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长江中游典型饮用水水源中药物的时空分布及风险评价 武俊梅,魏琳,彭晶倩,何鹏,施鸿媛,汤冬梅,吴振斌



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广西十万大山背景点 $PM_{2.5}$ 中非极性有机气溶胶组成及来源解析

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摘要:大气细颗粒物($PM_{2.5}$)中的非极性化合物包括多环芳烃(PAHs)和正构烷烃(n-alkanes)等,通常用于识别污染来源,且对人体健康和环境有很重要的影响.为探究广西背景点 $PM_{2.5}$ 中非极性有机气溶胶的污染特征及来源,于 2017 年 11 月至 2018 年 10 月,对野外采集的 $PM_{2.5}$ 样品分析了其中 17 种多环芳烃和 20 种正构烷烃.结果表明,多环芳烃和正构烷烃全年的平均值分别为(4.28 ± 4.25) $ng\cdot m^{-3}$ 和(13.7 ± 14.72) $ng\cdot m^{-3}$;季节变化规律均是:冬季[(7.86 ± 5.19) $ng\cdot m^{-3}$ 和(27.51 ± 16.9) $ng\cdot m^{-3}$] >春季[(2.73 ± 1.76) $ng\cdot m^{-3}$ 和(7.64 ± 4.71) $ng\cdot m^{-3}$] >秋季[(2.34 ± 1.45) $ng\cdot m^{-3}$ 和(7.01 ± 4.55) $ng\cdot m^{-3}$] >夏季[(1.91 ± 1.67) $ng\cdot m^{-3}$ 和(3.98 ± 3.12) $ng\cdot m^{-3}$]. PAHs 中5 环和6 环的分子占比较大,超过60%,其次是中低环分子(4 环和3 环); n-alkanes 中高分子量占比较高(P220 > P231 > P270),且奇偶碳数有明显差异. 另外结合特征比值法、主成分分析法和后向轨迹相互验证发现:冬季非极性有机气溶胶中41.5%受港口船舶交通排放、海洋源输送影响,36.7%的污染贡献来自燃煤和局地生物质燃烧;春季污染气团中有25.2%来自生物质燃烧传输影响,45.0%来自研究区域南部的海洋输送,高等植物蜡排放污染较高;夏季53.4%气团污染来自船舶交通源传输,10.6%的污染来自西南泰国的跨境输送.广西背景点有机气溶胶受到局地排放和传输源的共同影响.

关键词:非极性化合物;多环芳烃(PAHs);正构烷烃(n-alkanes);后向轨迹分析;主成分分析中图分类号: X513 文献标识码: A 文章编号: 0250-3301(2022)06-2895-11 **DOI**: 10.13227/j. hjkx. 202110031

Organic Aerosols and Source Analysis of Fine Particles in the Background of Shiwanda Mountain, Guangxi

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Abstract: The non-polar compounds in atmospheric fine particles (PM_{2.5}) mainly include polycyclic aromatic hydrocarbons (PAHs) and normal alkanes (*n*-alkanes), etc., which are usually used to identify the source of the air pollution and have a great important impact on human health and the environment. To study the pollution characteristics and sources of non-polar organic aerosols in the background point PM_{2.5} in Guangxi, from November 2017 to October 2018, 17 types of PAHs and 20 types of *n*-alkanes were analyzed on PM_{2.5} samples collected in the field. It was found that the annual average concentrations of PAHs and *n*-alkanes were 4. 28 ±4.25 ng·m⁻³ and 13.7 ±14.72 ng·m⁻³, respectively. The seasonal change was as follows: winter [(7.86 ±5.19) ng·m⁻³, (27.51 ±16.90) ng·m⁻³] > spring [(2.73 ±1.76) ng·m⁻³], (7.64 ±4.71) ng·m⁻³] > autumn [(2.34 ±145) ng·m⁻³, (7.01 ±4.55) ng·m⁻³] > summer [(1.91 ±1.67) ng·m⁻³], (3.98 ±3.12) ng·m⁻³]. In PAHs, 5-ring and 6-ring molecules accounted for more than 60%, followed by low- and medium-ring molecules (4-ring and 3-ring). The high molecular weight of *n*-alkanes was relatively high (C29 > C31 > C27), and the odd and even carbon numbers were significantly different. In addition, combined with the feature ratio method, principal component analysis method, and backward trajectory joint verification, it was found that 41.5% of non-organic aerosols in winter were affected by maritime traffic emissions and ocean source transportation, and 36.7% of the pollution was explained by the coal burning and local biomass burning; 25.2% of the pollution in spring came from biomass combustion and transportation, and 45.0% was attributed to marine transportation in the southern part of the study area and higher plant wax emissions pollution; 53.4% of pollution in summer came from polluted ship emissions, and 10.6% of pollution came from transportation in Southwest Thailand Source transportation. The organic aerosols at the background sites in Guangxi were affected

Key words: non-polar compound; polycyclic aromatic hydrocarbons (PAHs); normal alkanes (n-alkanes); backward trajectory analysis; principal component analysis

 $PM_{2.5}$ 是指大气颗粒物中动力学直径 \leq 2.5 μm 的粒子,其来源和组成复杂,对人体健康和环境质量有很大的影响. 面对我国日趋严重的大气污染,近年来针对 $PM_{2.5}$ 的研究已成为各部门关注的焦点. 其中,有机气溶胶(organic aerosols, OAs)是大气颗粒

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物的重要组成部分,占 $PM_{2.5}$ 质量的 $20\% \sim 60\%^{[1,2]}$,由来自不同源的化合物组成,有着不同的物理、化学特性[3,4],在不同程度上影响着气候效应和人体健康[5].

有机气溶胶组成复杂,多环芳烃(polycyclic aromatic hydrocarbons, PAHs)和正构烷烃(normal alkanes, n-alkanes)是其中重要的非极性化合物部 分. 正构烷烃的主要来源是生物体的脂肪酸、蜡质 和烃类物质[6],水生藻类和微生物是碳数低于20的 短链正构烷烃的主要贡献者[7],而 C26~C36 这类 高分子量正构烷烃来源于陆生高等植物蜡[8]. 其中 高分子量的正构烷烃危害性相对低分子量更高,更 易吸附在细颗粒物表面,对人体呼吸系统有重要的 影响[9,10]. 多环芳烃占比虽然较低, 却不容忽视, 主 要来源于生物质的不完全燃烧和化石燃料燃烧等人 为过程[9~11],另外也有自然排放,包括火山喷发 等[12,13],具有致癌、致毒和致畸性[14,15]等特点,美 国环保署(U.S. Environmental Protection Agency, EPA) 将其列为优先控制的环境污染物,共 16 种[16~18]

目前国内外对长三角、北京和东北等地区有机 气溶胶的组成以及来源已有较多研究,采用多种分 析方法,包括特征比值法[19~22]、主成分分析法 (principal component analysis, PCA) [23~25]、有机分 子标志物法^[26,27]和同位素分析法等^[28]. Liu 等^[29]的 研究利用生物质燃烧示踪剂与卫星数据结合以及模 式运算的方式,发现长三角的生物质燃烧信号来自 东南地区短时大面积的秸秆燃烧活动. Chen 等[30] 的研究综合运用特征比值法和正定矩阵因子分解 (positive matrix factorization, PMF), 发现在观测期 间青藏高原东南部受人为污染和自然排放的共同影 响,冬季家庭采暖会排放大量正构烷烃.本地多环芳 烃来源依次为生物质燃烧、汽油燃烧、住宅燃烧和 煤燃烧,另外,还受印度生物质燃烧传输的影响. Zheng 等[31]研究了东南亚春季生物质燃烧对东京湾 附近海洋有机气溶胶的影响,结合稳定碳同位素方 法,分析了5类化合物发现:生物质燃烧是东京湾上 空有机气溶胶的重要来源. 此外, 左旋葡聚糖常与其 他化合物进行相关性分析,是植物纤维素热解产生 的一种脱水糖,作为生物质燃烧最常用的指示 剂[32].

大多有机气溶胶的研究集中在长三角、珠三角和京津冀等发达地区^[20,33,34],对于我国西南地区有机气溶胶的分析较少. 另外东南亚作为生物质燃烧旺盛区域,燃烧集中期在干燥的 3 月和 4 月,此时盛行偏西风^[31,35],广西不仅毗邻东南亚生物质燃烧旺

盛的地区,可能受到传输影响,还靠近珠三角经济发达的贸易区,可能受到船舶排放影响,选取广西作为采样点有十分重要的意义. 另外采样站点位于十万大山腹地,人烟稀少,受到本地人为排放污染小,可作为背景点参考. 因此,本研究在广西十万大山腹地布设站点,通过采集 PM_{2.5}颗粒物,分析多环芳烃和正构烷烃的浓度变化特征及来源,结合后向轨迹和主成分分析法探讨污染物的潜在来源,对华南地区污染防控有重要意义.

1 材料与方法

1.1 研究区域与样品采集

采样点位于广西防城港市郊区南屏乡常隆村 $(20^{\circ}48'35''N, 107^{\circ}38'02''E)$,海拔高度为 759 m,处在十万大山腹地,靠近东南亚生物质燃烧旺盛区域,并且临近珠三角经济发达的贸易区. 采样时间为 2017 年 11 月至 2018 年 10 月,一周采一次,流量为 $1.05~\text{m}^3\cdot\text{min}^{-1}$,每次采样时长为 48~h (崂应 2031,青岛崂山应用技术研究所). 共采集 45~h PM_{2.5}样品,冬季 16~h ,春季 10~h ,夏季 11~h ,秋季 8~h ,采集完成后放入 -26~h 的环境中冷藏待测.

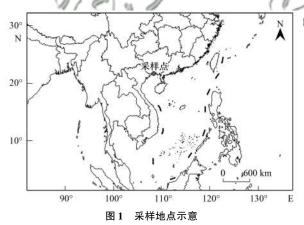


Fig. 1 Sampling location indication

1.2 实验方法

1.2.1 OC/EC 测定

碳质组分的测定使用全自动半连续式 OC/EC 分析仪(Sunset Laboratory,美国),切取 1.5 cm^2 样品膜在石英炉纯氦气环境下经不同加热程序挥发有机碳(OC),随后升温在氦氧混合气(10% 氧气)中氧化元素碳(EC)生成气态氧化物,两种气体经催化转化为 CO_2 ,后用非散射红外法定量.每个样品测定后使用 CH_4/CO_2 标准气校正仪器,每日进样前使用蔗糖标准溶液定标.

1.2.2 左旋葡聚糖测定

左旋葡聚糖(levoglucosan, Lev)利用离子色谱 (ICS 5000 + ,Thermo Scientific)进行测定,样品预处

理方法为:用打孔器(punch)切 $1 \sim 2$ 片 20 mm 直径的样品膜,并加入 8 mL 超纯水(Milli-Q Reference, 美国),经过 30 min 的超声萃取,再用 0.22 μ m 的过滤器(水相针式,聚四氟乙烯滤膜)过滤,最后进行仪器测定.

1.2.3 非极性化合物测定

采用 安 捷 伦 气 相 色 谱 质 谱 仪(Agilent GC7890B/MS 5977A)的 热 脱 附(TD)方 法 测 定 PAHs 和 n-alkanes 化合物浓度. 使用 30 m × 0. 25 mm × 0. 25 μm 规格的毛细管柱,用打孔器切下半径 为 0. 15 cm 的样品膜 2 ~ 3 片,放进 TD 内衬管,进样口初始温度为 35 ℃,运行 5 s,然后以 12 ℃·min $^{-1}$ 升温到 300℃;柱箱初始温度为 35 ℃,运行 3 min,然后以 10 ℃·min $^{-1}$ 升温到 120℃,运行 11. 5 min,再以 4 ℃·min $^{-1}$ 升温到 146℃,运行 18 min,最后以 8 ℃·min $^{-1}$ 升温到 310℃,保持 16 min 并运行 54. 5 min. 高纯氦气作为载气的流速为 1. 0 mL·min $^{-1}$,进样方式为分流比 5: 1的分流进样,进样体积为 1 μ L,

将六甲基苯作为内标物质,采用内标法进行定量. 共测定了 17 种 PAHs: 萘(Nap)、苊烯(Acy)、二氢苊(Ace)、芴(Flu)、菲(Phe)、蒽(Ant)、荧 蒽(Flua)、芘(Pyr)、苯并[a]蒽(BaA)、萹(Chry)、苯并[b] 荧蒽(BbF)、苯并[k] 荧蒽(BkF)、苯并[e]芘(BeP)、苯并[a]芘(BaP)、二苯并[a,h]蒽(DahA)、茚并[1,2,3-cd]芘(IcdP)和苯并[g,h,i] 芤(BghiP); 20 种 C16 ~ C35 的 n-alkanes.

1.2.4 特征比值法分析

特征比值法常用于有机气溶胶的朔源研究中^[12,33,36]. n-alkanes 的诊断方法包括两种:最大碳数(Cmax)和碳偏好指数(carbon preference index, CPI),用来评估 n-alkanes 的相对贡献. CPI 为奇数碳烷烃与偶数碳烷烃浓度和的比值,常作为估量化石源和生物源排放贡献的一个定性指标:当 n-alkanes 以植物蜡源为主时 CPI 值通常较高(CPI > 5),而当 n-alkanes 以化石源排放为主时 CPI 值接近 $1^{[7,37]}$:

$$CPI = \frac{(C17 + C19 + C21 + C23 + C25 + C27 + C29 + C31 + C33 + C35)}{(C16 + C18 + C220 + C22 + C24 + C26 + C28 + C30 + C32 + C34)}$$
(1)

WaxCn 为含碳数为 n 的正构烷烃的植物蜡源部分的浓度贡献:

WNA 为植物蜡的贡献率(%),计算如下:

WNA =
$$\frac{Cn - 0.5 \times [C(n-1) + C(n+1)]}{C(n-\text{alkanes})}$$

$$\times 100\% \tag{3}$$

FFNA(fossil fuel *n*-alkanes, FFNA)为化石燃料 贡献率(%):

$$FFNA = 1 - WNA \tag{4}$$

1.2.5 主成分分析

主成分分析法(PCA)是一种最广泛的数据降维计算的统计学方法. 现通常用来分析研究有机气溶胶组分的来源^[19,21,36,38],本研究使用 SPSS 25 进行计算,再通过因子旋转和回归分析,最后确定不同源的贡献率.

1.2.6 后向气流轨迹分析

本研究利用拉格朗日计算模型: HYSPLIT-4 进行后向轨迹分析,选择 500 m 高度上向后 48 h 轨迹模拟,每隔 6 h 输出一条轨迹,再进行聚类分析,气象数据是美国国家海洋和大气管理局(NOAA)在全球数据管理系统(GDAS)上发布的. 火点数据从FIRMS (fire information for resoutce management system)网站获得.

1.3 控制变量

每次采样前需对仪器状态进行检查,包括流速、流量和时间的检查,采样期间每周需要对切割头进行清洗,每个月进行一次野外空白采样,作为平行样品.实验过程中,每天需要打入一个2mg·L⁻¹的标准和一个空白,以此来检验仪器状态是否正常.每切一个样品膜之前需对镊子和打孔器进行清洗,使用二氯甲烷:甲醇体积比为2:1的混合溶液,减少实验误差.另外参考美国环境保护署(EPA)方法^[16],进行实验室回收率实验,并对所有样品进行空白校正,空白样品低于真实样品中对应目标化合物浓度的20%,对后续目标化合物定量无明显影响.各化合物的标准曲线相关系数达0.99以上,满足后续测样要求,氘代内标的回收率为70%~120%,满足定量分析的要求,所有物质均高于检测限.

2 结果与讨论

从 2017 年 11 月至 2018 年 10 月进行野外样品 采集,分析了 $PM_{2.5}$ 中的相关化学组分: PAHs 和 n-alkanes, 其相关浓度见图 2 和表 1. 采样期间, $\rho(PM_{2.5})$ 的年平均值为(35. 31 ± 21. 86) $\mu g \cdot m^{-3}$,变 化范围在 13. 9 ~ 91. 2 $\mu g \cdot m^{-3}$,其中有 21 d 低于我 国大气环境质量标准 24 h 日均一级浓度限值(35 $\mu g \cdot m^{-3}$)[39], 41d 低于二级浓度限值(75 $\mu g \cdot m^{-3}$).

研究区域的 OC 和 EC 年平均值分别是(2.90 ± 2.5) μ g·m⁻³和(0.41 ±0.34) μ g·m⁻³,远远低于北京夏季农村地区^[40,41], PAHs 和 n-alkanes 的年平均值分别是(4.28 ± 4.25) ng·m⁻³和(13.70 ± 14.72) ng·m⁻³,低于其他背景站点如长三角背景点临安

 $[\rho(PAHs): 25 \text{ ng·m}^{-3}, \rho \text{ (} n\text{-alkanes}\text{)}: 60 \text{ ng·m}^{-3}]^{[42]}$,高于其他城市地区如拉萨西郊 $[\rho(n\text{-alkanes}): (10 ± 6.6) \text{ ng·m}^{-3}]^{[43]}$,与喜马拉雅相当 $[\rho(PAHs): 5.57 \text{ ng·m}^{-3}]^{[44]}$,表明该区域适宜作为背景点进行研究.

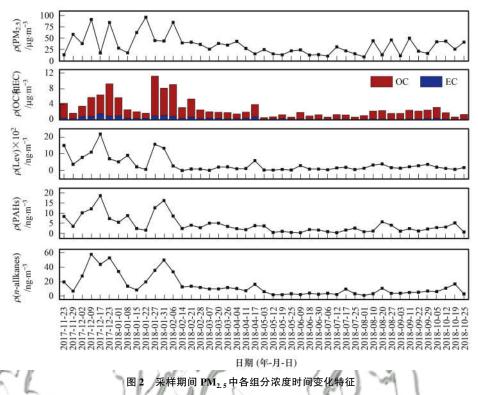


Fig. 2 Temporal variation in the concentration of each component in PM_{2.5} during the sampling period

OC 的主要来源是燃烧排放[41,45],也可以是植 物孢子、花粉和藻类等微小生物直接排放形成气溶 胶[46],在冬季明显高于其他季节,但远低于东南亚 地区[45],表明冬季相比其他季节受到的燃烧或者生 物排放影响更大,而整个地区受到的影响较小.结合 图 2 和表 1 可知, PAHs、n-alkanes、Lev、OC 和 EC 的变化趋势一致,其中 PAHs 和 n-alkanes 各季节的 总浓度变化均为:冬季[(7.86 ± 5.19) ng·m⁻³, (27.51 ± 16.9) ng·m⁻³] > 春季[(2.73 ± 1.76) $\text{ng} \cdot \text{m}^{-3}$, (7.64 ± 4.71 $\text{ng} \cdot \text{m}^{-3}$] > 秋季[(2.34 ± 1.45) $\text{ng} \cdot \text{m}^{-3}$, (7.01 ± 4.55) $\text{ng} \cdot \text{m}^{-3}$ > 夏季 [(1.91 ± 1.67) ng·m⁻³, (3.98 ± 3.12) ng·m⁻³]. 具 体来说,中高环 PAHs(4~6环)的浓度高于低环(2 环和 3 环) 浓度. 高分子量(含碳数 > 25) 的 nalkanes 浓度高于低分子量的浓度(含碳数 < 20),总 体年平均值为(17.70 ± 14.72) ng·m⁻³, n-alkanes 整体浓度相较于 PAHs 要高出 3~4倍,在 PM,5中 所占含量极低,仅0.39%,却不容忽视.Lev作为生 物质燃烧的常用示踪剂[47],在化合物中占有较高的 数量级,是植物热解产生的一种脱水糖^[32],与 OC 有着较好的相关性(r=0.69),对研究该区域的污染 来源有一定的指示作用. 在春季的 Lev 浓度最低,这与临安背景地点不同^[38],表明在春季受到的生物质燃烧影响较低.

为探究研究区域不同季节下有机污染物的传输 模式,对四季进行后向轨迹分析.结果如图 3 所示, 冬季气团轨迹分成了5个聚类,其中聚类2和聚类 3 分别占了 20% 和 30%, 有 50% 的气团污染来自低 空海洋传输,采样点靠近广西最南部的北部湾,临近 港口,既沿海又沿边,与东盟各国海陆相连,经济贸 易加速发展中,推测有交通运输的影响.聚类1占比 24%,主要来自采样点附近,结合图 2, 12 月 22 日 OC 浓度较高,且该日前后在本地有极明显的火点, 属于本地源排放,聚类4和5共占26%,来自北方高 空远距离传输.春季有5个聚类,有69%来自海洋 传输,有8%的气团来自泰国,图2中显示在4月16 日污染较高,通过检查火点数据发现该目前后有密 集的生物质燃烧,且从单日气流轨迹可知是来自泰 国、越南和老挝等地,还有19%来自本地源排放.夏 季,在季风的影响下有37%气溶胶来自泰国,此外 还有19%的气溶胶来自港口地区,18%来自东部南 海,26%来自广西中部地区.秋季有4个聚类,43%

表 1 采样期间不同组分不同季节的平均值1)

Tabla 1	Avorage	annaantrations o	fdifferent	components in different	accorna d	unina	compline:	nariad

项目	冬季	春季	夏季	秋季	全年
OC	5.17 ± 3.01	1.66 ± 0.99	1.41 ± 0.57	1. 99 ± 0. 77	2.90 ± 2.53
EC	0.73 ± 0.38	0.34 ± 0.18	0.16 ± 0.04	0.20 ± 0.09	0.41 ± 0.34
PM _{2.5}	50.03 ± 26.91	26.4 ± 10.08	23.52 ± 13.17	31.75 ± 14.4	35.31 ± 21.86
Lev	7.32 ± 6.5	1.42 ± 1.75	1.79 ± 1.16	2.01 ± 0.94	3.71 ± 4.78
$\sum { m PAHs}$	7.86 ± 5.19	2.73 ± 1.76	1.91 ± 1.67	2.34 ± 1.45	4.28 ± 4.25
$\sum n$ -alkanes	27. 51 ± 16. 9	7.64 ± 4.71	3.98 ± 3.12	7.01 ± 4.55	13.7 ± 14.72
Nap(2)	0.02 ± 0.01	0.01 ± 0	0.01 ± 0	0.01 ± 0.01	0.01 ± 0.01
Ace(3)	0.01 ± 0.01	0 ± 0	0 ± 0	0.01 ± 0.01	0.01 ± 0.01
Acy(3)	0.01 ± 0	0 ± 0	0 ± 0	0 ± 0	0 ± 0
Flu(3)	0.01 ± 0.01	0.01 ± 0	0 ± 0	0 ± 0	0.01 ± 0
Phe(3)	0.18 ± 0.22	0.02 ± 0.01	0.01 ± 0.01	0.02 ± 0.02	0.07 ± 0.15
Ant(3)	0.03 ± 0.03	0.01 ± 0.01	0.01 ± 0	0.01 ± 0	0.02 ± 0.02
Flua(4)	0.6 ± 0.7	0.09 ± 0.06	0.04 ± 0.02	0.09 ± 0.1	0.26 ± 0.48
Pyr(4)	0.58 ± 0.68	0.1 ± 0.07	0.04 ± 0.02	0.11 ± 0.12	0.26 ± 0.47
BaA(4)	0.57 ± 0.51	0.13 ± 0.09	0.08 ± 0.09	0.14 ± 0.13	0.28 ± 0.38
Chr(4)	0.8 ± 0.67	0.2 ± 0.14	0.11 ± 0.11	0. 19 ± 0. 16	0.39 ± 0.51
BbF(5)	1.05 ± 0.61	0.43 ± 0.32	0.29 ± 0.3	0.35 ± 0.21	0.6 ± 0.54
BkF(5)	0.51 ± 0.3	0.21 ± 0.17	0. 15 ± 0. 16	0. 18 ± 0. 11	0.3 ± 0.27
BeP(5)	0.76 ± 0.42	0.31 ± 0.23	0.23 ± 0.24	0.26 ± 0.14	0.44 ± 0.38
BaP(5)	0.61 ± 0.43	0.21 ± 0.13	0. 15 ± 0. 12	0.19 ± 0.15	0.33 ± 0.34
DahA(5)	0.1 ± 0.05	0.04 ± 0.03	0.03 ± 0.03	0.03 ± 0.02	0.06 ± 0.05
IcdP(6)	0.93 ± 0.49	0.42 ± 0.31	0.34 ± 0.32	0.33 ± 0.19	0.57 ± 0.45
BghiP(6)	1. 11 ± 0. 43	0.54 ± 0.3	0. 4 ± 0. 29	0.41 ± 0.17	0.68 ± 0.46
C16	0.78 ± 1.03	0.08 ± 0.03	0.06 ± 0.03	0.08 ± 0.05	0.33 ± 0.69
C17	0. 29 ± 0. 21	0.12 ± 0.06	0.09 ± 0.07	0.12 ± 0.09	0.17 ± 0.16
C18	0. 12 ± 0. 07	0.05 ± 0.02	0.04 ± 0.03	0.15 ± 0.29	0.09 ± 0.13
C19	0.1 ± 0.05	0.11 ± 0.09	0.09 ± 0.08	0.69 ± 1.34	0.2 ± 0.58
C20	0. 14 ± 0. 12	0.04 ± 0.02	0.04 ± 0.02	0.13 ± 0.23	0.09 ± 0.13
@ C21	0. 31 ± 0. 27	0.09 ± 0.06	0.08 ± 0.08	0.23 ± 0.26	0.19 ± 0.22
C22	0. 55 ± 0. 57	0.07 ± 0.06	0.04 ± 0.02	0.11 ± 0.14	0.24 ± 0.41
C23	1 ± 0. 89	0.26 ± 0.14	0.07 ± 0.06	0.11 ± 0.05	0.45 ± 0.67
C24	1. 14 ± 0. 83	0.19 ± 0.1	0.09 ± 0.05	0.13 ± 0.07	0.49 ± 0.69
C25	1.81 ± 1.1	0.47 ± 0.29	0.22 ± 0.14	0.34 ± 0.11	0.86 ± 0.98
C26	1.89 ± 1.14	0.44 ± 0.27	0.29 ± 0.25	0.41 ± 0.18	0.91 ± 1.01
C27	2. 99 ± 1. 84	0.87 ± 0.61	0.41 ± 0.3	0.68 ± 0.28	1.48 ± 1.61
C28	2.34 ± 1.51	0.63 ± 0.4	0.38 ± 0.33	0.71 ± 0.4	1. 19 ± 1. 28
C29	3.65 ± 2.39	1. 4 ± 1. 02	0.54 ± 0.38	0.9 ± 0.56	1.9 ± 2.02
C30	1.98 ± 1.32	0.56 ± 0.42	0.36 ± 0.34	0.56 ± 0.32	1. 02 ± 1. 1
C31	3.41 ± 2.25	1. 16 ± 0.77	0.46 ± 0.39	0.71 ± 0.46	1.71 ± 1.9
C32	1. 49 ± 1. 01	0.41 ± 0.29	0.24 ± 0.26	0.32 ± 0.23	0.74 ± 0.84
C33	1. 84 ± 1. 2	0.44 ± 0.29	0.24 ± 0.23	0.32 ± 0.27	0.87 ± 1.03
C34	0.86 ± 0.59	0.13 ± 0.09	0.14 ± 0.16	0.15 ± 0.1	0.4 ± 0.5
C35	0.81 ± 0.67	0.13 ± 0.11	0.12 ± 0.14	0.18 ± 0.11	0.38 ± 0.52

1) OC、EC 和 $PM_{2.5}$ 的单位为 $\mu g \cdot m^{-3}$,其余的单位为 $ng \cdot m^{-3}$,括号内的数字表示 PAHs 的环数

来自广东西部地区短距离传输,27%来自广西北部的湖南,另外还有来自海洋上污染排放的影响.总体来说,采样点的污染源分为海洋源,远距离传输和本地源.

2.1 正构烷烃来源解析

本研究测定了 20 种 n-alkanes (C16 ~ C35),表 1 给出了这些化合物不同季节以及年平均值. 从图 4 (a)中 n-alkanes 的季节变化规律和图 4(b)中不同

碳数 n-alkanes 的占比可知,冬季和春季平均 n-alkanes 占比较高,别占到 59.6% 和 16.6%,其次是秋季和夏季,占 15.2% 和 8.6%.其中,全年含量最高的是 C29,其次是 C31 和 C27,各季节的 Cmax 为冬季(C31)、春季(C29)、夏季(C29)和秋季(C31),体现了植物源的排放特征^[8]. C19 在秋季有明显较高的占比,Tian 等^[48]的研究指出燃煤和锅炉燃烧会在 C16 ~ C19 产生一个较高的峰值. n-

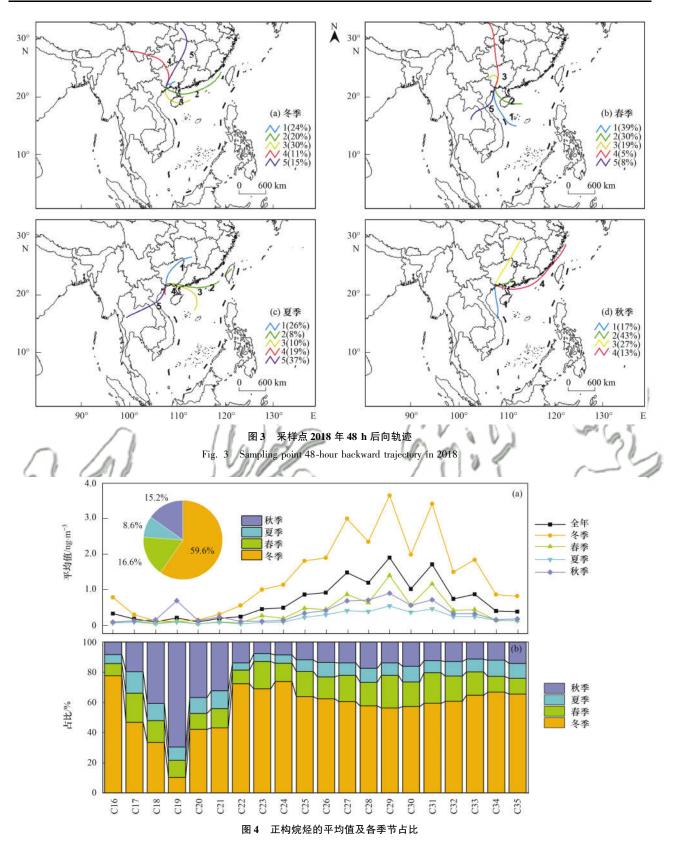


Fig. 4 Average of n-alkanes and the percentage of each season

alkanes 随碳数的变化在四季有一定的相似规律,其来源贡献可以解释丰度变化模式,从 C26 开始有明显的奇偶差异,且奇数碳烷烃高于偶数碳烷烃,表明高等植物蜡的排放强于人为排放的影响^[49].

有研究表明,人为源和生物源均是大气中 n-

alkanes 的来源^[36,37],人为源主要贡献包括化石燃料燃烧和生物质燃烧^[8],而植物蜡排放、花粉和生物体内的脂肪酸是生物源的主要贡献者^[24]. 化石燃料燃烧主要排放含碳数低于 25 的 *n*-alkanes^[28];而高等陆生植物主要排放含碳数高于 26 的 *n*-alkanes,奇

数碳烷烃的含量远高于偶数碳的含量^[50].该采样点作为广西背景点,不仅受到本地源的影响,还有远距离输送带来的人为污染影响.同时采样点处在十万大山腹地,植被茂盛,因此会释放大量的脂肪族化合物^[33].为比较化石源和自然源的相对贡献,本研究计算了 CPI、WNA 和 FFNA^[1,13,37],结果如表 2显示.

观测期间,冬季 CPI 值为 1.45,春季 1.90,夏季 1.50,秋季 1.53,这表明采样期间正构烷烃是由化石源和自然源共同作用的^[49].春季 CPI 值接近 2,表

明在春季生物源的贡献会相对较高,在冬季化石燃料燃烧等人为排放影响较大;各季节生物源的贡献分别为:春季(30.6%)>秋季(20.6%)>夏季(19.2%)>冬季(18.9%),这与CPI值的研究结果一致.在春季植物生长旺盛,此外东南亚处于生物质燃烧旺盛的季节,图3中后向轨迹发现有小部分气团污染来自泰国等地,影响了春季高等植物蜡的排放贡献.夏季气团较为干净,但化石源占比较高,结合后向轨迹发现,来自海洋传输,推测是船舶交通运输的影响.

表 2 采样点不同季节 PM_{2.5} 中正构烷烃和多环芳烃的诊断值

Table 2 Diagnostic ratios of n-alkanes and PAHs in PM _{2.5} in different seasons at sampling site									
项目	冬季	春季	夏季	秋季	全年	文献数据			
Cmax	C31	C29	C29	C31	C29				
CPI	1. 45 ± 0.23	1.9 ± 0.24	1. 5 ± 0.37	1. 53 ± 0.26	1. 58 ± 0.32	$1.35 \pm 0.43^{[36]}$			
WNA/%	18.93 ± 6.06	30.6 ± 4.82	19. 21 ± 10.48	20.59 ± 7.35	21.89 ± 8.57				
FFNA/%	81. 07 \pm 6. 06	69.4 ± 4.82	80.79 ± 10.48	79. 41 ± 7.35	78. 11 \pm 8. 57	/3			
IdcP/(IdcP + BghiP)	0.44 ± 0.05	0.4 ± 0.08	0. 41 ± 0. 1	0. 42 ± 0. 08	0.42 ± 0.08	0.5~0.57(冬)[51]			
BaA/(BaA + Chr)	0.4 ± 0.05	0.4 ± 0.05	0.44 ± 0.02	0.4 ± 0.03	0.41 ± 0.05	0. $2 \sim 0.35^{[37]}$			
Flua/(Flua + Pyr)	0.51 ± 0.02	0.48 ± 0.03	0.45 ± 0.02	0.47 ± 0.05	0.48 ± 0.04	0.46~0.48(冬)[51]			
BaP/BghiP	0.49 ± 0.23	0.36 ± 0.12	0.33 ± 0.11	0.43 ± 0.19	0.41 ± 0.19	$0.9 \sim 6.6^{[37]}$			
(BaP + BeP)/BghiP	1. 14 ± 0.38	0.86 ± 0.28	0.81 ± 0.3	1.05 ± 0.31	0.98 ± 0.35	$1.2 \sim 1.7^{[52]}$			
BeP/(BeP + BaP)	0.59 ± 0.08	0.58 + 0.06	0.58 ± 0.05	0 61 + 0 08	0.59 ± 0.07	0.73[33]			

oble 2. Diagnostic ratios of nullance and PAHs in PM. in different seasons at sampling site

2.2 多环芳烃来源解析

大气中 PAHs 是一种混合物^[8],主要由煤和石油等化石燃料及木材和烟草等含碳氢化合物不完全燃烧或热解所形成^[51].本研究测定了17 种 PAHs 在颗粒物样品中的丰度,并确定了每种物质在不同季节的浓度变化情况. 从表 1 和图 5(a)可知,其中浓度大小顺序为: BghiP(6 环) > BbF(5 环) > IcdP(6

环) > BeP(5 环) > BaP(5 环), 从图 5(b) 和 5(c) 可知,各物质在冬季中含量最高,表明在冬季受到的燃烧影响最大,总 PAHs 在各季节中的占比分别为冬季(53%) > 春季(18.4%) > 秋季(15.7%) > 夏季(12.9%). 从图 5(d) 可知,各季节中 5 环 PAHs 的占比最高,其次是 6、4、3 和 2 环.

为了确定 PAHs 的来源,采用诊断率来进行具

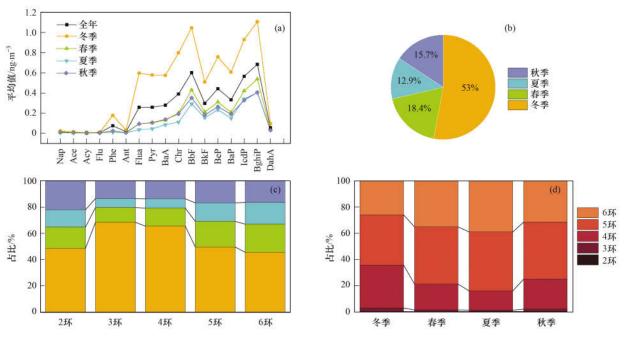


图 5 PAHs 的平均值和占比

Fig. 5 Average and percentage of PAHs

体解析,本研究通过计算 IdeP/(IdeP + BghiP)、BaA/(BaA + Chr)、Flua/(Flua + Pyr)、(BaP + BeP)/BghiP 和 BaP/BghiP 来探究广西背景点不同季节中多环芳烃的潜在来源污染,具体计算结果见表 2. 当 IdeP/(IdeP + BghiP) < 0. 2 时,代表石油源,在 $0. 2 \sim 0. 5$ 之间表示污染来自石油燃烧和燃煤的混合源,> 0. 5 时表示生物质燃烧 [13];当 BaA/(BaA + Chr) < 0. 2 时代表了石油源, $0. 2 \sim 0. 35$ 时表示燃煤源和汽油源, $0. 35 \sim 0. 5$ 来自燃煤排放,> 0. 5 时表示生物质燃烧 [11];当 Flua/(Flua + Pyr) < 0. 4 时代表了石油源, $0. 4 \sim 0. 5$ 代表了化石燃料燃烧,> 0. 5 时表示生物质燃烧 [37].

本研究 IdcP/(IdcP + BghiP) 四季的均值在 0.4 左右,表明 IdcP 和 BghiP 多来源于燃煤以及化石燃 料燃烧; BaA/(BaA + Chr)也在 0.4 左右,表明这两 种污染物多由燃煤排放; Flua/(Flua + Pyr)四季均 在 0.5 左右波动, 是化石燃料、燃煤、生物质燃烧的 混合源. 另外,通过计算 BeP/(BaP + BeP)来判断气 溶胶的光解老化程度^[9], BeP 光学性质稳定,不易 老化分解, BaP 更容易光解老化, 比值越大说明老 化程度越大. 本研究中 BeP/(BaP + BeP) 四季均在 0.6 左右,说明本次研究中气溶胶老化程度相对较 高. 冬季和秋季污染程度相对较高, 气溶胶光解老化 程度相对较低,冬季和秋季 BghiP 主要由生物质不 完全燃烧产生. 而夏季清洁大气加上高温加速了气 溶胶光解老化,导致(BaP + BeP)/BghiP 和 BaP/ BghiP 相对冬季和春季要低,主要是交通排放污染. 而春季气溶胶受远距离污染输送的影响,光解老化

程度升高,(BaP + BeP)/BghiP 和 BaP/BghiP 也是相对较低,这与上文所得的结论一致.

2.3 主成分分析(PCA)

特征比值法只能定性表征污染潜在来源,为了更有效识别不同季节下污染源的贡献,利用 SPSS 25对研究区域不同季节进行主成分分析(PCA). 其中2 环和3 环的 PAHs 浓度过低,且在传输过程中易降解^[44], BaP 易光解氧化^[13],因此选取了中高环的多环芳烃进行分析; 低碳数的 *n*-alkanes 受温度影响较大^[53],各季节中 Cmax 贡献较高的是 C29 和 C31,因此选取典型低分子量 C23 和 C24 以及高分子量 C29 和 C31 进行分析. 另外 Lev 作为典型的生物质燃烧示踪剂^[32],增加了结论的可靠性. 由于秋季样品量过少,导致分析结果出现较大误差,这里就不予讨论.

PCA 结果如表 4 所示,提取了特征值大于 1 的物质,并进行旋转,各季节的因子均能解释总变量的90%以上.其中冬季提取了 3 个因子,因子 1 解释了总变量的 41.5%,其中载荷较高的化合物是 BbF、BkF、BeP、IcdP、DahA 和 BghiP,BbF 和 BkF 作为柴油排放的指示物[37],BeP、IcdP 和 BghiP 作为汽油燃烧排放指示物[51],此外采样点靠近北部湾,与东盟国家进行经济贸易,船舶运输较多,结合后向轨迹结果分析[图 3 (a)]和表 2 可知:有 50% 的气团来自东部和南部的海洋,CPI 为 1.45,IdcP/(IdcP+BghiP)的值为 0.44,是石油和化石燃料燃烧的混合源,也验证了之前的猜想,因此因子 1 解释为汽油和柴油燃烧;因子 2 载荷较高的是 Flua、Pyr、BaA和

表 4 不同季节中多环芳烃和正构烷烃的旋转因子载荷1)

		Table 4 Rotation factor loading of PAHs and n-alkanes							
1万日	冬季			春季			夏季		
项目	1	2	3	1	2	3	1	2	3
Flua	0. 34	0.86	0. 32	0. 21	0. 95	0. 22	0.89	_	_
Pyr	0.35	0.86	0.31	0. 23	0. 92	0.31	0.83	_	_
BaA	0.60	0.78	0. 11	0.59	0. 63	0.48	0.76	_	0. 24
Chr	0. 57	0.78	0. 19	0.67	0.64	0.35	0.79	_	0. 24
BbF	0.82	0.41	0.39	0.94	0. 27	0. 18	0.99	_	0.04
BkF	0.88	0.41	0. 21	0.96	0. 27	0. 12	0.98	_	0.01
BeP	0.87	0.40	0. 28	0.95	0. 25	0. 17	0.99	_	0.02
IcdP	0.91	0.35	0. 22	0.96	0. 20	0. 22	0.98	0.01	_
DahA	0.86	0.46	0. 21	0.97	0. 20	0. 13	0.98	0.05	_
BghiP	0.89	0. 24	0. 23	0.88	0. 21	0.43	0.98	_	_
C23	0. 28	0.85	0. 37	0.35	0.62	0.65	0.90	_	0.34
C24	0.38	0.74	0. 52	0.34	0.61	0.67	0.73	0.39	0. 19
C29	0.30	0. 28	0. 90	0.38	0.30	0.85	0.66	0. 69	_
C31	0. 20	0. 25	0. 94	0.39	0. 53	0. 67	0.72	0. 63	_
Lev	0.62	0.70	0.03	0.00	0. 18	0.96	0.39	0.48	0. 62
累计贡献方差/%	41.5	78. 1	96. 6	45. 0	71. 9	97. 1	53. 4	81.9	92. 5
来源	交通源	燃煤、生物 质燃烧1	自然源	交通源	燃煤	生物质 燃烧1	交通源	自然源	生物质燃烧2

1)"一"表示载荷为负,不显示;黑体字表示相对载荷较高的值

Chr,这4种物质通常用来指示燃煤排放^[48],且表2显示Flua/(Flua + Pyr)的值为0.51,表明是生物质燃烧的影响,另外低分子量的 n-alkanes 与 Lev 的荷载也相对较高,主要是生物质燃烧排放,因此因子2归为燃煤源以及生物质燃烧源;因子3中高分子量n-alkanes的荷载较高,然而 PAHs 的荷载都比较低,左旋葡聚糖与之相关性也不高,因此因子3解释为本地自然源排放,主要包括植物蜡排放.

春季提取了3个因子,因子1和2与冬季相似,但在因子2中左旋葡聚糖与其他物质相关性不高,因此因子2解释为燃煤源;因子3中多环芳烃的贡献较低,其大气寿命小于左旋葡聚糖^[32],C29和C31与左旋葡聚糖荷载较高,相关性也好,图3(b)中显示有8%的气团来自泰国和老挝地区,表明本地受到了远距离传输的影响,在传输过程中低环的多环芳烃降解,另外春季是东南亚生物质燃烧旺盛的季节^[35,54],燃烧也会产生高分子量的n-alkanes,因此因子3解释为生物质燃烧,解释了变量的26.1%,这与上文结论一致.

夏季提取了3个因子,因子1解释了变量的53.4%,后向轨迹结果中显示有37%来自海洋传输[图3(c)],与冬季相似,柴油排放的指示物的荷载较高,且结合表2结果:IdcP/(IdcP+BghiP)的值为0.41,因此因子1归为交通源;因子2中荷载较高的是C29和C31.左旋葡聚糖与之相关性不高,表明来自高等植物蜡排放,因此解释为自然源;因子3中除左旋葡聚糖荷载较高外,其余贡献较低,图3中除左旋葡聚糖荷载较高外,其余贡献较低,图3中除左旋葡聚糖荷载较高外,其余贡献较低,图3个少显示聚类5有37%来自泰国和老挝地区,此时处于非生物质燃烧旺盛的季节,是在季风的影响下,气团污染传输到本地,虽然传输量大,但影响较小,解释了变量的10.6%,因此因子3归为生物质燃烧传输影响(生物质燃烧2).综合表明,广西郊区的空气污染主要来源于燃煤、交通排放、生物质燃烧以及植物排放.

3 结论

(1) 2018 年广西 $\rho(PM_{2.5})$ 年平均值为(35.31 ±21.86) $\mu g \cdot m^{-3}$, $\rho(OC)$ 和 $\rho(EC)$ 平均值依次为(2.9 ±2.53) $\mu g \cdot m^{-3}$ 和(0.41 ±0.34) $\mu g \cdot m^{-3}$, 分别占 $PM_{2.5}$ 的 8.7% 和1.8%. 左旋葡聚糖是重要的生物质燃烧示踪剂, 在冬季有较高的浓度. 多环芳烃和正构烷烃的季节变化规律一致, 均是冬季和春季浓度高, 秋季和夏季浓度低. PAHs 中5 环和6 环的分子占比较大, 超过 60%, 其次是中低环分子(4 环和3 环); n-alkanes 中高分子量占比较高(C29 > C31 > C27), 且奇偶碳数有明显差异. 其在 $PM_{2.5}$ 颗

粒中虽占比较低,不足 1%,但由于较大的危害性而不容忽视.

(2) 综合运用特征比值法、正定矩阵因子分解 法和后向轨迹发现:冬季非极性有机气溶胶中 41.5%受港口船舶交通排放、海洋源输送影响, 36.7%的污染贡献来自燃煤和局地生物质燃烧;春 季,污染气团中有25.2%来自生物质燃烧传输影响, 45.0%来自研究区域南部的海洋输送,高等植物蜡排 放污染较高;夏季,53.4%气团污染来自船舶交通源 传输,10.6%的燃烧污染来自西南泰国的跨境输送; 秋季燃煤占比较高.广西虽然靠近东南亚生物质燃烧 旺盛区域,但在春季受到的影响较低.因靠近珠三角 地区,受海上船舶排放的传输影响较高.

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