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不同水源补给河流浮游植物群落结构特征及其与环境因子的关系

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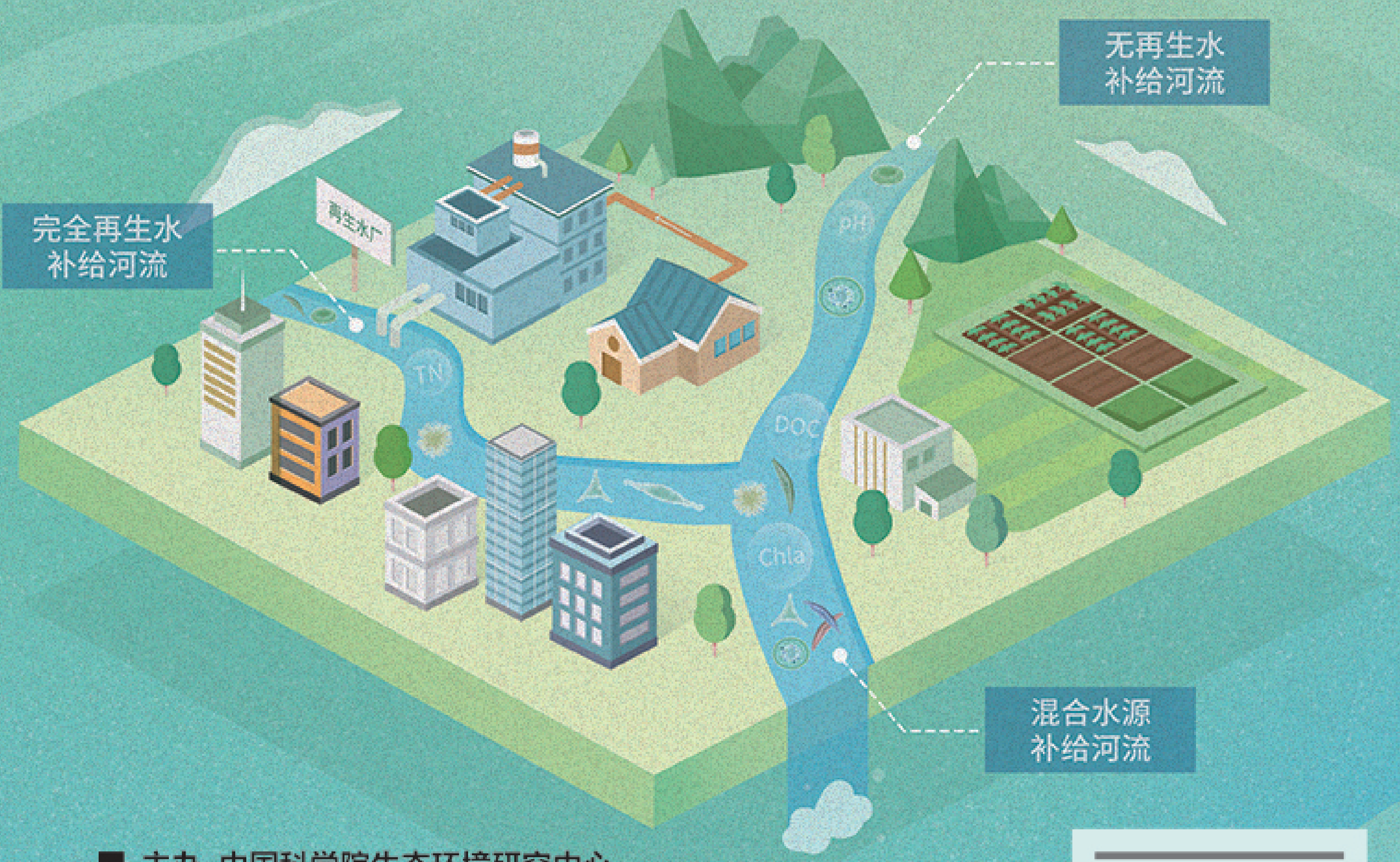

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缸内直喷汽油车颗粒物化学组分特征

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摘要: 随着缸内直喷汽油车(GDI)的大量使用,其尾气颗粒物排放问题日益凸显.研究缸内直喷汽油车尾气颗粒物构成组分是有效控制其颗粒物排放的重要前期工作之一.选取了5辆满足国六排放标准的缸内直喷汽油车进行了尾气颗粒物碳质成分、水溶性离子和多环芳烃(PAHs)组分分析.结果发现,各类碳质是测试车辆尾气颗粒物中的主要成分,平均占比约70%.颗粒物碳质中有机碳质(OC)多于核态碳质(EC),OC/EC比值在1.03~3.43之间.测量了颗粒物中多种水溶性离子的含量,结果表明 Ca^{2+} 和 SO_4^{2-} 是尾气颗粒物中的主要水溶性离子,主要来源均为机油添加剂.此外,结果还发现GDI车辆尾气颗粒物中高环PAHs排放占比较高,对人体健康危害大,需要重点关注.

关键词:轻型车; 缸内直喷汽油机; 尾气颗粒物; 碳质; 水溶性离子; 多环芳烃
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Chemical Characterizations of Particles from Direct-injection Gasoline Vehicles

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Abstract: With the widespread use of direct-injection gasoline vehicles, their tailpipe particulate emissions are becoming a serious problem for the urban atmosphere. The study of tailpipe particulate components from direct-injection gasoline vehicles is an important preliminary work to control particulate emissions effectively. In this study, five direct-injection gasoline vehicles meeting the China-6 emission standard were tested to analyze the carbonaceous components, water-soluble ions, and polycyclic aromatic hydrocarbons of tailpipe particulate emissions. It was found that carbonaceous substances were the major components of exhaust particles, accounting for approximately 70%. More organic carbon than elemental carbon was found in the particulate emissions of direct-injection gasoline vehicles, with OC/EC ratios of 1.03 to 3.43. The study also measured various water-soluble ions of particles. The total mass proportions of water-soluble ions were 11.3% to 17.6%. Ca^{2+} and SO_4^{2-} were the main water-soluble ions, with oil additives being the main source of these two ions. In addition, it was found that the PAHs emissions were significantly reduced with stricter regulations. PAHs emission factors of GDI vehicles meeting the China-6 standard were reduced by more than half of the emission factors of China-6-standard GDI vehicles. Moreover, GDI vehicles had a high proportion of high-cyclic PAHs emissions in exhaust particles, which damages public health and requires more attention.

Key words: light-duty vehicles; direct-injection gasoline engines; particles; carbonaceous substances; water-soluble ions; polycyclic aromatic hydrocarbons

$PM_{2.5}$是城市中重要的大气污染物之一,对大气环境和人体健康具有严重危害[1~3].机动车尾气排放是市区大气颗粒物的主要来源之一[4~7].为了继续强化 $PM_{2.5}$ 治理,降低市区机动车尾气中的 $PM_{2.5}$ 浓度,充分了解机动车颗粒物的构成组分是必要的前提条件之一.

目前许多国家及地区都针对当地机动车尾气颗粒物进行了组分分析.但由于试验条件及车辆排放水平的差异,不同地区不同时期机动车颗粒物排放结果差异较大[8~12]. Fujita等[13]在2007年研究了美国加州南部地区多辆轻型汽油车排放的颗粒物组分.结果表明碳质(TC)在颗粒物总质量中占比超过95%,且有机碳质(OC)是进气道喷射车辆排放颗粒物的主要成分. Fushimi等[14]使用日本JC08测试循环采集测量了4辆缸内直喷汽油车和一辆进气道喷射汽油车尾气颗粒物的化学组分.有研究发现TC在颗粒物总质量中平均占比约90%,水溶性离子和金属元素占比为2%~14%.核态碳质(EC)是4辆缸内直喷车辆尾气颗粒物的主要成分,且大部分来源于燃料不完全燃烧.

Yang等[15]采用新欧洲行驶循环(NEDC)对中国台湾地区4辆轻型汽油车尾气颗粒物化学成分进行了分析,其研究表明,颗粒物中总碳质平均排放为0.873 $mg \cdot km^{-1}$,OC/EC平均比值为2.29,水溶性离子平均排放为25.8 $\mu g \cdot km^{-1}$. Hao等[16]针对多辆国三及之前排放标准的轻型汽油车开展了尾气颗粒物化学组分分析,有机碳质在总颗粒物质量中占比为41%~64%,OC/EC平均比值为6.08,明显高于相同排放标准的轻型柴油车. Hao等[17]此后还对国四和国五排放标准的轻型汽油车尾气颗粒物进行了组分分析,结果表明随着排放法规的加严,颗粒物排放总质量下降,但水溶性离子和金属元素的占比有所上升.

此外,机动车尾气颗粒物中的多环芳烃类(PAHs)组分因具有高致癌性、致畸性和诱变毒性而受到特别关注[18~20].有研究对包括中国在内的多个地区的机动车尾气颗粒PAHs组分排放进行了相

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关测量研究[21~25].

随着机动车排放法规的加严和车辆排放控制技术的更新,市区范围内柴油车颗粒物排放明显减少,对市区颗粒物总排放贡献降低[26~31]. 同时,为了减少机动车燃油消耗,缸内直喷汽油车数量逐年增加. 缸内直喷汽油车相较传统进气道喷射车辆更易产生颗粒物排放,对市区颗粒物排放的贡献开始凸显[17,32,33]. 由于喷油方式相近,随着柴油机缸内喷射压力的提升,缸内直喷汽油车与柴油车缸内燃烧生成颗粒物具有许多相似的特征:颗粒物中核态碳质的比例较高,排放的总颗粒物质量偏低但颗粒物数量较高,颗粒物平均粒径较小[15,34,35]. 因此为了控制缸内直喷汽油车尾气颗粒物的排放,当前采用了与柴油车相似的技术路线,即进行缸内燃烧优化的同时使用尾气后处理装置(颗粒物拦截过滤装置)[36~38].

但目前针对满足国六排放标准的缸内直喷汽油车的颗粒物组分分析尚不充分,制约了我国现阶段本土化排放因子库的开发和排放清单的计算精度. 本研究针对 5 辆满足国六排放标准的缸内直喷汽油车进行了尾气颗粒物组分分析,以期为进一步降低颗粒物排放提供数据基础.

1 材料与方法

1.1 测试设备

图 1 为本试验的测试采样系统. 驾驶员依 GB 18352.6-2016 要求驾驶测试车辆在底盘测功机上运行全球轻型车测试循环(WLTC). 测试车辆的尾气全部排入全流稀释定容取样系统(CVS)与洁净空气进行稀释混合. 部分定容稀释后气体进入颗粒物质量(PM)测量采样托架并使用 47 nm 规格石英纤维滤纸进行尾气颗粒物采集. 采样前,石英纤维滤纸需在 550℃高温下灼烧 5.5 h 以去除其有机物,后密封避光保存.

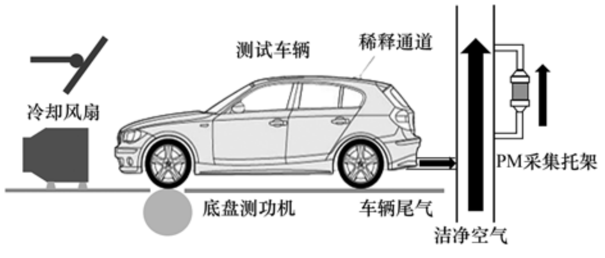

图 1 测试采样系统

Fig. 1 Test and sampling system

将称重后的石英滤纸取 1 cm^2 小样,使用 DRI Model 2001A 碳分析仪,依 IMPROVE A 协议规定的热光反射法(TOR)测量 OC/EC 比例和具体重量. 剩余滤纸加 15 mL 去离子水后超声提取 20 min,提取液经微孔滤膜过滤后使用 Thermofish Dionex 5000 + 型离子色谱仪进行颗粒物水溶性离子分析,其中阳离子分析柱为 Dionex Ion Pac CS12A,阴离子色谱柱为 Dionex Ion Pac AS11. 阳离子的检出限为 0.01 ~ 0.02 $\mu g \cdot mL^{-1}$,相对标准偏差小于 1.5; 阴离子的检出限为 0.01 ~ 0.1 $\mu g \cdot mL^{-1}$,相对标准偏差小于 3.

另有部分测试车辆称重后的石英滤纸被剪成 4 份,放入 20 mL 棕色样品瓶中,加入 10 mL 甲苯,在 60℃下超声萃取 60 min,再将萃取液浓缩定容至 2 mL 待测. 采用安捷伦公司 6890-5975C 型气相质谱分析仪(GC-MS)对美国 EPA-13A 方法中规定的 16 种 PAHs 进行定量分析,方法检出限为 2.5 $\mu g \cdot mL^{-1}$,仪器相对标准偏差小于 2.

1.2 测试车辆

本试验共选用 5 辆满足国六排放标准的缸内直喷汽油车,其中车辆 V1、V3 和 V4 采用增压进气方式,车辆 V2 和 V5 采用自然进气方式,仅车辆 V3 在尾气后处理系统配备了颗粒捕集器(GPF). 车辆进行试验时均采用符合国六标准的 92 号市售汽油. 试验对车辆 V1、V2 和 V3 采集的尾气颗粒物进行碳质分析和水溶性离子含量分析,对 V4 和 V5 的尾气颗粒物进行 16 种 PAHs 的含量分析. 5 辆测试车辆的具体信息如表 1 所示.

表 1 测试车辆主要技术参数

Table 1 Parameters of test vehicles

技术参数	V1	V2	V3	V4	V5
发动机排量/L	1.4	1.6	2.0	1.4	1.6
进气增压	是	否	是	是	否
后处理装置	TWC	TWC	TWC + GPF	TWC	TWC
最大功率/kW	116	94	160	116	94
压缩比	10.5	13	10	10.5	13
整车质量/kg	1 340	1 270	2 055	1 506	1 411
行驶里程/km	566	3 610	7 321	408	785

1.3 测试循环

5 辆测试车辆均使用常温冷启动 WLTC 循环进行测试. WLTC 是法规认证循环,由低速段、中速段、高速段和超高速段共 4 个部分组成. WLTC 平均速度为 46.08 $km \cdot h^{-1}$,总行驶里程共 23.26 km,最高车速达到 131.3 $km \cdot h^{-1}$,其中加速工况比例为 35.06%,循环总耗时1 800 s.

2 结果与讨论

图 2 为车辆 V1 ~ V3 的尾气颗粒物各组分排放因子. V1 ~ V3 在 WLTC 循环 PM 总排放因子分别为 1.60、1.42 和 0.99 $mg \cdot km^{-1}$,都远低于轻型车国六阶段 PM 排放限值为 3.00 $mg \cdot km^{-1}$的法规要求. 碳

质是3辆测试车辆尾气颗粒物中的主要成分,尽管由于发动机及后处理技术不同,不同车辆TC排放因子差异较大(0.67~1.59 mg·km^{-1}),但TC在颗粒物质量中占比均达到约70.0%(67.7%~75.1%).此外,不同车辆TC中OC和EC占比差异较大,V1尾气颗粒物质量中OC占比为58.2%,EC占比为16.9%,OC/EC比值为3.43. V2和V3尾气颗粒物中OC占比分别为34.6%和38.7%,OC/EC比值分别为1.03和1.34,小于V1的结果. V1尾气颗粒物中的OC成分主要是沸点低于280℃的有机碳质,可能由汽油燃料的不完全氧化燃烧形成的挥发性/半挥发性有机物冷凝形成[35,39].

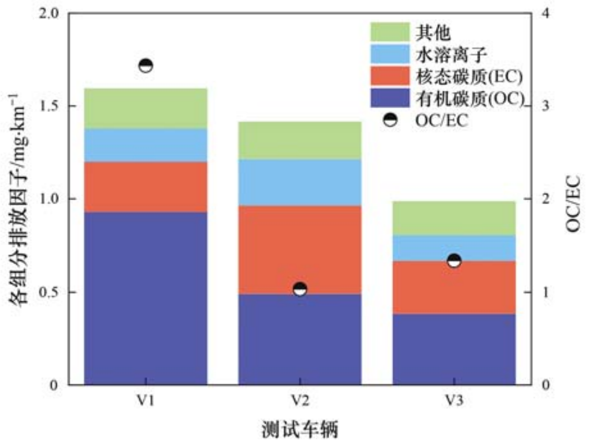

图2 测试车辆颗粒物组分排放因子

Fig. 2 Emission factors of PM components from test vehicles

3辆缸内直喷汽油车的各种碳质排放因子与其他研究的比较结果如表2所示.由于车辆使用燃料、排放标准和采样测试循环的不同,各研究中车辆平均排放因子差异较大,OC/EC比值在0.27~8.9区间内.此前有研究表明GDI车辆尾气颗粒物中OC/EC比值小于进气道喷射汽油(PFI)车辆.相关研究试验车队中因同时混有PFI和GDI车辆,OC/EC平均比值分布范围较大.当试验车队中PFI车辆比例较高时,OC/EC平均比值即明显高于本研究中GDI测试车辆的OC/EC平均比值.此外,表2中柴油车OC/EC比值较缸内直喷汽油车OC/EC比值更小,均小于1.采用缸内喷射燃料并混合的GDI车辆和柴油车辆,由于燃料和空气不能及时充分混合,常存在过浓区域,在燃烧时不易完全燃烧,从而生成包含大量核态碳质的未完全燃烧产物.

表2 相关研究颗粒物组分排放因子1)

Table 2 Components of PM in related studies

TC	OC	EC	水溶性离子	车辆排放标准	测试循环	燃料	文献
28.76 mg·mi^{-1}	24.13 mg·mi^{-1}	4.63 mg·mi^{-1}	—	—	UDC2)	汽油	[14]
0.37 mg·m^{-1}	0.20 mg·m^{-1}	0.17 mg·m^{-1}	—	Euro-33)	FTP4)	汽油	[40]
1.49 mg·km^{-1}	1.03 mg·km^{-1}	0.46 mg·km^{-1}	196.11 μg·km^{-1}	Euro-3	NEDC	汽油	[15]
1.29 mg·km^{-1}	1.16 mg·km^{-1}	0.13 mg·km^{-1}	约130 μg·km^{-1}	国四和国五	西安循环5)	汽油	[17]
0.74 g·km^{-1}	0.27 g·km^{-1}	0.47 g·km^{-1}	2.74 mg·km^{-1}	国三和国四	PEMS6)	柴油	[41]
50.4 mg·km^{-1}	10.8 mg·km^{-1}	39.6 mg·km^{-1}	154.49 μg·km^{-1}	—	NEDC	柴油	[15]

1)单位中mi表示英里(1 mi=1.609 km),“—”表示文献中没有相关数据; 2)NEDC循环市区段部分; 3)欧洲轻型车第三阶段排放标准; 4)美国联邦认证程序; 5)依据西安市区路段轻型车驾驶特征制定的驾驶循环; 6)便携式排放测试系统

图2还展示了3辆测试车辆尾气颗粒物中水溶离子的排放因子及占比. V1~V3尾气颗粒物中水溶性离子总排放因子分别为0.18、0.25和0.14 mg·km^{-1},质量分数达到11.3%~17.6%.其中各种水溶离子的排放因子如图3所示.在所测量的5种阳离子(Na^+、NH_4^+、K^+、Mg^{2+}和Ca^{2+})中4种金属离子都是机油特征离子[42~45],Ca^{2+}作为机油分散剂的重要组分,平均排放因子最大,为28.91 μg·km^{-1}. Na^+和NH_4^+的平均排放因子小于Ca^{2+},分别为12.23 μg·km^{-1}和11.09 μg·km^{-1}. Mg^{2+}在颗粒物中的含量最少,平均排放因子仅为2.56 μg·km^{-1}.在所测量的4种阴离子中(F^-、Cl^-、NO_3^-和SO_4^{2-}),SO_4^{2-}的平均排放因子远高于其余水溶性阴离子,为103.13 μg·km^{-1}.这是由于除机油中含有作为防氧化剂成分的硫元素外,汽油中也含有微量的硫元素,从而导致颗粒物水溶性阴离子中硫酸根占比最大.其余3种水溶性阴离子排放因

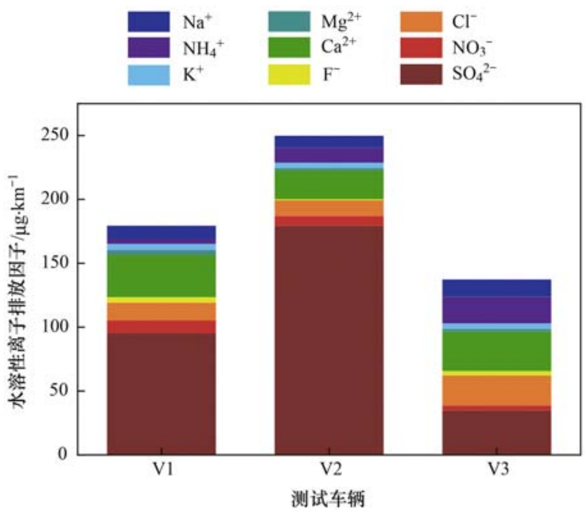

图3 测试车辆颗粒物中水溶离子排放因子

Fig. 3 Emission factors of water-soluble ions from test vehicle particles

子相对较小，F^-、Cl^-和NO_3^-的平均排放因子分别为2.93、16.51和7.18 μg·km^{-1}.

试验车辆 V4 和 V5 尾气颗粒物，依 EPA-13A 方法，对16种PAHs进行定量分析. 如图4所示，采集到的颗粒物样品中一共检测出其中8种PAHs，分别为萘(Nap)、菲(Phe)、蒽(Ant)、苯并[a]蒽(BaA)、苯并[b]荧蒽(BbF)、苯并[a]芘(BaP)、茚并[1,2,3-cd]芘(InP)和苯并[ghi]苝(BghiP).

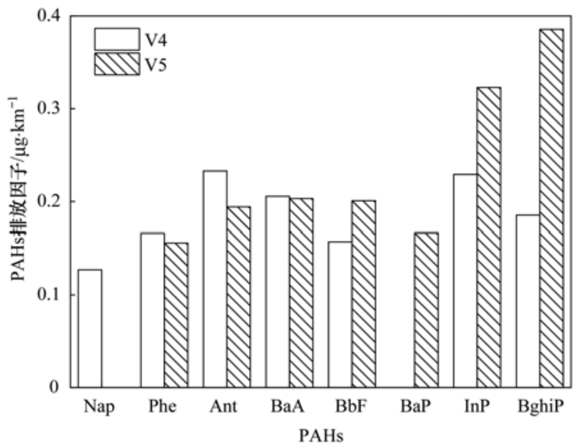

图4　测试车辆颗粒物中PAHs排放因子

Fig. 4　Emission factors of PAHs from test vehicle particles

V4 尾气颗粒物检测到的所有 PAHs 总排放因子为1.30 μg·km^{-1}，其中 Ant 和 InP 的排放因子最大，均为0.23 μg·km^{-1}，BaA 的排放因子稍小，为0.21 μg·km^{-1}. V5 尾气颗粒物检测到的所有 PAHs 总排放因子为1.63 μg·km^{-1}，其中 BghiP 排放因子最大，为0.39 μg·km^{-1}，InP 的排放因子仅小于 BghiP，为0.32 μg·km^{-1}. 表3为相关试验研究得到的不同类型车辆尾气颗粒物中 PAHs 排放因子. 对比结果可知，随着车辆尾气排放控制技术的提升，PAHs 的总排放量亦随之明显降低，相较于国五及之前排放水平的 GDI 车辆，本研究中满足国六标准的 GDI 车辆总 PAHs 的排放因子降低一半以上. 但采用不同排放标准和测试循环得到的 PAHs 排放结果差异较大. 同时由表3满足相同机动车尾气排放控制标准的轻型车使用汽油和柴油排放的 PAHs 不会产生超过一个量级的明显差异.

表3　相关研究颗粒物中PAHs含量

Table 3　PAHs of PM in related studies

PAHs含量/μg·km^{-1}	车辆排放标准	喷油方式	测试循环	燃料	文献
1.54	Euro-5	GDI	CADC-Cold Urban1)	汽油	[8]
1.10	Euro-5	GDI	CADC-Motorway2)	汽油	
2.46	国四	PFI	WLTC	汽油	[46]
5.78	国四	GDI	WLTC	汽油	
2.72	国五	GDI	NEDC	汽油	
3.88	国五	GDI	WLTC	汽油	
0.79	LEV Ⅲ SULEV303)	GDI	LA924)	汽油	[47]
0.92	LEV Ⅱ5)	GDI	LA92	汽油	
3.52	Euro-4	缸内喷射	CADC	柴油	[33]
2.27	Euro-5	缸内喷射	CADC	柴油	

1)通用阿耳忒弥斯驾驶循环冷启动测试市区段；2)通用阿耳忒弥斯驾驶循环高速段；3)美国加州低污染汽车标准第三阶段；4)洛杉矶92驾驶循环；5)美国加州低污染汽车标准第二阶段

图5所示为 V4 和 V5 颗粒物中不同种类 PAHs 在总 PAHs 中的质量分数. 试验车辆 V4 尾气颗粒物中3环和6环的 PAHs 质量分数最大，分别为30.6%和31.8%. V5 尾气颗粒物中6环的 PAHs 质量分数最大，为54.4%. 对比两车结果可以发现，V4 排放的相对分子质量较小的 PAHs 较多，2环和3环 PAHs 排放总质量分数为40.3%；而 V5 排放的相对分子质量较大的 PAHs 较多，5环和6环 PAHs 排放总质量分数为82.5%. 相对分子质量较小的多环芳烃多由燃料的不完全燃烧产物加聚重组生成. 相对分子质量较大的多环芳烃部分由燃料未完全燃烧剩余的不饱和烃多次加聚重组生成，或由机油的未完全燃烧产生. 与此前研究结果相似的是 GDI 车辆颗粒物中的高环 PAHs 在总 PAHs 中质量分数大. 由于高环 PAHs 的毒性和致癌性更大，GDI 车辆的 PAHs

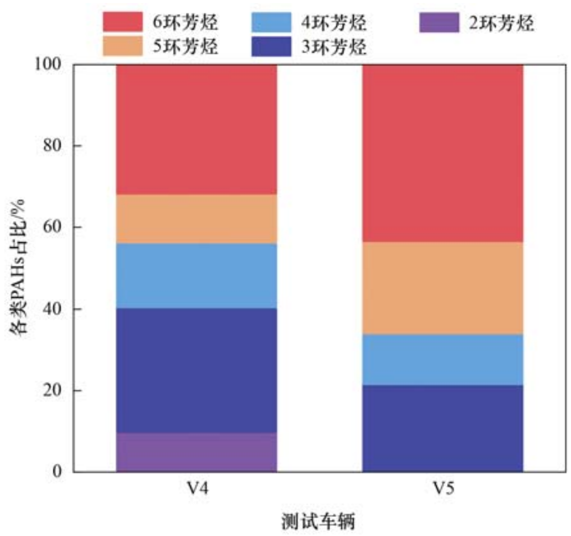

图5　测试车辆颗粒物中PAHs排放占比

Fig. 5　Emission percentages of PAHs from test vehicle particles

排放应受到更多的重视.

3 结论

(1)碳质(TC)是国六 GDI 车辆尾气颗粒物中的主要成分,平均占比约 70%,碳质中 OC/EC 比值在 1.03 ~ 3.43 区间.

(2)Ca^{2+} 和 SO_4^{2-} 是尾气颗粒物中的主要水溶性离子,主要来源均为机油添加剂.

(3)尾气颗粒物中 PAHs 排放质量随车辆排放标准的提升而降低,GDI 车辆高环 PAHs 排放占比高,对健康危害大,需重点关注.

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