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高原城市拉萨典型VOCs排放源成分谱特征

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摘要: 高海拔地区低的气压和含氧量会影响燃烧和溶剂挥发过程, 进而影响挥发性有机物(VOCs)的排放特征。然而, 人们对海拔3 000 m以上的燃烧源或挥发源的VOCs排放特征的认识有限。拉萨市正在经历快速的经济发展和城市化进程, VOCs排放源日趋复杂。采集拉萨市机动车尾气、汽油挥发、柴油挥发、液化石油气和燃香等排放源VOCs样品进行分析, 研究成分谱特征, 并与平原地区的相比较。发现液化石油气VOCs排放谱以丙烷和丁烷为主, 与平原地区的一致; 汽油挥发中1,2,4-三甲苯占比最高, 而汽油车尾气中异戊烷、正丁烷和丙烷占比高, 1,2,4-三甲苯占比较低, 这不同于平原地区的结果; 柴油挥发比汽油及汽油车排放具有更高占比的C7以上高碳化合物; 柴油车排放中含氧挥发性有机物(OVOC)占比最高, 而平原地区则是丙烯和C7以上烷烃占比高; 燃香排放中OVOC占比最高, 特征示踪物是乙腈, 且有不容忽视的异戊二烯排放。从甲苯与苯的比值上看, 汽油车及柴油车排放与平原地区的结果基本一致, 但两地区柴油挥发和燃香排放则不同。从苯、甲苯和乙苯比例三元图上看, 柴油挥发落在工业及溶剂排放区内, 有别于平原地区的结果。与其它城市一样, 烯烃和芳香烃化合物也是高原城市中具有高反应活性VOCs组分, 二者分别对臭氧和二次有机气溶胶的生成有重要的贡献。初步研究结果表明, 高原城市地区不同VOCs源的组分特征与平原地区有所差异, 需要进一步深入研究, 以应对潜在的环境和健康风险。

关键词: 挥发性有机物(VOCs); 源谱; 成分谱; 排放特征; 羟基消耗速率(L_{OH}); 臭氧生成潜势(OFP)

中图分类号: X511 文献标识码: A 文章编号: 0250-3301(2024)04-2011-08 DOI: 10.13227/j.hjxx.202305048

Composition Characteristics of Typical VOCs Sources in the Highland City of Lhasa

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Abstract: The low air pressure and oxygen content at high altitudes affect combustion and solvent volatilization processes, which in turn affect the emission characteristics of volatile organic compounds (VOCs). However, there is very limited knowledge about the VOCs emission characteristics of combustion sources or volatile sources over 3000 m above sea level (asl). Lhasa City has been undergoing rapid economic development and urbanization, where VOCs emission sources are becoming increasingly complex as a result of energy consumption changing from biomass burning to fuel combustion. Samples from major VOCs emission sources in Lhasa City, including motor vehicle exhaust, gasoline volatilization, diesel volatilization, liquefied petroleum gas (LPG), and incense burning emissions, were collected for analysis to study their compositional spectra and compare them with those in the plains. The VOCs emission profile of LPG was found to be consistent with those in previous studies in the plain area, with dominant propane and butane; gasoline evaporation had the highest proportion of 1,2,4-trimethylbenzene, whereas gasoline vehicle emissions had high proportions of isopentane, n-butane, and propane and a low proportion of 1,2,4-trimethylbenzene, unlike what has been reported in the plain area. Diesel volatilization had a higher proportion of C7 and higher carbon compounds than gasoline volatilization and gasoline vehicle emissions. Diesel vehicle emissions had the highest ratio of oxygenated VOC (OVOC), which was different from the high compositions of propylene and alkanes above C7 in the plains. Incense burning had the highest percentage of OVOC emissions, with acetonitrile as the tracer that distinguished it from other emission sources; it also had a small amount of isoprene emissions, which should not be overlooked. As for the ratio of toluene to benzene (T/B) in gasoline and diesel vehicle exhaust, they were generally consistent with that in the plain area. However, both T/B ratios in diesel evaporation and incense burning were >1, which differed from that in the plain area. In the ternary diagram of benzene, toluene, and ethylbenzene ratios, diesel volatilization fell within the industrial and solvent emission zones, which differed from that in the plain area. Like other cities, alkenes and aromatics were also highly reactive VOCs species in highland cities, contributing significantly to ozone and secondary organic aerosol generation, respectively. Preliminary results indicated that the composition characteristics of VOCs sources in highland areas differed from those in the plains, and further in-depth studies are needed to address their potential environmental and health risks.

Key words: volatile organic compounds (VOCs); source profiles; component spectrum; emission characteristics; hydroxyl radical loss rate(L_{OH}); ozone formation potential (OFP)

挥发性有机物(volatile organic compounds, VOCs)对近地面臭氧(O_3)和二次有机气溶胶(secondary organic aerosol, SOA)的生成有重要影响, 从而影响环境与气候^[1,2]。在全球尺度上, VOCs的天然源占绝对优势, 而在城市及经济发达地区, 人为源的贡献更大^[3]。人为源VOCs成分谱及组分清单存在着较高不确定性^[4]。因此, 获得准确的VOCs排放源成分谱是一项重要的基础工作。我国逐步构建了

涵盖工业、交通运输、溶剂使用、生物质燃烧和餐饮等主要行业VOCs排放源成分谱, 建立了不同尺度VOCs组分排放清单, 并广泛应用于城市或区域 O_3 污染防控研究^[5]。然而, 已开展的大量VOCs排放

收稿日期: 2023-05-06; 修订日期: 2023-06-25

基金项目: 国家自然科学基金项目(21876214)

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源谱的研究工作,主要集中在低海拔地区,尤其是京津冀、长三角和珠三角等经济发达地区^[6]。此外,以往的VOCs源谱中常常缺少含氧有机物(OVOC)成分。然而,有研究表明柴油汽车排放的OVOC可占总VOCs的20%~70%^[6]。同时,由于OVOC具有较高的化学活性,OVOC在清单中的缺失,导致其重要性被低估^[7]。近年来,基于化学反应活性的VOCs污染控制思路逐渐受到重视,完整的VOCs排放源谱是亟需完善的重要基础数据^[8]。

海拔高于3 600 m的城市拉萨正在经历快速的经济发展和城市化进程,污染物排放逐年增加,VOCs由早期相对简单的生物质燃烧排放^[9]逐渐发展成为复杂的多源排放^[10]。拉萨市处于受高山包围的河谷地带,人为排放的污染物在强的太阳辐射和高的臭氧背景的加持下,存在着光化学污染风险^[11-13]。在高海拔地区,低的气压和含氧量会影响燃烧和溶剂挥发过程,从而影响VOCs的排放特征。然而,人们对海拔高于3 000 m以上的燃烧源或挥发源等VOCs排放特征的知识非常有限。本研究通过对拉萨主要VOCs排放源,包括机动车排放、汽油挥发、柴油挥发、液化石油气和燃香排放的VOCs组分进行分析,计算不同源的各组分对O₃和SOA生成的潜在贡献,并与低海拔地区的结果进行比较,以期在今后在高原城市进行VOCs及其相关的污染控制提供科学的依据。

1 材料与方法

1.1 样品采集

使用3 L苏玛罐(Entech Instruments Inc., USA)进行负压全空气样品实地采样,采集汽油车尾气、汽油挥发、柴油挥发、液化石油气(LPG)挥发、柴油车尾气和燃香排放等。参照美国EPA TO-15方法,采样前使用清洁装置(Entech Instruments Inc., USA)和加湿氮气进行清洗和预处理。苏玛罐入口使用无水硫酸钠颗粒去除O₃等氧化物,并使用聚四氟乙烯膜过滤颗粒物^[14,15],负压瞬时采样,采样时间不超过10 s。其中,在加油站汽车加油过程中采集汽油挥发样品1个;在柴油车熄火状态下,打开油箱采集柴油挥发样品1个;在大货车密集通过路口时采集柴油车尾气排放样品3个;在加气站汽车加气过程中采集LPG挥发样品1个;在封闭空间内燃烧当地常用香,采集燃香排放样品3个;于09:00早高峰时段在拉隆隧道(全长2 000 m,双向双车道,大型货运车辆限行)内距离入口处约150 m,离地面约1 m处采集汽油车尾气排放样品3个。同一源多样本的结果取平均值。

1.2 VOCs检测分析

所采集的罐样品采用低温预浓缩(鹏宇昌亚,北京)联合气相色谱-质谱(GC-MS)分析系统(GCMS2010, SHIMADZU, Japan)进行VOCs成分分析。其中,13种C₂~C₅化合物通过PLOT-Al₂O₃色谱柱(15 m × 0.32 mm ID × 3 μm, J&W Scientific, USA)分离后,用氢火焰离子化检测器检测;其他85种化合物(间-二甲苯和对-二甲苯未能分开,标记为“间/对-二甲苯”)通过DB624色谱柱(60 m × 0.25 mm ID × 1.4 μm, J&W Scientific, USA)分离后,用单四极杆质谱在选择性离子扫描模式下检测(EI源, 70 eV)。一个完整的分析循环包括:准备、采样及预浓缩、进样及分析和反吹等过程,详细可见文献^[16]。

结合PAMS混合标气(Linde Co., Germany)、EPA TO-15 OVOCs和卤代烃的混合标气(Spectra Gases Inc., USA)在0.25 × 10⁻⁹、0.5 × 10⁻⁹、1.0 × 10⁻⁹、2.0 × 10⁻⁹和4.0 × 10⁻⁹这5个体积分数下对仪器进行多点校准。质谱检测过程中也采用溴氯甲烷、1,4-二氟苯、氯苯d5和溴氟苯这4种化合物作为内标物辅助分析。VOCs的校正曲线R²值在0.995~1.000之间。为了检查仪器的稳定性,使用混合比为2.0 × 10⁻⁹的标准气体定期进行响应检查。

1.3 活性计算

采用最大增量反应活性(maximum incremental reactivity, MIR)法计算各VOCs组分的臭氧生成潜势(ozone formation potential, OFP),以评估不同VOCs组分对其O₃生成的贡献。对特定VOC组分*i*有:

$$\text{OFP}_i = \text{MIR}_i \times [\text{VOC}]_i \quad (1)$$

式中, MIR_{*i*}为组分*i*最大增量反应活性常数,其值为2010年更新的数据^[17,18], [VOC]_{*i*}为组分体积分数。

VOCs组分对·OH的消耗速率(loss rate of hydroxyl radical, L_{·OH*i*})利用式(2)计算:

$$L_{\cdot\text{OH}_i} = K_{\cdot\text{OH}_i} \times [\text{VOC}]_i \quad (2)$$

式中, K_{·OH_{*i*}}为VOCs组分*i*与·OH的反应速率常数^[19]。

VOCs组分对二次有机气溶胶的生成潜势(secondary organic aerosol formation potential, SOAP)利用下式计算^[20]:

$$\text{SOAP}_i = \Delta\text{SOA}_i / \Delta\text{SOA}_T \quad (3)$$

式中, ΔSOA_{*i*}和ΔSOA_{*T*}分别为VOC组分*i*和甲苯浓度增加导致的SOA生成潜势。

2 结果与讨论

2.1 排放源VOC组成特征

2.1.1 汽油蒸发

体积分数前5位的VOCs组分分别为:1,2,4-三

甲苯(13.2%)、甲苯(8.1%)、异戊烷(7.7%)、间/对-二甲苯(6.9%)和苯(5.6%),占汽油蒸发排放的总挥发性有机物(TVOCs)的40%;质量分数前5位的为:1,2,4-三甲苯(19.5%)、甲苯(9.1%)、间/对-二甲苯(8.8%)、异戊烷(6.7%)和苯(5.3%),占汽油蒸发排放的TVOCs的50%。这与陆思华等^[21]在北京测量的结果差异较大,后者体积分数前5的组分分别为异戊烷(17.6%)、正丁烷(4.8%)、2,2,4-三甲基戊烷(4.7%)、2-甲基戊烷(4.4%)和2-甲基-2-丁烯(4.3%)。与之相比,本研究中1,2,4-三甲苯占比最高,异戊烷占比略低。不过,高的1,2,4-三甲苯占比与区家敏等^[8]在珠江三角洲汽油车尾气成分分析结果类似。从分类上看,汽油挥发中芳香烃化合物占比最大(37.2%),其次为烷烃(24.0%)和OVOC(23.5%);从C数上看,C5~C6化合物的质量分数为26.1%,而北京测的汽油蒸发中C5~C6化合物占比高达35.0%^[21],相比略低。

2.1.2 汽油车排放

体积分数由高到低的前10种VOC分别为:丙烷(9.9%)、异戊烷(7.7%)、正丁烷(7.6%)、乙烯(7.5%)、丙酮(6.2%)、异丁烷(4.5%)、2,3-二甲基丁烷(4.2%)、乙烷(3.6%)、甲苯(3.3%)和乙醛(3.2%);质量分数由高到低的前10种分别为:异戊烷(8.4%)、正丁烷(6.7%)、丙烷(6.7%)、2,3-二甲基丁烷(5.5%)、丙酮(5.5%)、甲苯(4.6%)、甲基叔丁基醚(MTBE)(4.3%)、异丁烷(4.0%)、乙烯(3.2%)和正戊烷(2.7%)。图1对比了在北京和拉萨测得的汽油车尾气成分。北京汽油车尾气中乙烯(12.1%)、乙烷(12.0%)、正戊烷(10.9%)、间/对-二甲苯(7.6%)和2,3-二甲基丁烷(5.4%)是主要组分,拉萨汽油车尾气则以丙烷、异戊烷、正丁烷和乙烯为主要组成,且OVOC组分占比更高。Liu等^[22]用底盘测功机测得汽油车排放的VOCs主要有:乙烯、异戊烷、甲苯、苯、间/对-二甲苯、邻-二甲苯和1,2,4-三甲苯,但OVOC没有测量。与之相比,本汽油车排放研究中丙烷占比高,1,2,4-三甲苯占比低,其他主要组分基本一致。珠江三角洲汽油车尾气排放最主要的成分是:甲苯、苯、间/对-二甲苯、1,2,4-三甲基苯和异戊烷^[8]。本研究中甲苯、间/对-二甲苯和苯的占比也较高,但三甲苯占比很少。这可能与不同地区的机动车组成、油品的差异、不同海拔高度对车辆排放的影响和不同的测量方法有关。例如,之前的文献报道的油品为国Ⅱ~Ⅳ,而本研究中的油品已升级为国Ⅵ,品级更高。从分类上看,拉萨汽油车尾气排放中烷烃占比最大(48.1%),其次是OVOC(16.2%)。这与区家敏等^[8]报道的轻型

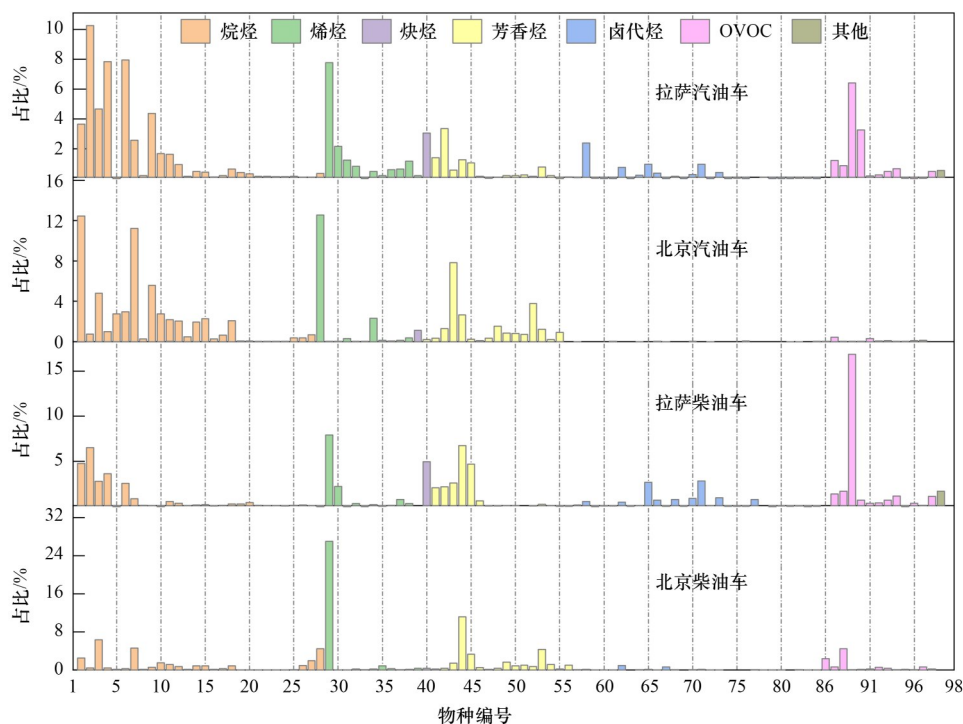
汽油车尾气以芳香烃为主(51.5%)、烷烃其次的组成特征不同。从C数上看,C2化合物体积分数最高,C5~C6质量分数最高,而北京测的汽油车尾气C2化合物质量分数最高^[21]。

2.1.3 柴油挥发

体积分数前10种化合物为:2,3-二甲基丁烷(6.4%),正丁烷(6.1%)、1,1,2-三氯乙烷(6.0%)、MTBE(4.9%)、甲基环己烷(4.8%)、正辛烷(4.2%)、甲苯(3.9%)、正壬烷(3.7%)、正戊烷(3.2%)和2-甲基庚烷(3.1%);质量分数高的化合物有:1,1,2-三氯乙烷(8.4%)、2,3-二甲基丁烷(5.8%)、正辛烷(5.0%)、甲基环己烷(5.0%)、MTBE(4.6%)和甲苯(3.8%)。相对于汽油挥发,柴油挥发组分中具有更高的烷烃占比(60.0%),且高碳组分占比高,如正十一烷的质量分数为3.1%。这与Liu等^[22]用顶空法测得的柴油挥发组分中有较高比例的高碳烷烃(正庚烷和甲基环己烷等)的结果一致,但其测得的柴油挥发中苯的占比远高于甲苯,而本研究中甲苯占比高于苯,这可能与油品的升级,苯含量降低有关。

2.1.4 柴油车排放

体积分数较高的化合物有:丙酮(15.3%)、乙醛(9.4%)、乙烯(7.2%)、间/对-二甲苯(6.1%)、丙烷(5.9%)、乙炔(4.5%)、乙烷(4.3%)、邻-二甲苯(4.3%)、正丁烷(3.3%)和1,2-二氯乙烷(2.6%)等;质量分数较高的化合物有:丙酮(13.3%)、间/对-二甲苯(9.7%),邻-二甲苯(6.8%)、乙醛(6.2%)、丙烷(3.9%)、1,2-二氯乙烷(3.8%)、乙苯(3.7%)、二氯甲烷(3.1%)、乙烯(3.0%)和正丁烷(2.9%)等。笔者在北京测得柴油车尾气主要成分为:乙烯(22.1%)、间/对-二甲苯(9.1%)、异丁烷(5.2%)、正戊烷(3.8%)、十一烷(3.7%)、丙酮(3.6%)、1,2,4-三甲苯(3.5%)、邻-二甲苯(2.7%)、乙烷(2.0%)和丙烯醛(2.0%)。二者相比,拉萨柴油车尾气中OVOC组分占比更高(31.4%),但奎烷、十一烷以及C9以上的芳香烃占比低。Liu等^[22]测得的柴油车尾气排放中,乙烯(9.0%)、丙烯(10.4%)、C7~C12烷烃(20.0%)和苯系物(BTEX, 15.0%)占比较高,但在拉萨柴油车排放中丙烯(2.0%)及C7~C11烷烃(1.9%)占比低。低的烯烃占比也明显不同于Dong等^[23]的结果,后者报道的柴油车排放中烯烃的占比高于50%,这一方面与油品升级烯烃含量的降低有关,另一方面,高原低氧的环境造成的燃烧不充分也有一定影响。高的OVOC占比与Yao等^[24]在北京道路测得的柴油车排放的结果相似。不过,在后者的结果中,甲醛占比最高,其次是乙醛。本研究没有对甲醛测量,测得的丙酮占比最高。其他组成较高的乙醛、乙烯、



物种编号 1~98 分别为:乙烷、丙烷、异丁烷、正丁烷、环戊烷、异戊烷、正戊烷、2,2-二甲基丁烷、2,3-二甲基丁烷、2-甲基戊烷、3-甲基戊烷、正己烷、2,4-二甲基戊烷、甲基环戊烷、2-甲基己烷、环己烷、2,3-二甲基戊烷、3-甲基己烷、2,2,4-三甲基戊烷、正庚烷、甲基环己烷、2,3,4-三甲基戊烷、2-甲基庚烷、3-甲基庚烷、正辛烷、正壬烷、正癸烷、十一烷、乙烯、丙烯、反-2-丁烯、正丁烯、顺-2-丁烯、1,3-丁二烯、1-戊烯、反-2-戊烯、异戊二烯、顺-2-戊烯、1-己烯、乙炔、苯、甲苯、乙苯、间/对-二甲苯、邻-二甲苯、苯乙烯、异丙苯、正丙苯、3-乙基甲苯、4-乙基甲苯、1,3,5-三甲苯、2-乙基甲苯、1,2,4-三甲苯、1,2,3-三甲苯、1,3-二乙基苯、1,4-二乙基苯、氟利昂 114、氯甲烷、氯乙烷、溴甲烷、氯乙烷、氟利昂 11、1,1-二氯乙烷、氟利昂 113、二氯甲烷、1,1-二氯乙烷、顺-1,2-二氯乙烯、氯仿、1,1,1-三氯乙烷、四氯化碳、1,2-二氯乙烯、三氯乙烯、1,2-二氯丙烷、二氯溴甲烷、反-1,3-二氯丙烯、顺-1,3-二氯丙烯、1,1,2-三氯乙烷、四氯乙烯、1,2-二溴乙烷、氯苯、1,3-二氯苯、1,4-二氯苯、苯基氯、1,2-二氯苯、乙醛、丙醛、丙酮、甲基叔丁基醚、异丁烯醛、正丁醛、甲基乙烯基酮、甲乙酮、2-戊酮、正戊酮、3-戊酮、正己醛和乙腈

图1 北京和拉萨机动车排放 VOCs 成分谱对比

Fig. 1 Comparison of VOCs composition profiles of motor vehicle emissions between Beijing and Lhasa

乙炔和二甲苯等占比较为接近。

2.1.5 LPG 挥发

按体积分数, 占比较高的化合物有丙烷 (59.9%)、异丁烷 (17.0%) 和正丁烷 (14.5%); 按质量分数, 丙烷 (52.9%)、异丁烷 (19.8%) 和正丁烷 (16.9%) 这 3 种化合物占 89.5%。这与平原地区的研究结果一致^[21,22]。

2.1.6 燃香排放

按体积分数, 乙烯 (15.2%)、乙醛 (12.1%)、丙烯醛 (11.4%)、乙烷 (10.1%)、丙酮 (8.4%)、丙烯 (7.7%)、1,3-丁二烯 (5.5%)、乙腈 (4.1%)、丙烷 (2.8%) 和甲基乙烯基酮 (2.4%) 等组分占比较高; 按质量分数, 丙烯醛 (12.8%)、乙醛 (10.8%)、丙酮 (9.8%)、乙烯 (8.6%)、丙烯 (6.6%)、乙烷 (6.2%)、1,3-丁二烯 (6.0%)、乙腈 (3.4%)、甲基乙烯基酮 (3.3%) 和丙烷 (2.5%) 占比较高。燃香排放中有很高比例的 OVOC 和烯烃。乙烯、乙炔、苯、丙烯和乙腈被认为是生物质燃烧排放 VOCs 的主要组分^[4]。本研究燃香排放的乙烯和丙烯也是重要组分, 但乙炔的体积分数和质量分数分别仅为 1.7% 和 0.9%,

列第 15 和 21 位。作为生物质燃烧示踪剂的乙腈^[25,26], 在燃香中体积和质量分数分别为 4.1% 和 3.4%。香炉闷烧会产生有毒的挥发性有机化合物, 如苯、1,3-丁二烯、乙醛、呋喃、苯乙烯、氯仿、甲苯和二甲苯等^[27]。本研究中燃香未采用焖烧, 但 1,3-丁二烯的占比也较高。异戊二烯体积分数和质量分数分别为 1.5% 和 2.1%, 是不可忽视的组分。焚香是甲醛和呋喃以及 BTEX 的重要来源^[28]。本研究未测量甲醛, 但燃香排放中较高的乙醛和丙烯醛与文献^[29]一致。此外, 燃香排放中 OVOC 和烯烃的占比较高, 不同于 Lu 等^[30]报道的燃香中烷烃和芳香烃占比高, 这可能与香的组成成分及燃烧条件有关。

2.2 不同排放源 VOCs 排放特征比较

2.2.1 主要物种及示踪物

识别不同排放源的特征物种对评估源的贡献非常重要。然而, 各类同源排放的 VOC 物种往往有所重叠^[22]。总体来看, 拉萨汽油挥发中 1,2,4-三甲苯占比最高, 其次是甲苯和异戊烷; 汽油车排放中异戊烷是重要的特征组分, C10 以上烷烃占比少; 柴油挥发中 C7 以上烷烃占比相比汽油挥发有明显增

加,且醛酮类化合物占比高.MTBE在柴油挥发及汽油车排放中均有一定的占比,但在柴油车排放中较低,可作为汽油车排放的示踪剂.LPG以C3~C4烷烃占有绝对优势.燃香排放中,醛酮类化合物占比高,乙腈是重要的示踪物种.异戊二烯是生物质燃烧排放的示踪剂,在夜间可区分没有植物排放的情况下的秸秆和垃圾燃烧^[15].本研究中燃香也是异戊二烯的重要来源,可作为夜间没有植物排放的异戊二烯溯源的根据.

2.2.2 特征比值分析

不同物种的比值可提供有用的VOCs源的信息^[31].VOCs组分配对,如正丁烷和异丁烷,甲苯和苯,乙苯和间/对-二甲苯,在环境测量中经常有很好的相关性^[32],意味着相同或相近的来源.基于物种的反应性差异,一些物种的比率可用来估计气团的光化学年龄^[22,33].

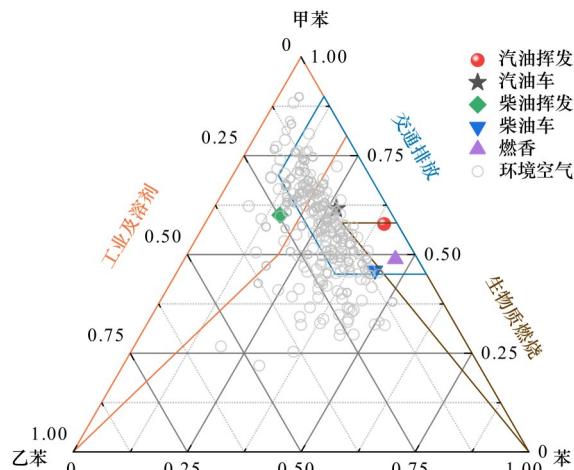
甲苯与苯的比值(T/B)常用来判断VOCs来源^[34~36].城市机动车排放,T/B一般在2~3之间^[37,38],工业区环境大气中比值为6~7^[39,40],溶剂涂料行业中的T/B>10^[41].本研究中汽油挥发、汽油车、柴油挥发、柴油车和燃香的T/B分别为1.5,2.4,4.0,1.1和1.1.拉萨汽油挥发及汽油车排放中具有较高的T/B,与以往研究一致;柴油挥发的T/B较高,而平原地区苯高于甲苯^[22];柴油车排放及燃香排放的T/B较低,苯的占比相对更高;燃香排放的T/B>1,不同于Silva等^[29]获得的小于1的T/B(0.17~0.50).异戊烷与正戊烷比值(0.06~1.46)可用于区分生物质燃烧和以汽油为主的交通与化石燃料燃烧排放(2.83~4.13)^[15].本研究中汽油车排放的异戊烷/正戊烷为3.1,汽油挥发为5.0,燃香为0.5,这与文献^[15]一致.汽油挥发中的异戊烷/正戊烷最高,而燃香排放中异戊烷占比低于正戊烷,可以作为区分化石燃料及交通排放与生物质燃烧的判断依据.

2.2.3 BTE三元图分布

Zhang等^[39]从文献^[22]中总结了不同排放源中苯、甲苯和乙苯的比值关系,并绘制三元图,如图2所示.可见,拉萨市的汽油挥发和汽油车排放位于交通排放的区域内,而柴油车排放位于交通排放区域的边界,柴油挥发则不在交通排放区域而是落在工业及溶剂区域内;燃香排放落于交通排放和生物质燃烧的公共区域内.尽管环境空气测量数据绝大部分落在了交通排放的区域,但仅用三元图分析显然会低估了柴油挥发以及柴油车排放的贡献.

2.3 不同VOCs排放源的物种活性贡献

不考虑化学反应活性仅考虑排放量的VOCs减排政策,会影响VOCs减排对改善空气质量的效



不同颜色实线指示交通排放、生物质燃烧、工业和溶剂排放特征比值集中的区域;不同形状的彩色实心点是本研究的结果,浅灰色空心圆为2021年同月拉萨环境空气的监测数据^[42]

图2 苯、甲苯和乙苯比例三元图

Fig. 2 Ternary diagram of the ratio of benzene, toluene, and ethylbenzene

果^[43,44].具有较高MIR值的芳香烃、烯烃和OVOCs组分,由于其高的化学反应活性,在排放量较低的情况下却有较高的臭氧生成潜势^[6].

图3是以有机物类别计算不同排放源VOCs的体积分数、质量分数、对 L_{OH} 、OFP和SOAP的贡献.烯烃化合物占比低,但反应活性较高,在各排放源中,相比体积分数和质量分数,对 L_{OH} 的贡献均有较大提升,而烷烃贡献均有所下降;同样,芳香烃对OFP贡献也明显提高,特别在汽油挥发、柴油挥发及柴油车排放中占绝对优势,而在汽油车排放和燃香中,烯烃对OFP贡献最大.已有报道表明,烯烃和芳香烃是城市空气中起主导作用的活性VOC物种,而机动车排放是大气反应活性的最大贡献者^[45,46].Guo等^[10]于2019年在拉萨以及本研究的结果均表明,烯烃和芳香烃是拉萨大气OFP的主要贡献者.从C数上来看(图略),汽油挥发和柴油挥发中以C6~C9化合物贡献为主;相对于汽油车排放,柴油车排放的高碳化合物对OFP的贡献更大.按碳数对 L_{OH} 贡献来看,汽油车以C5~C8物种为主,柴油车中C8化合物贡献最大,而燃香中以C2~C4为主.拉萨典型排放源的SOAP均以芳香烃为主,柴油挥发及柴油车排放中C8~C9的贡献比其他化合物更高.

3 结论

(1)通过分析拉萨市6种排放源的VOCs组成发现,汽油挥发中1,2,4-三甲苯占比最高,而汽油车尾气中异戊烷、正丁烷和丙烷的占比高,1,2,4-三甲苯占比较低.这与在北京测的汽油挥发中异戊烷占比最高和珠三角地区汽油车尾气1,2,4-三甲苯质

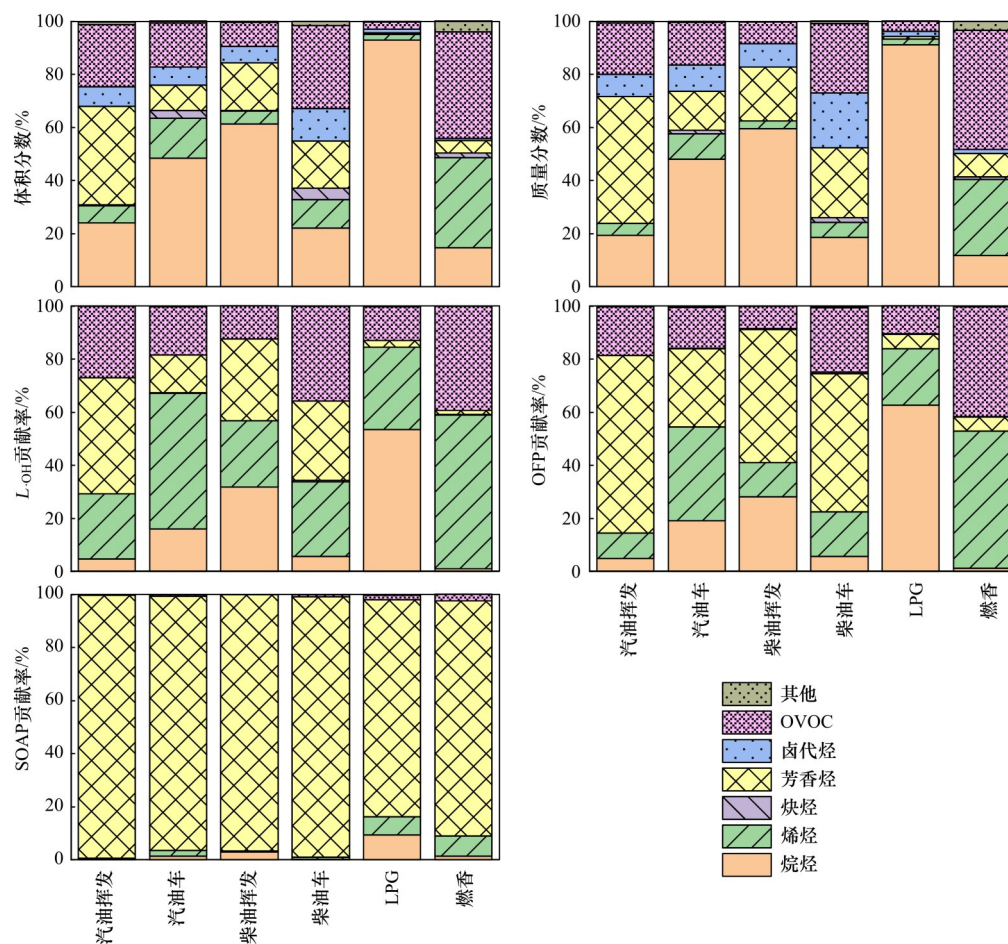


图3 拉萨各排放源中各类化合物的体积分数、质量分数及对 L_{OH} 、OFP 和 SOAP 的贡献

Fig. 3 Volume fraction, mass fraction and contribution to L_{OH} , OFP, and SOAP of various compounds from each emission source in Lhasa

量分数占比高不同. 柴油挥发比汽油挥发及汽油车尾气排放更高占比的 C7 以上高碳化合物, 且甲苯占比高于苯, 与平原地区柴油挥发中苯的占比远高于甲苯的结果不同. 柴油车排放中 OVOC 占比最高, 不同于平原地区丙烯和 C7 以上烷烃高占比的特征. MTBE 是汽油车排放的示踪剂, 在汽油车排放中占比明显高于柴油车, 而在 LPG 及燃香排放中基本没有. 高原与平原地区 LPG 挥发的 VOCs 一致, 主要是丙烷、丁烷. 燃香排放的乙烯、乙醛和丙烯醛占比高, 乙腈是区别于其他排放源的示踪物种, 异戊二烯占比虽少, 但不容忽视.

(2) 从 T/B 上来看, 汽油挥发、汽油车及柴油车的结果与平原地区的结果基本一致; 燃香的 T/B > 1, 这与平原地区生物质燃烧具有较低的 T/B 不同. 柴油挥发在 BTE 三元图上处在机动车排放区域外, 而更接近于工业及溶剂排放比值, 在后续研究中应加以重视. 烯烃化合物在不同排放源中占比低, 但因为较高的反应活性, 对 L_{OH} 和 OFP 贡献大. 各排放源中芳香烃对 SOAP 贡献均大于 80%, 这在柴油车排放中尤为突出. 在高原城市中, 烯烃和芳香烃同样是城市空气中起主导作用的活性 VOCs.

(3) 初步研究结果表明, 高原城市拉萨典型排放源的成分谱特征与平原地区有所差异, 需要进一步对 VOCs 排放源进行研究, 以应对潜在的光化学污染风险并有助于 O_3 污染控制.

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