地环流是中、小尺度(几千米至几十千米)的区域性环流,如山谷风环流、城市热岛环流、海陆风环流等。霾通常发生在天气系统弱、气压梯度小的天气条件下。在这种天气条件下,因复杂下垫面的热力差异而产生的局地环流成为影响气溶胶输送和累积的关键因素^[72-85]。以北京为例,北京三面环山,太行山、燕山和军都山形成的“弓状山脉”可引发局地山谷风环流。长时间序列资料显示了北京风场具有显著的山谷风环流特征:凌晨至上午偏北风占优势,午后至前半夜偏南风占优势^[74]。局地环流的这种日变化特征,与边界层过程一起,成为影响气溶胶浓度日变化的主要机制。夜间下山气流带来洁净的高层气流,导致山风影响区气溶胶浓度大幅下降,这也是形成北京地区“南北两重天”现象的重要原因^[82]。下山气流与偏南暖湿气流的交汇

处形成气流辐合线,辐合线的汇聚作用有利于污染物和水汽在辐合线附近累积,导致在辐合线附近形成高气溶胶浓度区,而辐合带的南北摆动可引起地面气溶胶浓度的剧烈变化^[79],这也是北京地区PM_{2.5}浓度出现短时间剧烈波动的主要因素之一。山谷风的强弱与影响范围受多种因素影响,包括山体高度、环流背景、辐射、大气稳定度等多种因素影响,并存在显著的季节变化^[78]。同时,近期研究还显示,气溶胶也是影响山谷风环流的强迫因素之一,气溶胶浓度的增加导致大气稳定度增加,致使山谷风减弱。城市热岛也是诱发局地环流的机制之一,城郊温度差异引起空气在城市上升、在郊区下沉,形成了城市与郊区间的热岛环流。以上局地环流还会相互影响,多种局地环流间的叠加或耦合作用进一步增加了对局地环流科学认识的难度。局地环流是影响PM_{2.5}浓度短期波动的重要因素,直接关系到短期(24 h)雾霾预报的准确率,而目前的模式对该部分的模拟效果并不理想,天气模式主要关注大气流场和降水过程,而空气质量模式多关注源排放和化学过程。因此未来需要在模式研发方面开展更加深入的工作,进而提高短期精细化预报能力。除此之外,还需要开展局地环流和气溶胶空间分布的协同观测研究,提高对于多种局地环流耦合作用的科学认识。

1.4 天气系统

近年来在霾天气对应的天气系统方面开展了大量研究工作,弱经向型环流、弱均压场、偏南暖湿气流增强被认为是产生霾天气的典型天气特征。Wu等人^[86]的研究结果指出,在经向环流控制下,中高层为偏西气流,低层则为偏南暖湿气流且风速较小,仅为1~2 m/s,南部暖湿气流推高了低层相对湿度,而中低层的下沉气流则导致边界层高度降低,进一步抑制了气溶胶的垂直输送。张人禾等人^[16]对2013年1月份持续性雾霾天气演变的动力热力影响分析进一步指出,大气对流层中低层的垂直混合与500~850 hPa之间水平风的垂直切变密切相关,水平风垂直切变偏小时,垂直混合偏弱,不利于污染物向高空扩散;而热力作用体现在对流层中低层大气层结稳定性与能见度的关系,地面层逆温程度越强,能见度越小,即雾霾天气越强。Chen等人^[87]对于长时间序列资料(1960~2012年)分析结果也识别出霾日自由对流层底层存在逆温异常,导致低层大气稳定性增加。马小会等人^[88]进一步梳理了北京地区出现重霾天气对应的天气类型,识别出低压辐合区、

高压后部、高压底部、弱气压场、高压前部、低压倒槽、弱高压、鞍形场和华北地形槽等多种类型,其中低压辐合型、高压后部型和高压底部型天气所占比重最大。

天气系统的基本特征是存在周期性变化。在中高纬地区,长波槽(罗斯贝波)的波动周期为6~7 d,这也是华北区域大气PM_{2.5}浓度呈4~7 d周期性变化^[11,19,35,70,89]的主要影响机制。在霾天气过程初期,受高压系统控制,整层大气以西北风为主,风速大、扩散能力强,空气质量好。之后逐渐转为静稳天气,主要表现在风速减小、相对湿度增加、边界层高度降低、大气稳定度增加^[11,70]。气溶胶在此过程中逐步累积、浓度增加,大气能见度降低,导致霾天气。在一次过程中PM_{2.5}浓度达到的峰值浓度与该次天气过程持续的时间密切相关,通常一次天气过程持续的时间越长,后期PM_{2.5}达到的峰值浓度也越高^[12]。李梓铭等人^[35]基于Morlet小波和交叉小波对连续5年PM_{2.5}和常规气象资料的周期性变化诊断结果证实了上述周期性变化。除此之外,还诊断出了1, 14以及40~60 d的周期性振荡。1 d的周期性变化主要由边界层过程和局地环流造成(1.2和1.3节),14 d左右的振荡周期则与大气准双周振荡密切相关,而40~60 d季节内振荡这主要归因于季风环流调整(1.5节)。纪忠萍等人^[90]对广州市空气污染指数和各要素的周期研究表明也发现了类似周期性变化,进一步证实了天气过程对霾天气周期性变化的影响。天气过程不仅影响到污染物的扩散能力,还会影响到污染物的区域输送。例如,在高压后部型天气条件下,华北区域低空以偏南气流为主,致使河南、河北中南部污染物和水汽向北输送,形成沿太行山脉的污染物积聚带,导致北京南部、保定、石家庄、邢台及邯郸一带为重霾污染区^[88]。

1.5 季风-大尺度环流

我国所在的东亚是一个主要季风气候区,温度、降水和大气环流等气候特征具有明显的季节变化。季风环流的调整会影响到大气污染物的输送、扩散以及化学过程^[91~101]。20世纪80年代后中国霾日数呈逐年增加的趋势,尤其是进入2000年之后霾出现日数明显增多^[8]。尽管人类活动导致的排放增加主导了中国东部地区霾的日益频发^[4],但研究显示,减弱的季风环流也对霾日频发有一定的贡献^[102]。季风环流的减弱导致冷锋活动次数减少,地表风速降低,相对湿度增加^[102,103],

以上要素变化均有助于霾日增加。长时间序列雾霾年际变化与大气环流指数的相关性分析结果显示二者存在负相关关系^[91,104],即弱(强)冬季风导致更多(少)雾霾天气。2013年1月份发生在中国中东部强雾霾事件即是冬季风异常和高污染物排放共同作用的结果^[16]。季风的强弱还会影响到我国气溶胶的区域分布。以夏季风为例,弱季风年气溶胶光学厚度高值区位于中国南部,而之后的强季风年,分布位置则会扩展至中国北方^[95]。我国夏季风环流对应的主要天气系统为副热带高压。副热带高压经常以高压脊的形势伸向大陆,高压范围内盛行大范围的下沉气流,以晴朗、少云、微风、炎热为主,低层普遍形成逆温层,因此副热带高压控制区及其外围是高气溶胶浓度区^[81]。副热带高压的南北波动是影响气溶胶区域分布的气候强迫因子。我国季风存在典型的季节变化特征^[105]。大气环流的季节转换也是造成我国东部气溶胶浓度季节性变化的核心影响机制。在夏季风影响下,风速大、垂直对流深厚,降水频繁,以上因素均有利于气溶胶扩散和沉降,冬季则与之相反。与之对应,我国东部地区霾日出现频率冬季普遍偏高,秋、春季次之,夏季最轻。而青藏高原气溶胶季节变化趋势则与之相反,伴随着南亚季风的建立,南部污染物向青藏高原输送,导致该区域在春夏季出现气溶胶高值^[106]。

有效的外强迫因子能够通过影响季风-大尺度环流来影响霾天气的发生。影响我国季风环流的强迫因子与驱动力是多方面的,包括太平洋年代际变率、海平面温度(SST)、北极海冰面积、欧亚大陆春季积雪、北大西洋涛动、北极涛动等^[107~113]。在气候学研究的基础上,部分学者进一步分析了影响季风环流的外强迫因子与霾天数的关系。例如,Wang等人^[110]分析了北极海冰面积和中国东部霾天数的关系,结果显示二者存在显著负相关关系。秋季北极海冰面积的缩减有助于中纬度欧亚大陆海平面气压正异常,气旋活动路径北移,进而导致中国东部北纬40°以南冬季罗斯贝波减弱。上述大气环流的调整致使大气变得更加稳定,有助于霾日数增加。另外,中国东部由青藏高原、黄土高原与东部平原构成的“三阶梯”特殊的大地形作用,以及高原的热强迫也会影响该区域大气热力结构及其环流变化,进而影响大范围霾长周期(年代际)变异现象^[117,114]。徐祥德等人^[117]的研究显示,青藏高原东侧“背风坡”呈季节性顺时针垂直环流结构。冬季、春季以及秋季,在西风带背景下高原大地形东侧背风坡环流存在类似的

顺时针垂直环流圈; 夏季背风坡则为上升气流区. 此类大地形背风坡特殊环流圈的下卷气流区恰对应背风坡弱风区、霾日频发峰值区, 表明大地形背风坡弱风区及其下卷气流不利于大气扩散或对流, 形成类似静稳天气的“避风港”效应.

可见, 季风对于霾日的发生频率、区域分布、季节性变化均可产生一定影响, 近几十年东亚夏季风的减弱^[92,93]对中国区域气溶胶浓度的增加有贡献, 这为重新认识中国区域的高气溶胶浓度特征提供了一个新的视角^[100]. 下一步要加强霾天气成因和季风环流影响关联性的深入研究, 为长期重污染天气的预报预测、污染治理评估提供应用理论与技术支撑.

2 气溶胶与多尺度过程的相互作用

多尺度过程之间存在潜在的相互影响. 由前面分析可知, 大尺度过程(季风环流)可影响天气过程^[81,100], 而天气过程又可影响局地环流和边界层结构^[16,69], 以上过程均可影响近地面气溶胶浓度; 同时, 气溶胶也可通过辐射、云降水等过程影响大气化学、边界层、天气和气候等物理化学过程. 在以上反馈过程中尤以气溶胶与不同尺度系统的相互作用最为复杂, 成为该领域研究的热点和难点. 气溶胶与不同尺度系统的相互作用主要包括以下方面.

2.1 气溶胶与边界层过程相互作用

大气气溶胶通过吸收、散射过程影响太阳辐射, 导致到达地面的辐射减弱. 从全球尺度上看, 高气溶胶区为入射辐射的主要衰减区, 年衰减率为 $0.41 \text{ W m}^{-2} \text{ a}^{-1}$ ^[115]. 周秀骥等人^[116]结合观测资料和中国区域气候模式指出, 中国地区气溶胶辐射强迫范围在 $-5.3 \sim -13 \text{ W m}^{-2}$ 之间. 王昕然等人^[117]对北京污染和清洁天的辐射收支平衡分析结果显示, 冬季污染天入射短波辐射削减量高达 55.8 W m^{-2} . 大气气溶胶对太阳辐射的削弱作用将减弱近地层感热和湍流动能, 不利于边界层发展, 导致边界层高度降低^[11,65]. 另外, 吸收性气溶胶可吸收太阳辐射, 致使边界层上部增温, 低层降温, 大气稳定度增加, 也有助于边界层高度降低^[118]. 而边界层高度的降低又会降低垂直扩散能力, 致使边界层内气溶胶浓度增加. 因此, 气溶胶-边界层之间存在以下物理反馈机制: 气溶胶浓度的增加降低了到达地面的太阳辐射, 减弱边界层内的热通量和湍流, 导致边界层高度降低; 而边界层高度的降低又进一步促进

气溶胶在边界层内累积, 浓度增加. 上述反馈机制被越来越多的观测资料和数值模式结果证实^[11,118-124]. 研究还显示, 该反馈机制与气溶胶浓度密切相关, 当气溶胶浓度较低时反馈效应不明显, 而当气溶胶浓度较高时反馈效应显著增加^[65]. Zhong等人^[125]针对中国6个典型区域的观测研究结果进一步证实了上述分析, 在污染较重区域(华北平原、关中平原和东北平原)的反馈效果显著高于污染较轻的区域(长三角、两湖流域和珠三角).

在上述物理反馈机制的基础上, Liu等人^[70]又提出了气溶胶-边界层化学反馈机制. 该反馈机制的核心是边界层过程改变了边界层内的水汽和相对湿度, 后者直接影响到液相非均相反应. 在霾天气过程后期, 由于气溶胶对太阳辐射的削弱作用, 大气光化学能力在一定程度上降低, 但气-粒转化速率却显著增加, 这意味着液相非均相反应在气-粒过程中发挥主导作用^[13,70]. Liu等人^[70]的研究显示, 边界层高度的下降在促进气溶胶在近地面累积的同时, 也会同时促进水汽在近地面的累积, 导致相对湿度增加, 后者可促进液相非均相反应加速气-粒转化, 进一步增加气溶胶浓度. 上述化学反馈过程与物理反馈过程一起, 构成了气溶胶-边界层物理化学综合反馈机制, 该机制也是造成目前我国霾天气过程后期气溶胶浓度快速增加的重要机制之一.

2.2 气溶胶与天气/气候过程相互作用

天气/气候过程可影响气溶胶输送和扩散, 进而影响气溶胶浓度(见1.4和1.5节). 而气溶胶也会在一定程度上对天气/气候过程产生影响. 气溶胶对天气/气候多尺度过程的影响主要体现在以下两方面: 一是气溶胶可以散射和吸收太阳短波辐射以及地球长波辐射, 影响地气系统的辐射平衡(直接效应)^[126]; 二是气溶胶粒子作为云凝结核改变云的物理和微物理特征, 改变云的辐射特征和降水过程, 影响太阳能在地气系统中的分配(间接效应)^[127-130]. 大气气溶胶直接辐射强迫可正可负, 取决于气溶胶反射和吸收太阳辐射的相对能力(单散射反射率因子)以及地面反照率等其他因子^[100]. 对于硫酸盐这样的几乎不吸收太阳辐射的气溶胶来说, 其直接辐射强迫是负的, 部分抵消温室气体的温室效应^[109]. 但是对黑碳这样的强烈吸收太阳辐射的气溶胶而言, 其直接辐射强迫将是正的, 它会像 CO_2 等温室气体一样, 使大气增暖^[131]. 气溶胶对云和降水, 以及季风

环流的影响更加复杂,这与气溶胶特性、云系动力热力结构特征、天气、气候、地形等因素密切相关^[132,133]. Song等人^[134]的研究显示,东亚地区高气溶胶引起地表冷却,这样就会导致海陆温差减弱和华北高压异常,从而使得东亚夏季风低层环流减弱.类似的气候模式模拟结果也显示,东亚冬季寒潮爆发次数的减少和西北盛行风风速的减弱也与气溶胶的增加有关^[103].气候模式中气溶胶辐射特性参数的设定会影响气溶胶对季风强弱的效果分析,例如当将气溶胶设为强吸收型(单散射反射率0.85),气溶胶增加将减弱夏季风,导致中国夏季南涝北旱^[131],而将气溶胶单散射反射率提高到0.90,则气溶胶增加后的综合作用令夏季风环流有微弱的增强^[109].除上述辐射效应外,气溶胶还可以作为云凝结核影响到云和降水过程,徐祥德等人^[17]分析了1961~2010年我国降水变化趋势,发现在我国东部降水历史变化趋势中小到中雨发生频率下降,大到暴雨发生频率增加,并且该变化趋势与霾出现频率存在强相关性.基于华北区域云系飞机探测结果进一步证实气溶胶对降水的影响与降水系统的强弱密切相关,在弱降水系统中,气溶胶增加导致云滴粒径减小(Twomey效应)^[135],难以达到降水云滴所需的临界值,因此会抑制降水;而在强降水系统中,Twomey效应导致降水推迟,系统发展的更加旺盛,因此会增强降水.该结果与我国东部伴随气溶胶增加表现出的小到中雨发生频率下降,大到暴雨发生频率增加的观测结果吻合,意味着我国东部高浓度气溶胶已经成为影响该区域降水的显著性因子.气溶胶时空分布的差异性、天气/气候系统本身的非线性以及影响因素的复杂性,导致目前在该领域观测和数值模式工作结果不尽相同,甚至相反的研究结论.例如,气溶胶增加后,云滴粒径增加^[136]还是减小^[135]?云滴离散度增加^[137]还是减小^[138]?降水增加还是减小^[129]?以上问题均有截然相反的观测研究结果,意味着对于气溶胶在天气/气候过程中所起作用的科学认识还存在非常大的不确定性,未来有待继续开展更加深入的研究工作^[100].

2.3 气溶胶与大气化学过程相互作用

气溶胶中的二次气溶胶与化学过程密切相关(见1.1节),而气溶胶也会对化学过程产生影响.气溶胶对大气化学的影响主要体现在以下两方面:一是气溶胶影响近地面光化学辐射通量^[139],从而影响光化学能力与化学过程;二是气溶胶粒子可作为反应介质参与非

均相反应过程^[140~144]. Tie等人^[143]的工作显示,中国东部由于气溶胶增加导致冬夏季臭氧光解速率 $J(\text{O}_3)$ 分别下降20%~30%和5%~20%,对应的地表 O_3 浓度下降2%~4%和2%. Li等人^[144]对我国中东部的数值模拟工作显示,在低层气溶胶高浓度区对光解率的影响最大,夏季白天臭氧的光解速率常数 $J(\text{O}^1\text{D})$ 在低层(<1 km)的下降幅度可达51%,可导致 O_3 浓度下降5.4%. Liao和Seinfeld^[140]发现气溶胶表面对过氧自由基的摄取可导致近地面 O_3 浓度下降25%~30%. Li等人^[145]基于改进的NAQPMS模式同时分析了气溶胶的光化学辐射效应和非均相反应效应,进一步揭示了东亚地区气溶胶对该区大气光化学能力的影响.

在对流层内,气溶胶主要分布于低层或边界层内,气溶胶垂直分布的不均匀性导致其对大气光化学能力的影响在对流层内存在显著差异.低层由于高气溶胶的影响,光化学能力减弱,气态前体物向颗粒物的转化速率减慢,这就会导致更多的气态污染物向上输送.Chen等人^[146]的观测研究工作显示,在自由对流层底部 O_3 形成潜势转为 NO_x 敏感区,因此更多 NO_x 向上输送会增强该区域的大气光化学能力,导致在北京上空1~1.5 km处出现 O_3 高值区,这与清洁区存在显著差异.气溶胶对于对流层内大气光化学能力的影响也相应地影响到对流层内新粒子生成与物质循环过程^[147].在边界层内部气溶胶浓度高,气溶胶对光化学的削弱效应与气溶胶本身的吸附碰并效应会抑制新粒子生成;而在自由对流层内,增强的大气光化学能力与减弱的气溶胶吸附碰并效应会促进新粒子生成,导致在高气溶胶区新粒子的易发生区由边界层内部提升至自由对流层底部(图2),这与清洁区存在显著差异.Quan等人^[147]基于飞机观测的结果证实了上述分析.上述过程也影响到大气污染物在对流层内的循环过程,以硫循环为例,低层光化学能力的减弱导致更多的 SO_2 向高空输送,在高空转变为硫酸盐(图2).上述分析近期也被基于在线AMS(气溶胶质谱)气溶胶化学组分垂直观测资料所证实(图3),例如在华北区域硫酸盐在总气溶胶所占比例高空(34%)远高于近地面(14%),而在其他洁净区高空和近地面硫酸盐所占比例基本一致^[148~155].

平流层化学过程与辐射和动力过程是紧密耦合在一起的,平流层与对流层之间也存在复杂的相互作用,导致气溶胶对平流层化学过程的影响是一个非常复杂的问题.研究工作显示,对流层气溶胶增加使得大气层顶和地表短波净辐射通量减少,影响对流层的温度和

图2 气溶胶浓度对于对流层内新粒子生成以及硫循环影响的概念图。左列中阴影区代表新粒子的易发生区, 阴影区右边线距离中线的距离代表新粒子生成能力强弱, 绿色、红色和蓝色箭头分别代表与新粒子生成密切相关的气态硫酸浓度、吸附系数和碰并系数, 箭头长短表征数值大小, 具体数值见文献[147]。右列表示 SO_2 向高空输送以及向硫酸盐转化

Figure 2 Schematic diagram summarizing the effect of aerosol on NPF and sulfur cycles in the troposphere. Shadows in the left lines represent NPF capacity, and the arrows represent H_2SO_4 (green), condensation sink (red) and coagulation sink (blue) under high and low aerosol. The right lines shows the vertical transport of SO_2 and the conversion of SO_2 to sulfate. The data in this figure are from Ref. [147]

大气环流, 进而影响行星波的上传, 使得平流层气候发生变化^[156]。Tilmes等人^[157]的研究工作还显示, 平流层气溶胶增加可影响平流层内的化学过程, 尤其是通过促进非均相反应加速中高纬度臭氧消耗, 导致南极臭氧洞恢复时间推迟。而对于北半球, 极地涡旋的增强与温度的降低也导致臭氧消耗增加。上述研究工作显示, 气溶胶浓度变化可影响边界层-对流层-平流层化学过程, 目前在影响机制机理以及影响程度等方面尚不完全清楚, 今后仍需开展深入研究。

3 未来研究展望

综上所述, 我国东部地区频发的霾天气是在西风带与阶梯型大地形背景下多尺度系统过程协同作用的结果。多尺度过程涉及多学科、多领域复合, 复杂的交叉影响效应, 尤其多尺度系统理化过程之间潜在的相互反馈问题。这既是目前科学认识我国霾天气形成机制面临的最艰巨挑战, 同时也是目前大气污染治理、污染调控以及重污染天气预报工作面临的主要难题。未来有待在以下方面继续开展更加深入的研究工作。

(1) 深入开展多要素协同观测试验, 尤其是对流层

图3 北京上空气溶胶化学组分垂直分布特征。黑线代表气溶胶质量浓度随高度变化, 饼图代表不同高度气溶胶化学组分。观测设备为机载气溶胶质谱仪(AMS), 数据资料为2011年11月7次飞机探测资料平均值, 观测设备与飞行方案等具体信息见文献[71]

Figure 3 Aerosol composition profiles over Beijing, including mass concentrations (black line) and their mass fractions (pie charts), measured by AMS during November 2011. The data in this figure are from Ref. [71].

内多要素协同垂直探测。目前的观测主要集中在地面开展, 未来需要综合飞机、地基遥感、系留汽艇、卫星遥感等多种探测手段, 开展包含气溶胶物理、光学及化学特性、辐射、大气光化学、气态污染物、云宏观特性、气象要素在内的大型集成观测试验。结合大气环流背景场, 深入认识气溶胶的空间分布、来源、光学与辐射特性以及空间输送特征与影响机制, 及其对云降水、天气、气候的影响。

(2) 加强大气物理-大气化学-天气/气候等多学科间的交叉性研究。霾天气的形成是多种物理化学过程综合影响的结果, 涉及多学科、多领域复合, 以及复杂的交叉影响效应。气象过程的分析有助于认识气溶胶的变化特征、演变趋势, 而化学过程的分析有助于解析气溶胶的来源与形成机制, 未来需要实现气象与化学有机结合, 以深入认识对流层内的大气物理化学过程。

(3) 加强气溶胶与大气化学、边界层、天气/气候等过程相互作用的数值模拟研究。结合精细化气溶胶理化特性观测资料, 完善气溶胶-辐射方案, 发展适用于中国地区的气溶胶谱分布和云凝结核谱的参数化方案, 改进天气/气候模式; 发展气象-化学反馈模式, 实现气象/气候模式与大气化学模式间的双向耦合过程, 进而提高对于气溶胶与大气化学、边界层、天气/气候等过程相互作用机制的科学认识。

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Summary for “影响我国霾天气的多尺度过程”

Multi-scale processes in severe haze events in China and their interactions with aerosols: Mechanisms and progresses

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Severe haze events occur frequently in China, characterized by exceedingly high concentration of fine particulate matter (smaller than 2.5 μm , or $\text{PM}_{2.5}$). These extremes are caused by synthetic physical-chemical processes, including emissions, chemical formation, planetary boundary layer processes, regional circulation, weather and climate. These processes are multi-scales, ranging from nanometers to thousands of kilometers. The complex interplays among these processes make it more difficult to understand the formation of severe haze events, which also influences model development and weather forecast. Here, we review the contributions of dominant mechanisms in severe haze events, especially their roles in temporal oscillations of $\text{PM}_{2.5}$ concentrations, and discuss the interplays among these atmospheric multi-scale processes. Previous studies indicate that: (1) Secondary aerosols become the dominant components in aerosols. Heterogeneous aqueous reactions play important roles in gas-to-particle conversion, especially in the late haze period; (2) $\text{PM}_{2.5}$ owns extensive temporal oscillations (on daily, weekly, to monthly timescales), which are caused by multi-scale processes; (3) high aerosols in China have already influenced photochemistry, boundary layer, weather and climate processes. The complex interplays among aerosols and the above processes make it more difficult to unravel the causes, mechanisms, and trends for haze pollution. In the future, the following issues should be considered: cooperative observations of aerosols, gas pollutants, photochemistry, and meteorological variables should be strengthened, especially their vertical distribution in the troposphere; multidisciplinary research should be strengthened, especially among atmospheric physics, chemistry, weather and climate; and model simulations related to the interplays among aerosols and atmospheric physical and chemical processes should be strengthened.

haze, aerosol, multi-scale processes, weather/climate processes

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