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不同国家农用地土壤环境标准比较与启示
李勳之, 姜榕, 王国庆, 陈玉东, 龙涛, 林玉锁

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基于气团老化程度对挥发性有机物分类改善 PMF 源解析效果

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摘要: VOCs 作为臭氧与二次有机气溶胶的关键前体物, 其来源解析对臭氧和颗粒物的协同控制至关重要。但多数 VOCs 具有反应性, 不能完全满足受体模式对污染源排放化学物质组成稳定的假设要求, 导致受体模式解析结果不能精准反映实际源贡献。为解决因不同 VOCs 反应活性不同而导致的组分相对变化与模型假设不符的问题, 引入 VOCs 老化程度来表征其光化学反应的状态。将乌海市超级观测站监测的 VOCs 依据乙苯与间/对-二甲苯的比值分为高、中和低这 3 种老化状态。结果表明, 分类后解析结果的模拟值与实测值的回归方程参数(斜率和截距)、标准误差、决定系数和残差合格率等模型诊断参数改善明显。因为老化程度与气团在大气中的传输时间和大气氧化性密切相关, 在一定程度上也反映了气团的不同来源信息。在高老化态样品中焦化源贡献率最高, 达 47.20%; 在低老化态样品中燃烧源和焦化源均较高, 分别为 28.67% 和 24.39%。按老化程度进行分类后, PMF 对 VOCs 源解析的结果更符合实际排放源的贡献。

关键词: 挥发性有机物(VOCs); 源解析; 正交矩阵因子分解法(PMF); 老化特征; 受体模型

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Improved Performance of PMF Source Apportionment for Volatile Organic Compounds Based on Classification of VOCs' Aging Degree in Air Mass

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Abstract: VOCs are the key precursors of ozone and secondary organic aerosols. The results of source apportionment for VOCs are very important for the coordinated control of ozone and second organic particulate matter. However, VOCs do not fully meet the assumption of the receptor model because the VOCs released from each source are relatively unstable in the transmission process for their reactivity. As a result, we do not accurately obtain the actual source contribution when the receptor model is used for the source apportionment of VOCs. In order to solve the problem that the relative changes in the components caused by VOCs reactivity are not consistent with the PMF model hypothesis, the aging degree of VOCs was introduced to distinguish the state characteristics after their photochemical reactions in the ambient air. According to the ratio of ethylbenzene to m/p-xylene, VOCs monitored at Wuhai were divided into three aging states: high, medium, and low. The results showed that the model parameters, such as regression equation parameters (slope and intercept), standard error, determination coefficient, and pass rate of residual error, were improved obviously compared to the sample set after classification. Because the degree of aging is closely related to the transport time of air mass and the atmospheric oxidation in the atmosphere, it also reflects the different sources of air mass to some extent. In the high-aging VOCs samples, the coking source occupied a high proportion (up to 47.20%). In the low-aging VOCs samples, the combustion source and coking source accounted for a higher proportion, 28.67% and 24.39%, respectively. After the classification according to the aging degree, the results of VOCs source apportionment by PMF are more consistent with the actual contribution of emission sources.

Key words: volatile organic compounds(VOCs); source apportionment; positive matrix factorization(PMF); aging characteristics; receptor model

以细颗粒物($PM_{2.5}$)和臭氧(O_3)为代表的复合污染是我国当前面临的主要环境空气污染问题^[1,2]。作为 O_3 与 $PM_{2.5}$ 重要前体物的挥发性有机物(volatile organic compounds, VOCs)对区域空气质量^[3]和人体健康影响显著^[4-6],其来源解析是改善空气质量的基础。VOCs 源解析方法主要有扩散模型和受体模型,但扩散模型常因缺乏本地化高精度排放清单在应用中受到限制。正定矩阵因子分解法(positive matrix factorization, PMF)是在 VOCs 源解析中广泛应用的受体模型^[7-9],相比于其它受体模型能更好地识别源类^[10]。PMF 模型的基本假设之一为污染源排放的污染物种类和比例在传输过程中不变^[11,12],但 VOCs 在传输过程中必然伴随着光化学反应^[13],导致其物种和比例发生变化,使源解析结

果不能真实反映实际污染源贡献,学者们尝试多种方法解决这一问题。Wadden 等^[14]的研将究受体模型应用于烃类解析时,发现数据不完全符合成分相对稳定的假设,会对机动车和溶剂使用等污染源的判别产生一定影响,提出应在相对稳定的源-受体关系下,即尽量保证污染源成分相对稳定的条件下使用该模型。Vestenius 等^[15]的研究引入 VOCs 氧化产物试图减少因 VOCs 反应性对 PMF 解析结果的影响,并应用于荷兰针叶林环境 VOCs 的来源解析,其中氧化产物的不确定同样也会导致结果可能存在一

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定的问题. Liu 等^[16]的研究则排除部分高反应活性的非甲烷碳氢化合物(NMHCs),利用 PMF 对剩余 17 种进行解析,确定了北京冬季大气 NMHCs 的 5 类主要贡献源,但在排除高反应活性物种的同时也可能丢失了这些物种数据所包含的有用信息使解析结果出现偏差. Zheng 等^[17]的研究考虑了 VOCs 与 $\cdot\text{OH}$ 的反应对所测物种进入环境空气后受到光化学反应的影响,利用一级化学反应动力学方程定量描述每种污染源排放 VOCs 的反应活性,由于源排放的 VOCs 在大气中的传输和被氧化程度非常复杂,导致该方法很难确定 VOCs 初始排放参数,作者确定初始 X/E 百分比为 95%,解析结果同样存在不确定性. 除此之外,还有一些学者则考虑各物种与 $\cdot\text{OH}$ 的反应速率常数^[18,19]和物种间相关性^[20]等进行解析,但鲜有研究综合考虑污染物传输速度及大气氧化性等因素共同影响下的 VOCs 光化学反应状态,在保留尽可能多 VOCs 物种的前提下,对不同反应状态的 VOCs 进行解析.

本研究以表征反应活性的 VOCs 气团老化特性为依据,对乌海市 2019 年 9 月至 2020 年 3 月超级观测站 VOCs 小时监测数据进行分类. 对于分类后的 VOCs 子数据集而言,不同污染源排放特征的差异成为影响 VOCs 样品中物种浓度变化的最重要因素,从而更准确地判别 VOCs 的来源,以期对 VOCs 精准减排提供科学依据.

1 材料与方法

1.1 研究区域与采样点位

乌海市及周边煤炭资源丰富,工业以焦化和化工行业为主,辖区内及周边分布着多个焦化和化工工业园区, VOCs 排放情况复杂. 采样地点(39.66°N, 106.83°E)如图 1 所示,属于交通居住混合区,采样器架设距离地面约 15 m,在研究时段内主要受南风和西北风控制,周围无明显固定污染源.

1.2 数据来源

采用乌海市超级观测站 2019 年 9 月至 2020 年 3 月自动监测小时数据,其中 VOCs 采用北京博赛德科技有限公司的 BCT-7800A PLUS 挥发性有机物在线监测系统,即对捕集到的 VOCs 利用预浓缩系统(ENTCH7200CTS-C2)浓缩后使用安捷伦科技有限公司的 GC-FID/MS 进行在线分析. C2 组分通过 FID 检测器检测, C3 及以上组分由 MS 检测,方法检出限(φ)为 $(0.03 \sim 0.09) \times 10^{-9}$,精密度在 10% 以内. 共测定烷烃 29 种、烯烃 9 种、卤代烃 34 种、芳香烃 17 种、醛类 9 种、酮类 4 种、醚类 3 种、酯类 3 种以及异丙醇、二硫化碳和乙炔. O_3 和 NO_2 光解



图 1 监测点位示意

Fig. 1 Location of the monitoring station

速率常数(J_{O_3} 和 J_{NO_2})由美国 Metcon 公司 UF-CCD 光解光谱仪测得. 仪器每日定时自动校准,每周人工校准一次.

1.3 分析方法

1.3.1 气团老化程度分析

气团老化程度可反映气团中 VOCs 的大气光化学反应状态,与 VOCs 排入大气后的传输时间和传输路径上大气氧化性的强弱有关^[21]. 通常以来源相近但光化学活性相差较大的两种 VOCs 物种浓度比,如乙苯:间/对-二甲苯、甲苯:苯、正丁烷:丙烷和丁烷:丙烷等来衡量气团的老化程度^[22]. 本文选取乙苯:间/对-二甲苯对样品进行分类.

1.3.2 PMF 模型

正定矩阵因子分解法(PMF)是基于最小二乘法的定量受体源解析模型,将实测样品数据分解为源成分谱矩阵、源贡献矩阵和残差矩阵. 基本原理如下^[23]:

$$\mathbf{E}_{ik} = \sum_{j=1}^k \mathbf{A}_{ij} \mathbf{B}_{jk} + \boldsymbol{\varepsilon}_{ik} \quad (i = 1, 2, \dots, n) \quad (1)$$

式中, \mathbf{E}_{ik} 为第 k 次观测的 VOCs 物种 i 的浓度; \mathbf{A}_{ij} 为污染源 j 排放的 VOCs 源成分谱中物种 i 的质量分数; \mathbf{B}_{jk} 为源 j 对受体点处样品 k 中 VOCs 总浓度的贡献值; $\boldsymbol{\varepsilon}_{ik}$ 为模型对样品 k 中物种 i 浓度拟合的残差.

Q 表示模型对样品物种浓度拟合残差与其不确定度比值的平方和. PMF 通过梯度下降法,求得 Q 取最小值时的各类源的贡献浓度和成分谱:

$$Q = \sum_i^m \sum_k^n \left(\frac{\varepsilon_{ik}}{\sigma_{ik}} \right)^2 \quad (2)$$

式中, σ_{ik} 为样品 k 中物种 i 浓度测量的不确定度与模型不确定度的总和^[24].

2 结果与讨论

2.1 VOCs 日均值变化特征

利用 2019 年 9 月至 2020 年 3 月乌海超级站 VOCs 日均体积分数数据及 Ehhalt 等^[25]的研究提出

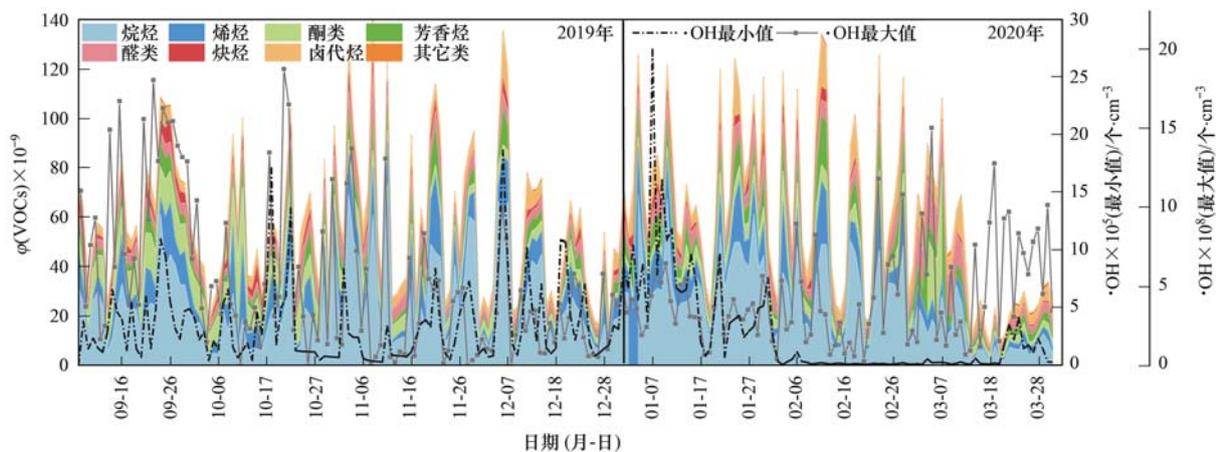


图 2 采样期间各类 VOCs 体积分数和·OH 浓度日变化

Fig. 2 Diurnal variation in VOCs volume fraction and ·OH concentration during sampling

2.2 基于气团老化的环境空气中 VOCs 分类

本研究选择以乙苯:间/对-二甲苯(相关系数为 0.79)为气团老化程度的判断指标,其比值越大,老化程度越高.根据乙苯:间/对-二甲苯将样品分为高、中和低这 3 种老化状态,各类样品数占样品总数的比例及对应性质见表 1.分类后乙苯与间/对-二甲苯相关系数的提升,表明

的公式,计算所得每日·OH 浓度的最大值和最小值,如图 2 所示.烷烃体积分数占比(41%)最大,C7~C12 的长链烷烃可反映重型柴油车排放的影响^[26~28].2020 年 3 月 2~14 日出现一次以丙酮体积分数上升为主的污染过程,丙酮为重要的溶剂^[29],故可能受到工艺过程源影响.VOCs 体积分数与·OH 浓度变化剧烈,大气氧化性差异较大,因此,在解析过程中考虑样品在大气中发生光化学反应后的老化程度十分必要.

同类样品中 VOCs 来源具有较高一致性.为进一步确定气团分类的合理性,基于实测 NO_2 、 J_{NO_2} 和 J_{OH} 数据,利用 Ehhalt 等^[25]提出的·OH 浓度计算公式得出各类样品对应·OH 浓度.高老化样品经历的光化学过程最长,消耗了更多的·OH;低老化样品则消耗最少的·OH,这与比值法的判断一致.

表 1 不同老化程度类别样品性质和·OH 浓度¹⁾

Table 1 Properties and ·OH concentration of samples with different aging degrees

类别	未分类	高老化态	中老化态	低老化态
乙苯:间/对-二甲苯	—	> 0.60	0.20 ~ 0.60	< 0.20
乙苯与间/对-二甲苯的相关系数	0.79 **	0.98 **	0.88 **	0.89 **
各类别样本数占总样本数的比例/%	—	2.30	90.85	6.85
·OH 数浓度/个·cm ⁻³	—	2.71×10^5	5.44×10^5	5.82×10^5

1) ** 表示相关性在 0.01 水平上显著(双尾)

2.3 结果对比验证

考虑其老化特性分类后,利用 PMF 5.0 对 VOCs 小时监测数据进行解析,因子筛选在 3~9 个,模型运行 100 次以上,使解析结果的残差值尽量处于 -3~3,且符合正态分布,模型结果趋于稳定的同时 Q 值接近自由度后,从 4 个方面进行比较:①模拟值与实测值线性回归方程的斜率与截距,斜率越接近 1 越好,截距则越接近于 0 越好.②标准误差是

模拟值与实测值偏差的平方和观测次数比值的平方根,用来衡量观测值与真值之间的偏差.标准误差越小表示拟合程度越好.③模拟值与实测值线性回归的决定系数越接近 1,表示实测值的变动能被模拟值解释的程度越高.④残差表示了实测值与模拟值之间的差.本研究将残差在 -3~3 定为合格,将合格样品数占样品总数的比例称为残差合格率.与未分类结果相比残差合格率增加的物种数占总物种数

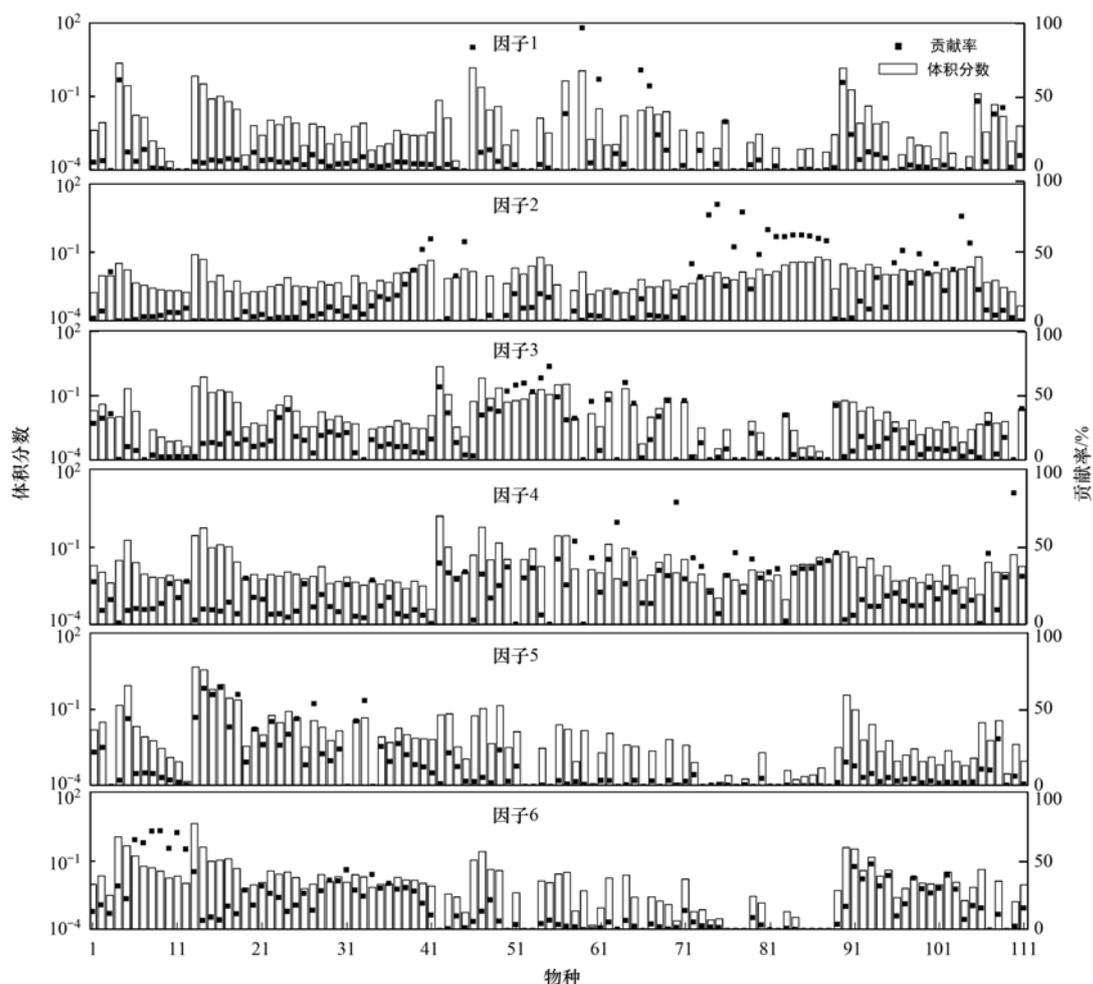
的比例称为残差合格提升率. 残差合格提升率越高, 代表残差合格的物种越多, 模拟结果越好.

结果表明(表 2), 相比于未分类的结果, 分类后解析结果的各项模型参数均有改善.

表 2 模型诊断参数对比

Table 2 Comparison of model diagnostic parameters

分类	截距	斜率	标准误差	决定系数	残差合格提升率/%
未分类	0.24	0.51	0.46	0.59	—
高老化态	0.12	0.71	0.36	0.74	88
中老化态	0.24	0.55	0.45	0.60	59
低老化态	0.21	0.60	0.36	0.68	79



1. 乙酸乙酯, 2. 乙酸乙酯, 3. 甲基丙烯酸酯, 4. 乙烯, 5. 丙烯, 6. 正丁烯, 7. 丁二烯, 8. 反式-2-丁烯, 9. 顺式-2-丁烯, 10. 1-戊烯, 11. 反式-2-戊烯, 12. 顺式-2-戊烯, 13. 乙烷, 14. 丙烷, 15. 异丁烷, 16. 正丁烷, 17. 异戊烷, 18. 正戊烷, 19. 2,2-二甲基丁烷, 20. 环戊烷, 21. 2,3-二甲基丁烷, 22. 2-甲基戊烷, 23. 3-甲基戊烷, 24. 正己烷, 25. 甲基环戊烷, 26. 2,4-二甲基戊烷, 27. 环己烷, 28. 2-甲基己烷, 29. 2,3-二甲基己烷, 30. 3-甲基己烷, 31. 2,2,4-三甲基己烷, 32. 正庚烷, 33. 甲基环己烷, 34. 2,3,4-三甲基己烷, 35. 2-甲基庚烷, 36. 3-甲基庚烷, 37. 正辛烷, 38. 正壬烷, 39. 正癸烷, 40. 十一烷, 41. 十二烷, 42. 丙酮, 43. 2-丁酮, 44. 4-甲基-2-戊酮, 45. 2-己酮, 46. 乙炔, 47. 乙醛, 48. 丙醛, 49. 丙醛, 50. 正丁醛, 51. 丁醛, 52. 戊醛, 53. 己醛, 54. 苯甲醛, 55. 间-甲基苯甲醛, 56. 二氟二氯甲烷, 57. 氯甲烷, 58. 1,1,2,2-四氟-1,2-二氯乙烷, 59. 氯乙烯, 60. 溴甲烷, 61. 对-乙基甲苯, 62. 一氟三氯甲烷, 63. 1,1,二氯乙烷, 64. 二氯甲烷, 65. 1,2,2-三氟-1,1,2-三氯乙烷, 66. 反式 1,2-二氯乙烷, 67. 1,1-二氯乙烷, 68. 三氯甲烷, 69. 1,2-二氯乙烷, 70. 1,1,1-三氯乙烷, 71. 1,2-二氯丙烷, 72. 一溴二氯甲烷, 73. 三氯乙烯, 74. 反式 1,3-二氯-1-丙烯, 75. 顺式 1,3-二氯-1-丙烯, 76. 1,1,2-三氯乙烷, 77. 二溴一氯甲烷, 78. 1,2-二溴乙烷, 79. 四氯乙烯, 80. 氯苯, 81. 三溴甲烷, 82. 1,1,2,2-四氯乙烷, 83. 氯代甲苯, 84. 1,4-二氯苯, 85. 1,3-二氯苯, 86. 1,2-二氯苯, 87. 1,2,4-三氯苯, 88. 1,1,2,3,4,4-六氯-1,3-丁二烯, 89. 四氯化碳, 90. 苯, 91. 甲苯, 92. 乙苯, 93. 间/对-二甲苯, 94. 苯乙烯, 95. 邻二甲苯, 96. 异丙苯, 97. 正丙苯, 98. 对-乙基甲苯, 99. 间-乙基甲苯, 100. 1,3,5-三甲基苯, 101. 邻-乙基甲苯, 102. 1,2,4-三甲基苯, 103. 1,2,3-三甲基苯, 104. 间-二乙基苯, 105. 对-二乙基苯, 106. 萘, 107. 异丙醇, 108. 二硫化碳, 109. 四氢呋喃, 110. 1,4-二氧六环, 111. 甲基叔丁基醚

图 3 中老化 VOCs 样品中各因子的物种贡献体积分数及其对物种的贡献率

Fig. 3 Species volume fraction of each factor and its contribution rate to the species in VOCs samples of a medium-aging state

2.4 解析结果

如图 3 所示,以中老化态为例对本研究提出的基于气团老化分类解析的因子识别进行介绍. 中老化态共解得 6 个因子,因子 1 含有丰富的乙烷、丙烷、苯、甲苯和乙炔,其中乙炔是燃烧源的重要示踪物,故该因子识别为燃烧源^[30]; 因子 2 含有丰富的正辛烷、正癸烷、十一烷、十二烷、丙烯和大量的苯系物,其中长链烷烃由重型柴油车排放,故该因子可能为柴油车源^[31,32]; 因子 3 含有丰富的丙烯、反式-2-丁烯、丙烯醛和丙酮,其中丙烯、丙烯醛和丙酮为餐饮类主要排放^[33,34],故该因子识别为餐饮源; 因子 4 含有丰富的丙烷、正丁烷、异丁烷、乙炔和乙烯,其中 C2~C6 直链烷烃是汽油车尾气排放的标志物,乙烯和乙炔是汽油燃烧的产物,故因子 4 识别为汽油车源^[35,36]; 因子 5 含有丰富的苯、甲苯、异戊烷、1,3,5-三甲基苯、1,2,4-三甲基苯、甲基环戊烷、对乙基甲苯和异丙醇,故该因子判为焦化源^[37,38];

因子 6 中含有丰富的丙酮、2-丁酮、反-2-戊烯、苯、甲苯、乙苯、乙醛、丙醛和间/对-二甲苯,其中较高的丙酮、苯、甲苯和乙醛源于工艺过程^[39~41],故该因子可能为除焦化外其它工业及溶剂的混合源.

不同老化态和总解析结果如表 3 所示. 研究期间对乌海市 VOCs 贡献较大的四大污染源分别为焦化源(27.65%)、燃烧源(21.49%)、除焦化外其它工业及溶剂的混合源(17.85%)和餐饮源(18.26%); 高老化气团中焦化源贡献率最大,达 47.20%,柴油车源贡献率最小,为 1.40%; 低老化态中燃烧源与焦化源贡献率相差不大,分别为 28.67%和 24.39%. 对比不同老化程度气团的解析结果,来源具有一定差距,这反映了不同污染源排放 VOCs 的物种活性和传输时间的差异在大气环境氧化性变化的情况下,使受体点处 VOCs 的贡献比例发生不规则变化,这也说明了解析时考虑 VOCs 在大气环境中反应状态的合理性.

表 3 各类 VOCs 样品的污染源贡献率及加权总贡献率/%

Table 3 Contribution rate of pollution sources in all kinds of VOC samples and weighted total contribution rate/%

源类	高老化态	中老化态	低老化态	各类别加权总结果
焦化	47.20	27.40	24.39	27.65
餐饮	20.80	18.30	16.87	18.26
燃烧源	15.70	21.10	28.67	21.49
柴油车	1.40	2.40	5.48	2.59
汽油车	4.50	12.70	7.58	12.16
除焦化外其它工业和溶剂混合源	10.40	18.10	17.01	17.85

3 结论

(1) 考虑 VOCs 传输过程中的大气光化学反应状态,按气团老化程度分类后进行源解析,其结果相较于未分类的模拟值与实测值的标准误差和决定系数等模型诊断参数改善明显.

(2) 基于不同气团老化特性的分类方法,将反应状态相近的 VOCs 样品进行归类,极大地减少了样品间因 VOCs 物种反应变化引起的差异,使输入数据更符合受体模型物种相对稳定的假设.

(3) 乌海市 VOCs 主要来源于焦化源和燃烧源,占比分别为 27.65%和 21.49%; 在老化 VOCs 样品中焦化源贡献率最高,达 47.20%; 在未反应的 VOCs 样品中燃烧源和焦化源占比均较高,分别为 28.67%和 24.39%,分类后的解析结果更符合源排放的实际情况.

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