VOCs源解析模型方法

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报告大纲

- 介绍
- VOCs采样与分析
- 源解析
  - 已知源谱
  - 未知源谱
- 总结

介绍

- 为什么VOCs很重要?
    - 直接健康影响
        - 急性 –羰基化合物与刺激反应
        - 慢性 – 多环芳烃（PAHs）和致癌
    - 参与大气化学过程
        - 生成臭氧
        - 生产二次有机气溶胶（SOA）

介绍

- 为什么VOCs很重要?
  - 多环芳烃（PAHs）
  - 活性烃类化合物
    - 烯烃包括萜烯和异戊二烯
    - 芳烃如苯、甲苯、二甲苯
    - 脂肪烃（在较小程度上）

采样与分析

- 在美国，设置了站点来监测与臭氧生成最为相关的碳氢化合物—**光化学评估监测站 (Photochemical Assessment Monitoring Stations， PAMS)**
- 介绍可见：http://www3.epa.gov/ttnamti1/pamsmain.html

采样与分析

- 采样
  - 半连续在线采样与分析
    - 自动气相色谱仪(Auto-GC)
  - 全组分样品 (离线采样)
    - 采样罐采样后带回实验室进行分析

采样与分析

VOC 测量技术

采样罐

VS

自动气相色谱仪

- 数据为采样时段平均值
- 低建设成本
- 会不断产生实验室/样品运送成本
- 耗费人力
- 采样罐带来的误差

- 小时数据
- 更高的建设成本
- 分析数据需要更高的技术水平
- 一些物种难以识别

采样与分析

碳氢化合物
乙烯
乙炔
乙烷
丙烯
丙烷
异丁烷
1-丁烯
正丁烷
t-2-丁烯
c-2丁烯
异戊烷
1-戊烯类
正戊烷
异戊二烯
T-2-戊烯
c-2-戊烯
2,2-二甲基丁烷
环戊
2,3-二甲基
2-甲基戊烷

3-甲基戊烷
2-甲基-1-戊烯
正己烷
甲基
2,4-二甲基
苯
环己烷
2-甲基己烷
2,3-二甲基戊烷
三甲基己烷
2,2,4-三甲基戊烷
正庚烷
甲基环己烷
2,3,4-三甲基
甲苯
2-甲基庚烷
3-甲基庚烷
正辛烷
乙苯
m&p二甲苯

苯乙烯
邻二甲苯
正壬烷
异丙基苯
正丙
m-乙基甲苯
对乙基甲苯
1,3,5-三甲苯
邻乙基甲苯
1,2,4-三甲基苯
正癸烷
连三甲苯
m-二乙基苯
对二乙苯
正十一烷

羰基
甲醛
丙酮
乙醛

源解析用什么采样方法最合适?

- 自动气相色谱仪(auto-GC)：可每小时采样一次，一天24个样本。
- 采样罐：每3小时采样1次，每天8次。但是大多数采样罐站点每天就采样2次（上午、下午各1次），没有晚上的样本。

因此，源解析研究多选用
自动气相色谱仪。

源解析方法

- 所有的源解析方法都是基于质量平衡法
- 我们观测到的浓度是一系列相对独立的源类别贡献的加和。
- 因此，我们要做质量平衡分析。

质量平衡

质量平衡方程可以写作计算n个样品中的全部m种化学物种来自p个独立排放源的方程。

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj}$$

其中 $i = 1,\ldots, n$ 样品, $j = 1,\ldots, m$ 物种， $k = 1,\ldots, p$ 排放源

受体模型

- 要解开这一方程式，接下来的问题是要了解什么是*已知的*?

- 将问题分为两类：
  - 已知源谱
  - 未知源谱

受体模型

- **已知源谱**
- **化学质量平衡模型**
  - **多元校正方法**
  - 偏最小二乘法
    - 人工神经网络
    - 模拟退火算法
    - 遗传算法

化学质量平衡模型

质量平衡方程可以改写为一个回归问题，其中源谱和环境浓度是已知的。

$$x_j = \sum_{k=1}^{p} g_k f_{kj} + e_j$$

现在，这一方程每次只能代表一个样本。

化学质量平衡模型 (CMB)

- 那么CMB模型主要是回归问题。 然而，由于模型的因变量与自变量均存在误差，因此需要在计算中加入适合的误差模型，以确保计算结果的精确。
- 美国环保局(EPA) 通过将方差最小二乘法与新一代CMB模型(CMB 8)结合，有效地解决了误差问题（详情请见 www.epa.gov/ttn/SCRAM）。

化学质量平衡模型

- 化学质量平衡（CMB）模型已被广泛用于美国西部的PM10来源解析和细颗粒有机碳解析中，这一模型是基于对已故格伦·卡斯（Glen Cass）和他的学生通过一些源排放试验而确定的特定有机物（“分子标记”）的解析而建立的。

化学质量平衡模型 (CMB)

- 应用CMB模型的前提条件是要有源谱。
- 但是排放源采样很难并且成本较高，只有很少的源进行了采样。因此，所需的一些特定污染源类别，可能并没有源谱数据。
- 而对特定排放源类别的源谱组分差异也知之甚少。

受体模型

- 源谱未知
  - 因子分析
    - 主成分分析
    - 绝对主成分分析
    - SAFER/UNMIX
    - 正定矩阵因子分解

因子分析

- 如今我们可能没有时间详究不同方法的细节。
- 有一种最为广泛使用的方法就是正定矩阵因子分解(PMF)模型，因为该方法可以更多地利用数据，并已知相关的测量不确定性。

正定矩阵因子分解模型

- 用最小二乘法来解决因子分析问题
- 各个数据点的权重
- 考虑自然与其他方面的约束
- 建立更为复杂的模型的灵活性

正定矩阵因子分解模型

- 目标函数Q，被定义为

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{\sigma_{ij}} \right]^2$$

其中 σ_{ij} 是x_{ij} 中不确定性的估值

正定矩阵因子分解模型

- 美国环保局PMF (第 5版) 模型可在此链接下载：https://www.epa.gov/air-research/positive-matrix-factorization-model-environmental-data-analyses

正定矩阵因子分解模型

- 有一些将PMF应用于VOC解析的研究， 一个好的例子:
- 中国上海VOCs特征分析与源解析，发表于《大气环境》。（Characteristics and source apportionment of VOCs measured in Shanghai, China, Changjie Cai, Fuhai Geng, Xuexi Tie, Qiong Yu, Junlin An, Atmospheric Environment 44 (2010) 5005 – 5014. ）

采样与分析

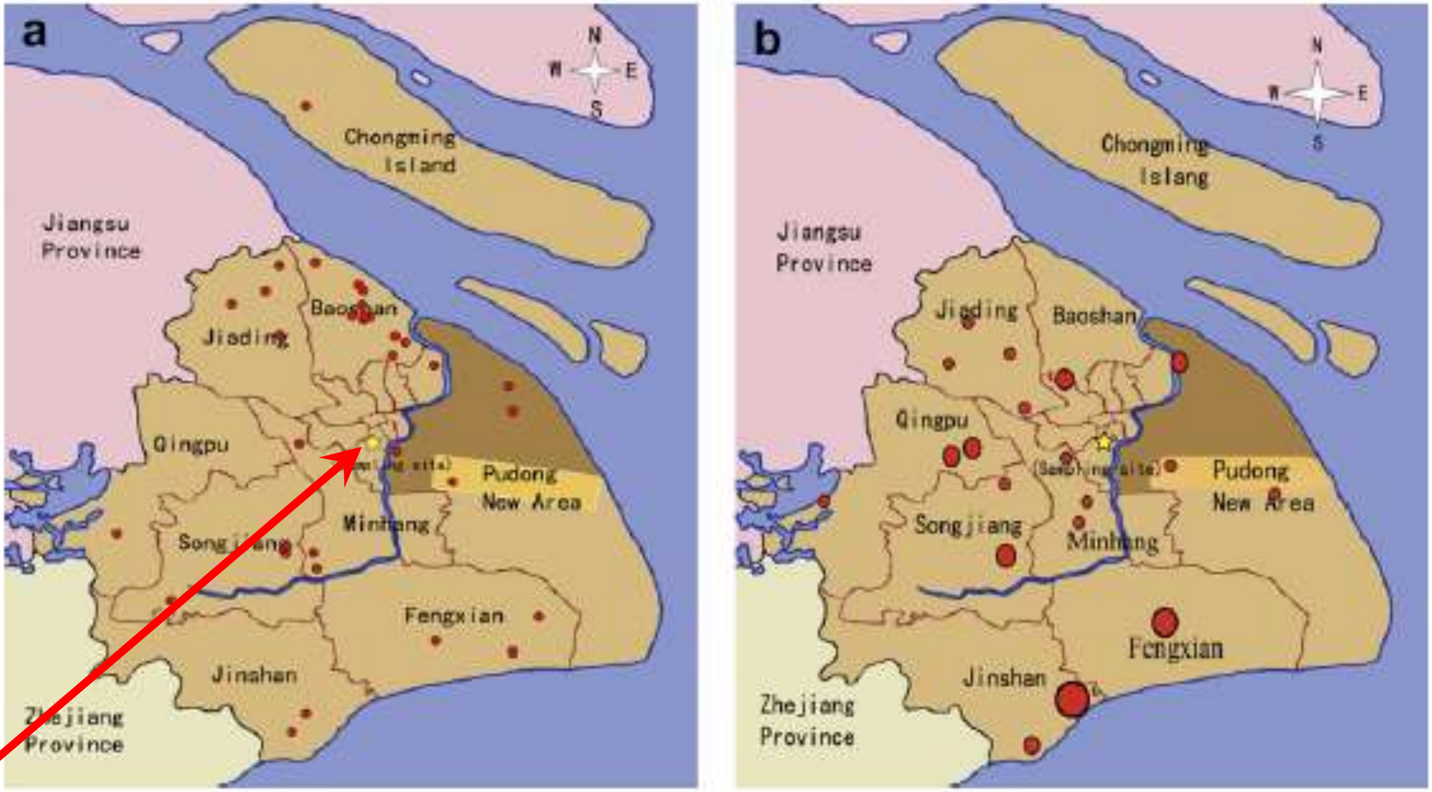


图1. 采样站点位置为地图中黄色五角星，图中还标出了上海大型冶炼厂与钢厂（a）与大型化工园区(b)的位置。

采样与分析

- 采样站点设在上海的中心商业区徐家汇。
- VOCs采样时间为2007年1月至2010年3月，上午6:00-9:00 采样3小时，使用6升容量的带有硅烷化采样阀的采样罐（美国Entech公司生产29-10622 型）。
- 为了研究昼夜变化，2009年8月25到9月20日期间进行VOC密集采样，(一天8个样品，3小时1个)。

采样与分析

- 使用Entech Model 7100 对VOC气体样品进行浓缩预处理。
- 用气相色谱/质量选择检测器分析。
- 对32种物质进行源解析研究。

风数据

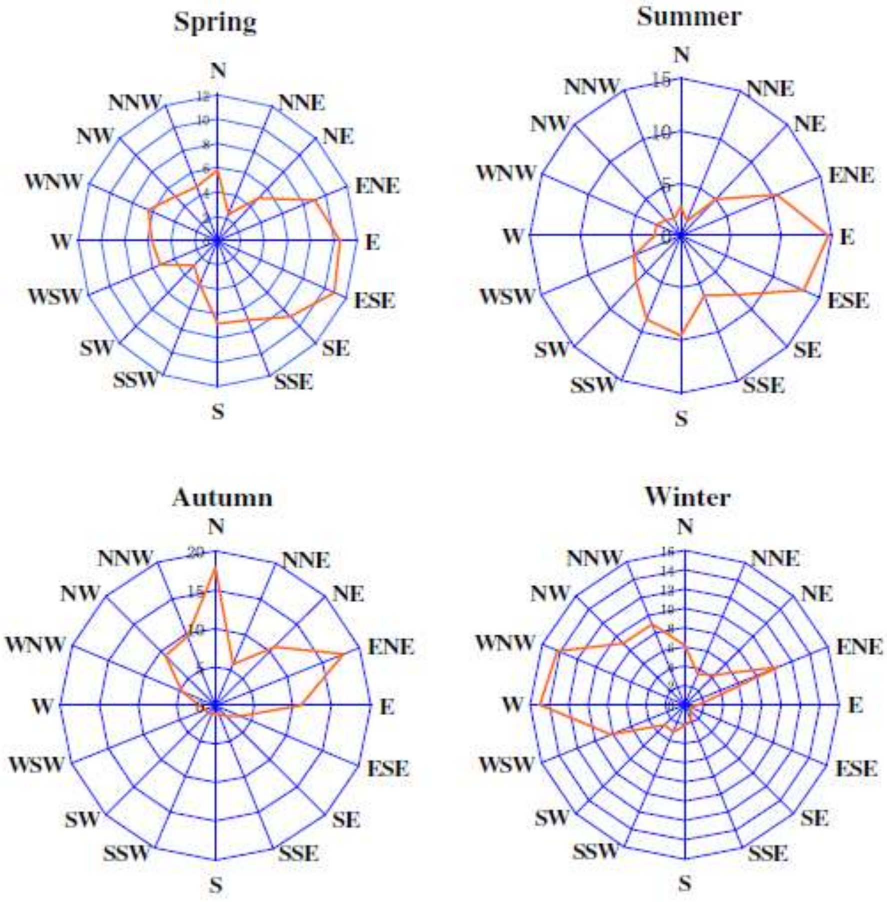


图2： 上海不同季节的盛行风向

风数据

- 全年的不同风向数据很有用，因为在不同时间不同的源处于上风向或下风向。

- 这种变化为PMF分析提供了更强的数据基础。

正定矩阵因子分解(PMF)模型细节

- 在正定矩阵因子分解模型之前,研究人员应首先设定一些因子 (p)。
- p值的选择基于以下几项参数：
  - VOC浓度的误差平方和 (Q值)；
  - VOC物质的归一化残差分布数据；
  - 得到源谱的物理解释性。

PMF细节

- 数值的不确定性分配：
  - $$U = \sqrt{(EF\ \ x\ conc)^2 + (MDL)^2}$$ Conc > MDL
  - 其中，EF代表错误率（EF=不确定性比例/100）；MDL代表方法的检出限。
- 如果污染物浓度小于或等于 MDL，则偏差计算公式为：
  - U = (5/6)*MDL Conc < MDL

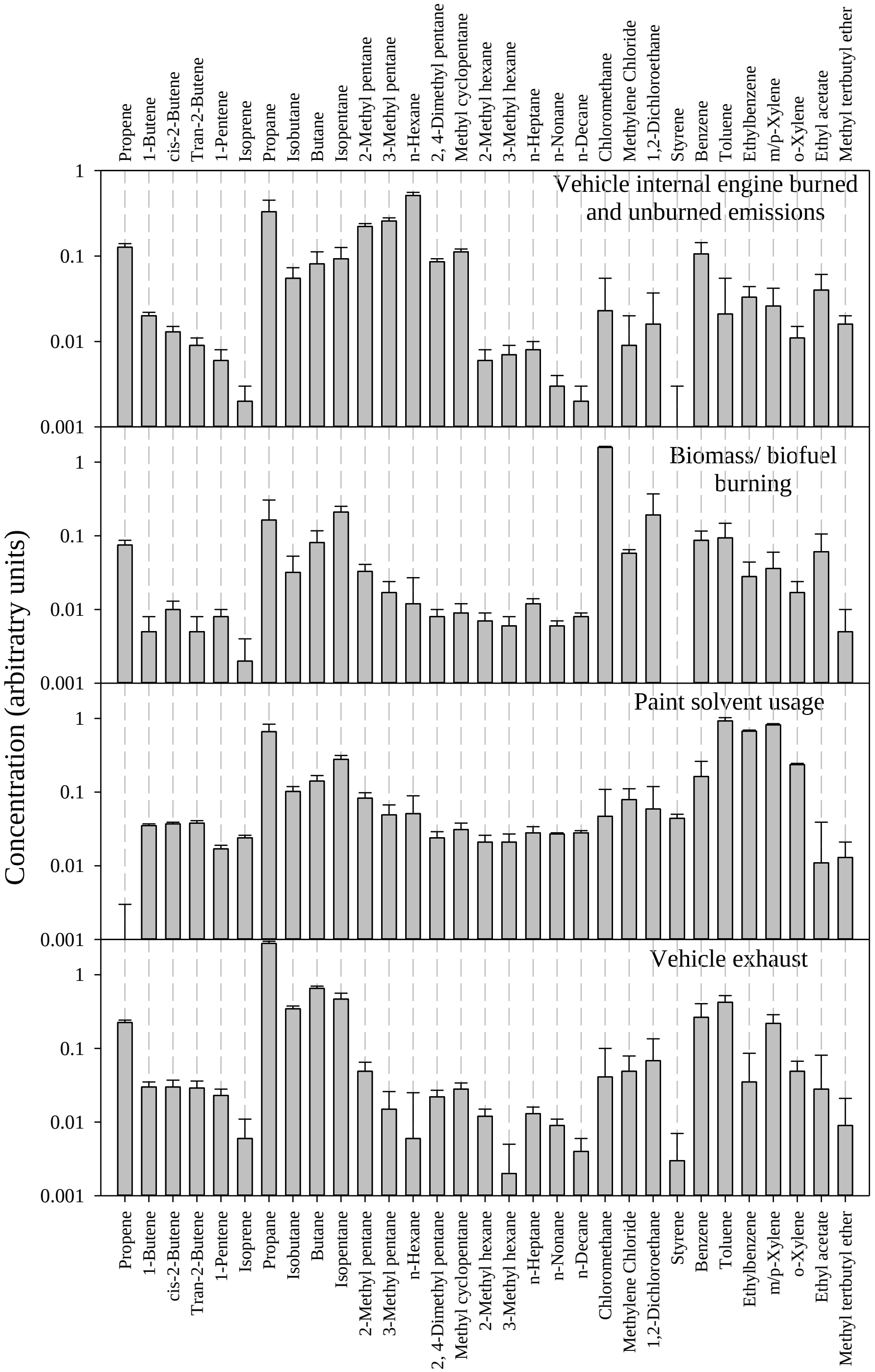
PMF 结果

- 基于得出的Q值与源谱的解释性，选取了8项因子 。
- 为了更好的将移动源归类，该项研究识别了与机动车排放VOCs相关的3种重要来源，包括机动车尾气（约40%），油储罐挥发（约40%），以及发动机内部燃烧排放与曲轴箱通风的非燃烧排放（约20%）。

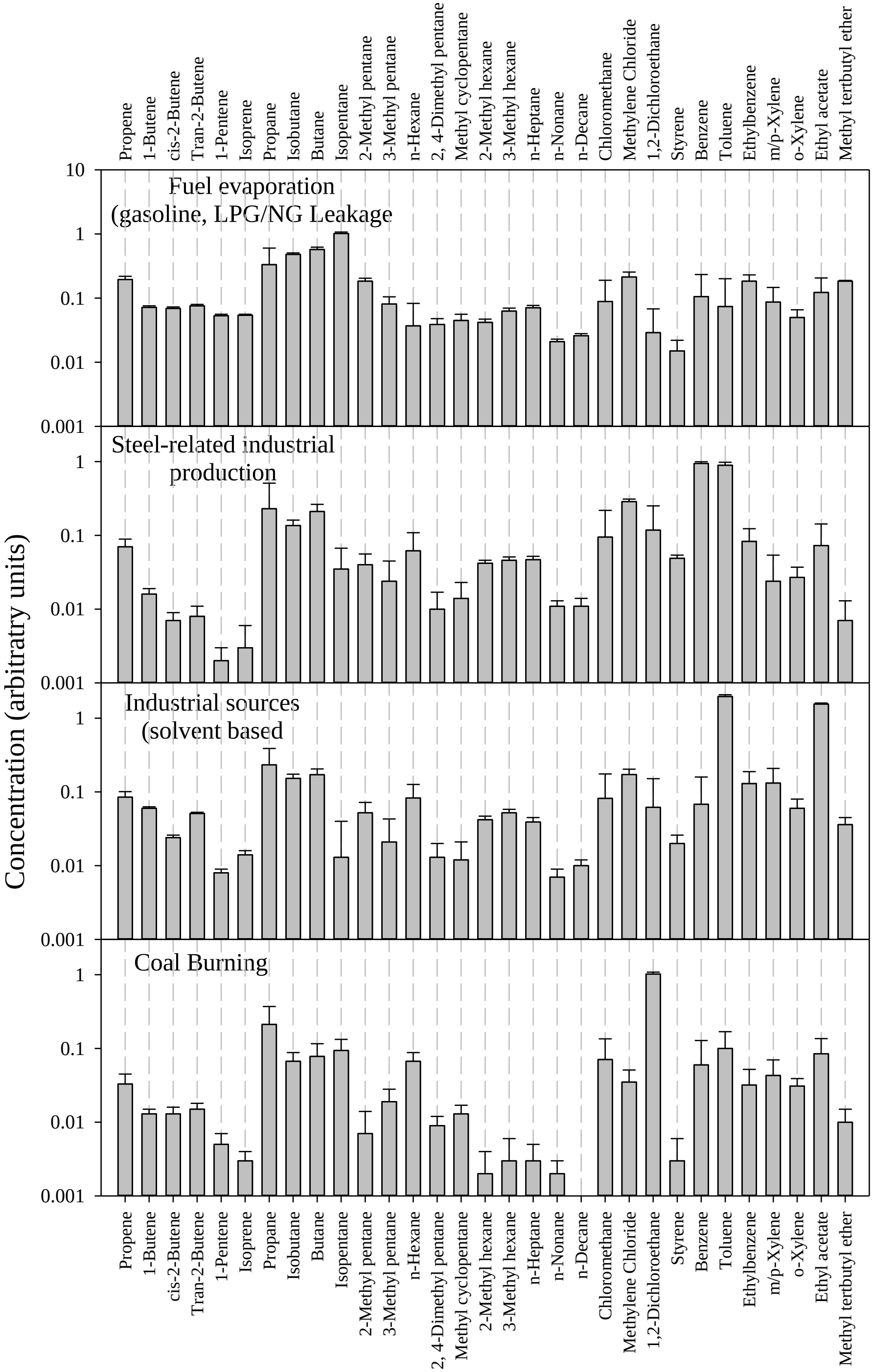
PMF 结果

- Cai et al. 发表的文章中具体说明了如何将源谱对应到不同的排放源。
- 今天没有时间具体探讨各个源谱是如何归因到相应的排放源类别的。

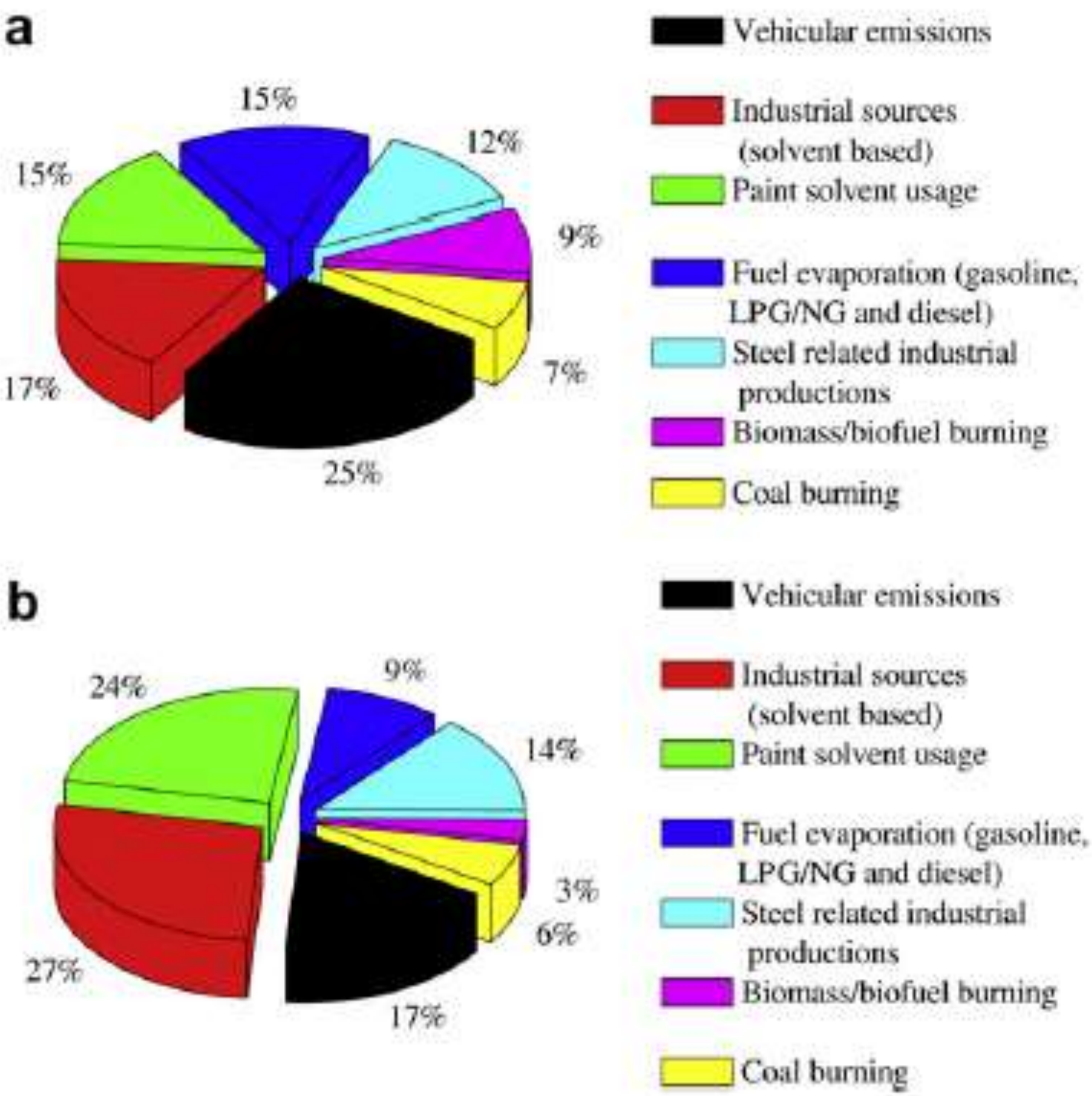
PMF 结果


Vehicle internal engine burned and unburned emissions
Biomass/ biofuel burning
Paint solvent usage
Vehicle exhaust
Concentration (arbitratry units)
1
0.1
0.01
0.001
Propene
1-Butene
cis-2-Butene
Tran-2-Butene
1-Pentene
Isoprene
Propane
Isobutane
Butane
Isopentane
2-Methyl pentane
3-Methyl pentane
n-Hexane
2, 4-Dimethyl pentane
Methyl cyclopentane
2-Methyl hexane
3-Methyl hexane
n-Heptane
n-Nonane
n-Decane
Chloromethane
Methylene Chloride
1,2-Dichloroethane
Styrene
Benzene
Toluene
Ethylbenzene
m/p-Xylene
o-Xylene
Ethyl acetate
Methyl tertbutyl ether

PMF 结果


Fuel evaporation (gasoline, LPG/NG Leakage
Steel-related industrial production
Industrial sources (solvent based
Coal Burning
Concentration (arbitratry units)
10
1
0.1
0.01
0.001
Propene
1-Butene
cis-2-Butene
Tran-2-Butene
1-Pentene
Isoprene
Propane
Isobutane
Butane
Isopentane
2-Methyl pentane
3-Methyl pentane
n-Hexane
2, 4-Dimethyl pentane
Methyl cyclopentane
2-Methyl hexane
3-Methyl hexane
n-Heptane
n-Nonane
n-Decane
Chloromethane
Methylene Chloride
1,2-Dichloroethane
Styrene
Benzene
Toluene
Ethylbenzene
m/p-Xylene
o-Xylene
Ethyl acetate
Methyl tertbutyl ether

PMF 结果



a.VOC测量浓度的不同来源贡献
b.与a相同，除了臭氧生成潜力

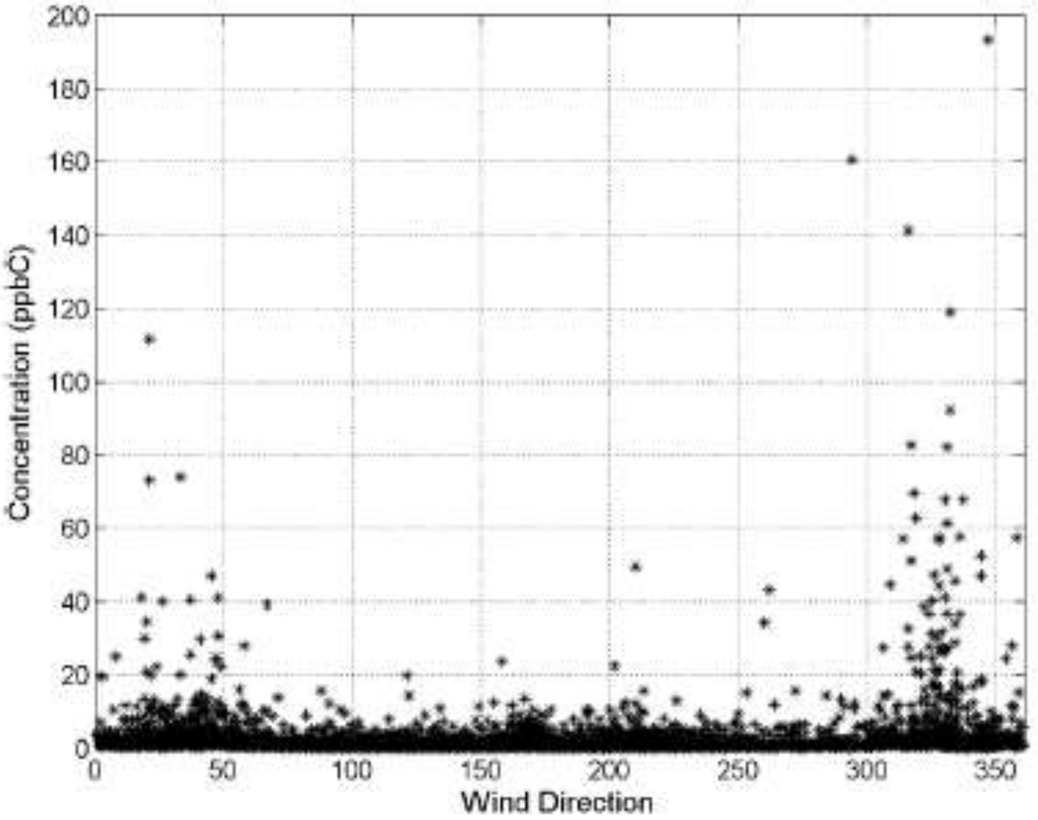
其他方法

- 如果有自动气相色谱系统得出的更高时间分辨率的数据，就可以使用其它方法来进行VOCs溯源。
- 一个例子是发表在《大气环境》的Henry 等人的一项研究成果，使用非参数回归模型识别美国德克萨斯州休斯顿市的VOCs主要排放源(Atmospheric Environment 36 (2002) 2237–2244) 。

非参数回归模型

- 该方法结合了给定物质在观测时间内的浓度数据和风向数据。

非参数回归模型



鹿园测量的环己烷小时浓度与风向方位

非参数回归模型 (NPR)

- 分析污染源贡献 vs 风向。
- 如果风向已知，则应用无参数的回归模型计算污染物浓度。
- *设滑动窗的宽幅为$\Delta\theta$*，中心点位置为 θ，则平均污染物浓度为：

$$\bar{C}(\theta,\Delta\theta)=\frac{\sum_{i=1}^{n}K((\theta-W_i)/\Delta\theta)C_i}{\sum_{i=1}^{n}K((\theta-W_i)/\Delta\theta)}$$

W_i：测定风向

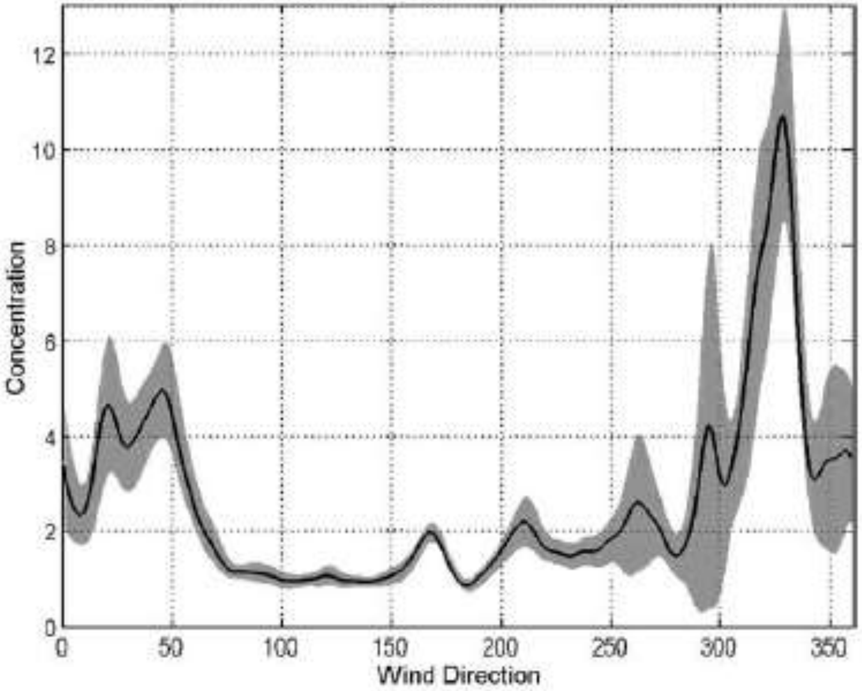
C_i：测定样本 i 浓度

n：样本量

- 应用高斯核函数［$K(x)$］为观测结果分配不同的权重。$K(x)$的计算公式为：

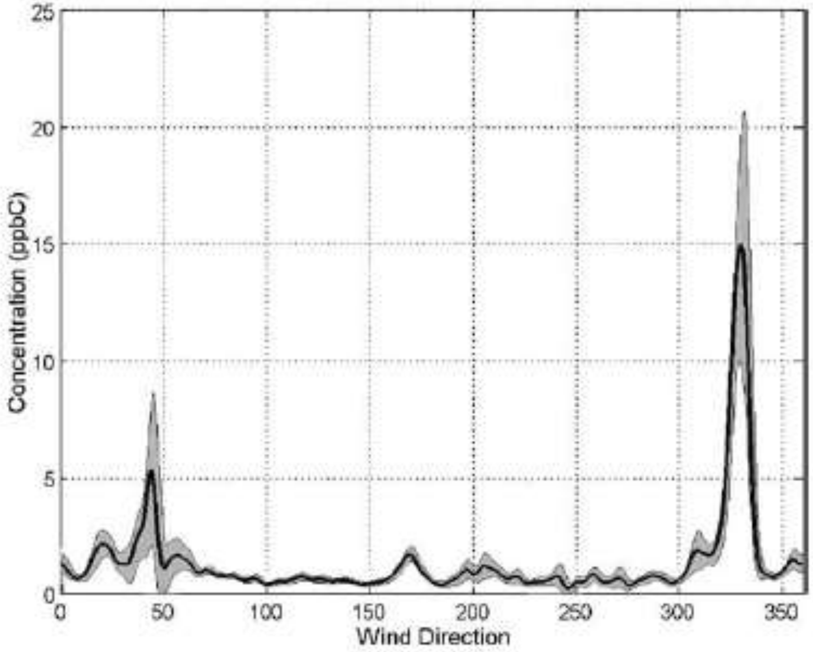
$$K(x)=\frac{1}{\sqrt{2\pi}}\exp(-0.5x^2)$$

非参数回归模型 (NPR)

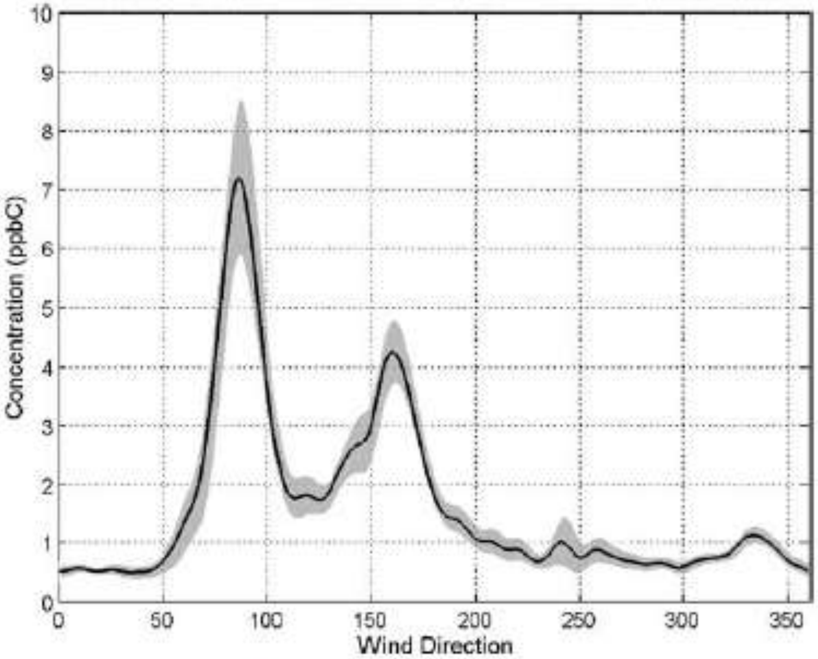


使用高斯核函数对环己烷与风向进行非参数回归（10°半峰全宽），剔除了风速小于每小时1英里的数据。
灰色区域是95%置信区间。

非参数回归模型 (NPR)



使用高斯核函数对环己烷与风向进行非参数回归（5°半峰全宽），采用风速大于每小时6英里（从最大的排放源到监测站点需要1小时输送时间）的数据。灰色区域是95%置信区间。



使用高斯核函数对环己烷与风向进行非参数回归（10°半峰全宽），采用风速大于每小时5英里（从最大的排放源到监测站点需要1小时输送时间）的数据。灰色区域是95%置信区间。

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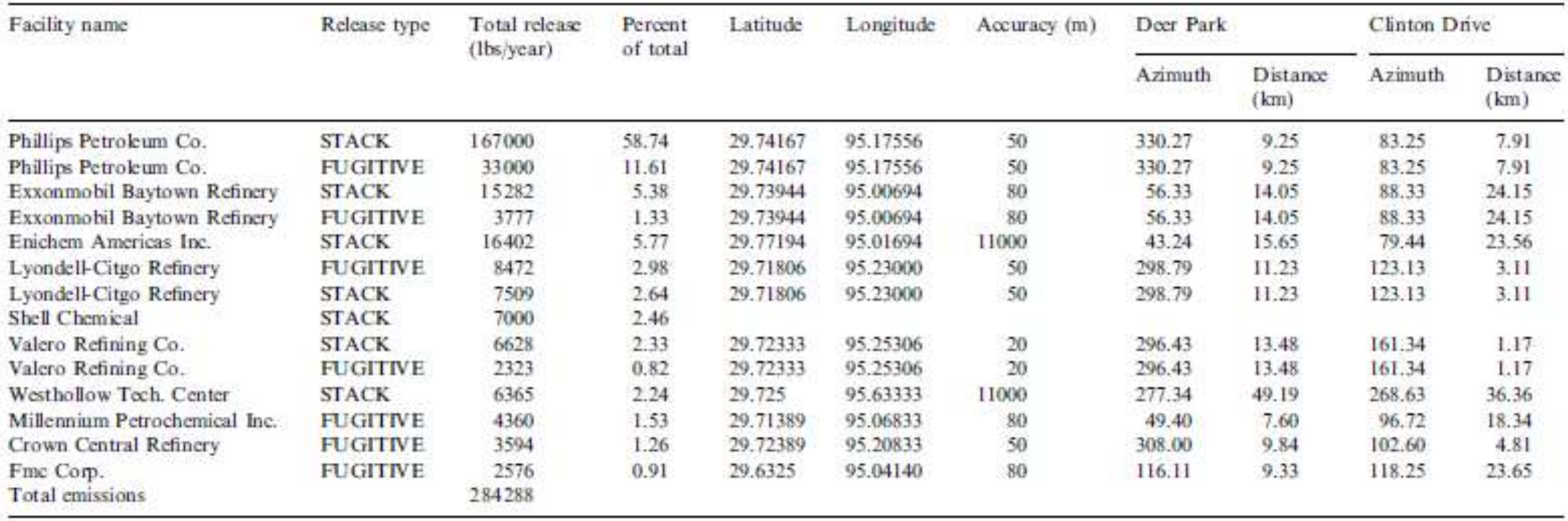
非参数回归模型 (NPR)

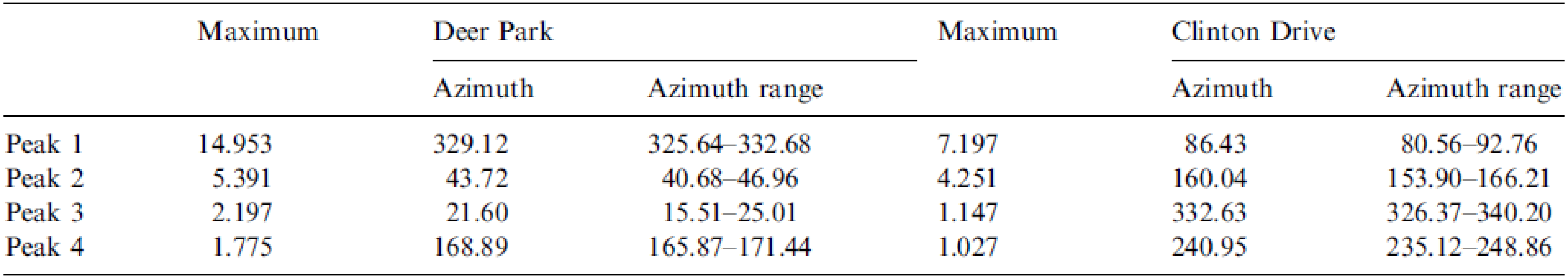
表格1
德克萨斯州Harris公司1997年环己烷排放量



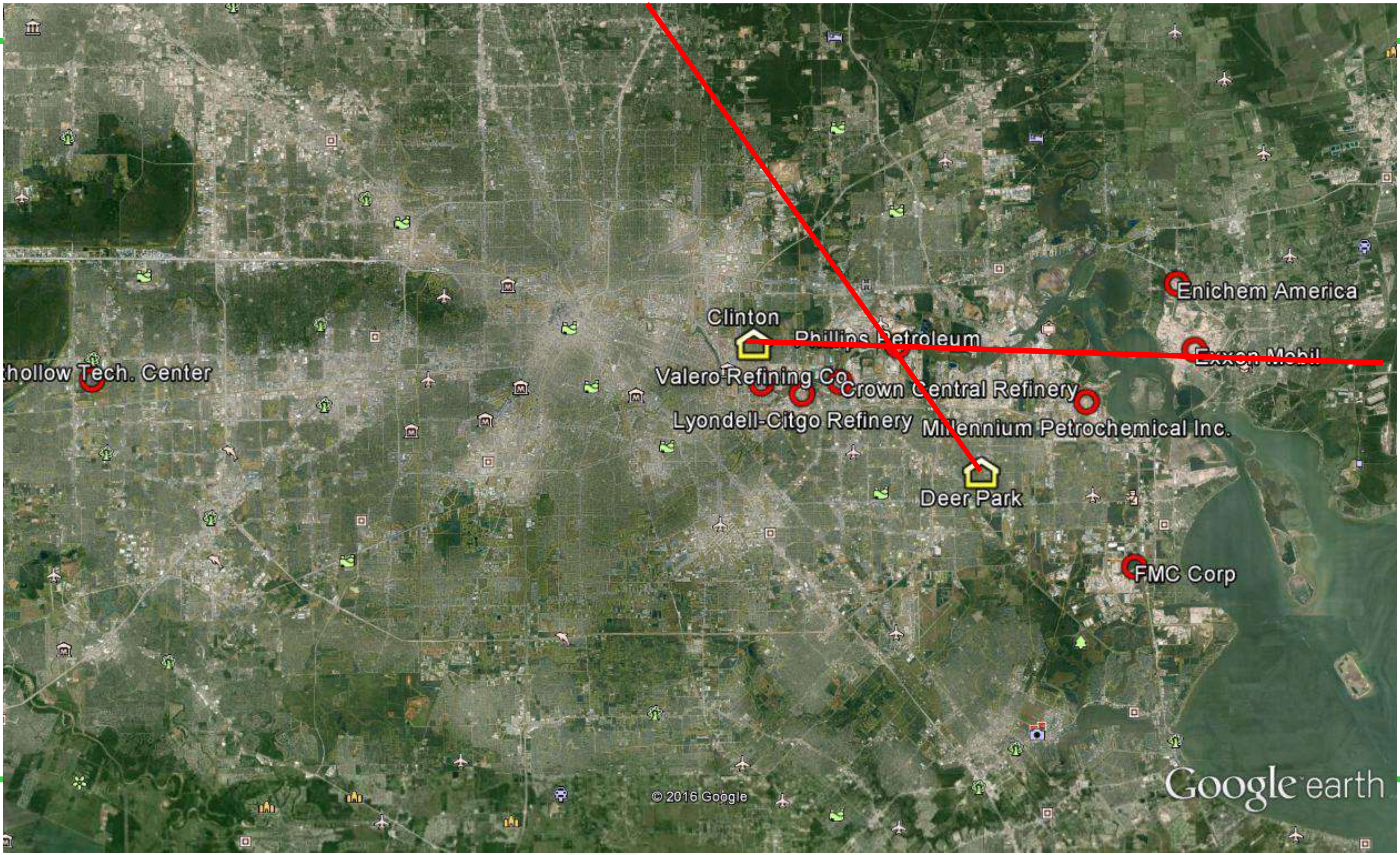
Facility name	Release type	Total release (lbs/year)	Percent of total	Latitude	Longitude	Accuracy (m)	Deer Park		Clinton Drive	
							Azimuth	Distance (km)	Azimuth	Distance (km)
Phillips Petroleum Co.	STACK	167000	58.74	29.74167	95.17556	50	330.27	9.25	83.25	7.91
Phillips Petroleum Co.	FUGITIVE	33000	11.61	29.74167	95.17556	50	330.27	9.25	83.25	7.91
Exxonmobil Baytown Refinery	STACK	15282	5.38	29.73944	95.00694	80	56.33	14.05	88.33	24.15
Exxonmobil Baytown Refinery	FUGITIVE	3777	1.33	29.73944	95.00694	80	56.33	14.05	88.33	24.15
Enichem Americas Inc.	STACK	16402	5.77	29.77194	95.01694	11000	43.24	15.65	79.44	23.56
Lyondell-Citgo Refinery	FUGITIVE	8472	2.98	29.71806	95.23000	50	298.79	11.23	123.13	3.11
Lyondell-Citgo Refinery	STACK	7509	2.64	29.71806	95.23000	50	298.79	11.23	123.13	3.11
Shell Chemical	STACK	7000	2.46							
Valero Refining Co.	STACK	6628	2.33	29.72333	95.25306	20	296.43	13.48	161.34	1.17
Valero Refining Co.	FUGITIVE	2323	0.82	29.72333	95.25306	20	296.43	13.48	161.34	1.17
Westhollow Tech. Center	STACK	6365	2.24	29.725	95.63333	11000	277.34	49.19	268.63	36.36
Millennium Petrochemical Inc.	FUGITIVE	4360	1.53	29.71389	95.06833	80	49.40	7.60	96.72	18.34
Crown Central Refinery	FUGITIVE	3594	1.26	29.72389	95.20833	50	308.00	9.84	102.60	4.81
Fmc Corp.	FUGITIVE	2576	0.91	29.6325	95.04140	80	116.11	9.33	118.25	23.65
Total emissions		284288								

Nonparametric regression (NPR)

表格2
对环己烷与风向进行非参数回归的最大峰值



	Maximum	Deer Park		Maximum	Clinton Drive	
		Azimuth	Azimuth range		Azimuth	Azimuth range
Peak 1	14.953	329.12	325.64–332.68	7.197	86.43	80.56–92.76
Peak 2	5.391	43.72	40.68–46.96	4.251	160.04	153.90–166.21
Peak 3	2.197	21.60	15.51–25.01	1.147	332.63	326.37–340.20
Peak 4	1.775	168.89	165.87–171.44	1.027	240.95	235.12–248.86


Enichem America
Clinton
Phillips Petroleum
Exxon Mobil
hollow Tech. Center
Valero Refining Co
Crown Central Refinery
Lyondell-Citgo Refinery
Millennium Petrochemical Inc.
Deer Park
FMC Corp
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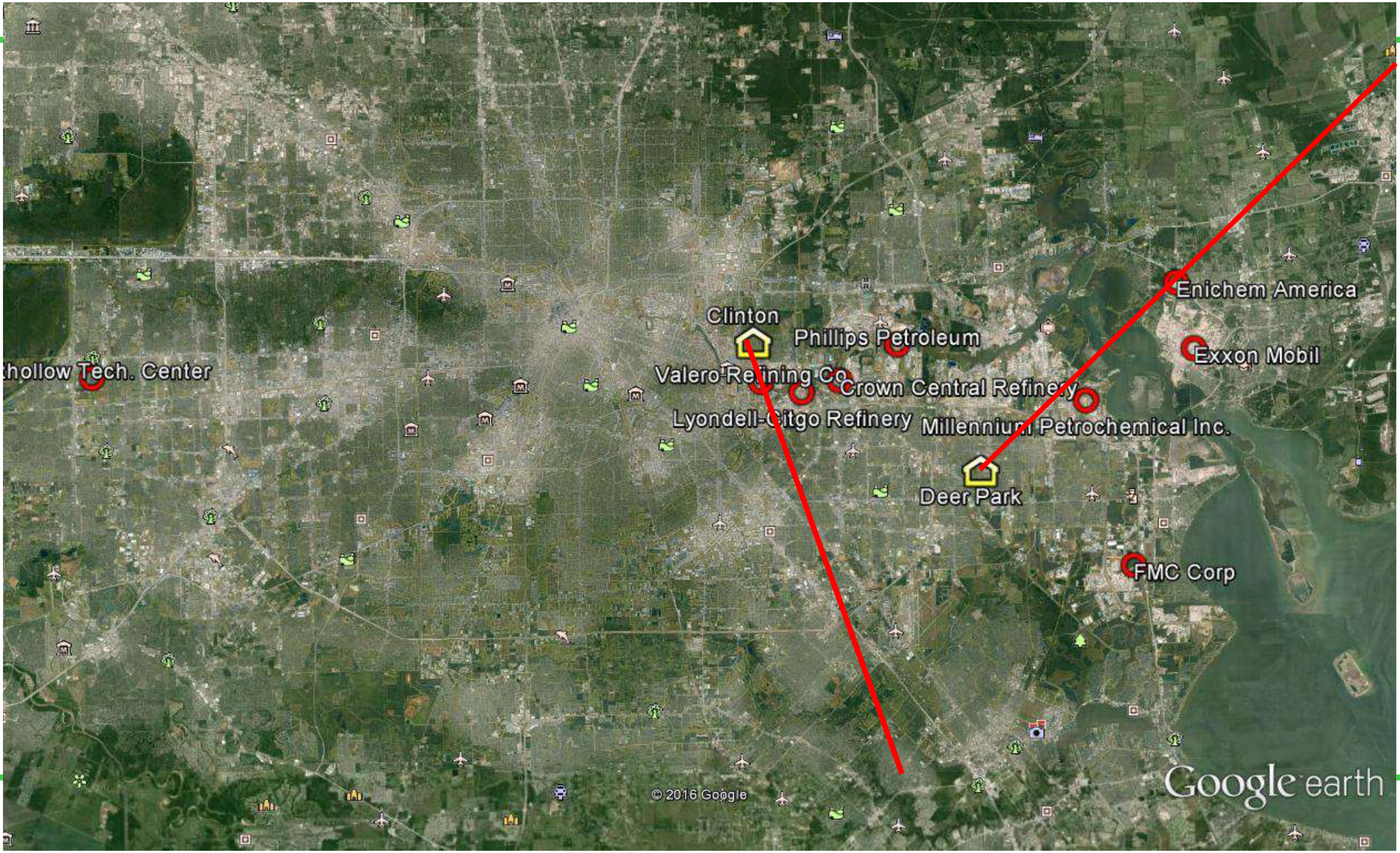
Clinton
Phillips Petroleum
Valero Refining Co
Crown Central Refinery
Lyondell-Citgo Refinery
Millennium Petrochemical Inc.
Enichem America
Exxon Mobil
Deer Park
FMC Corp
hollow Tech. Center
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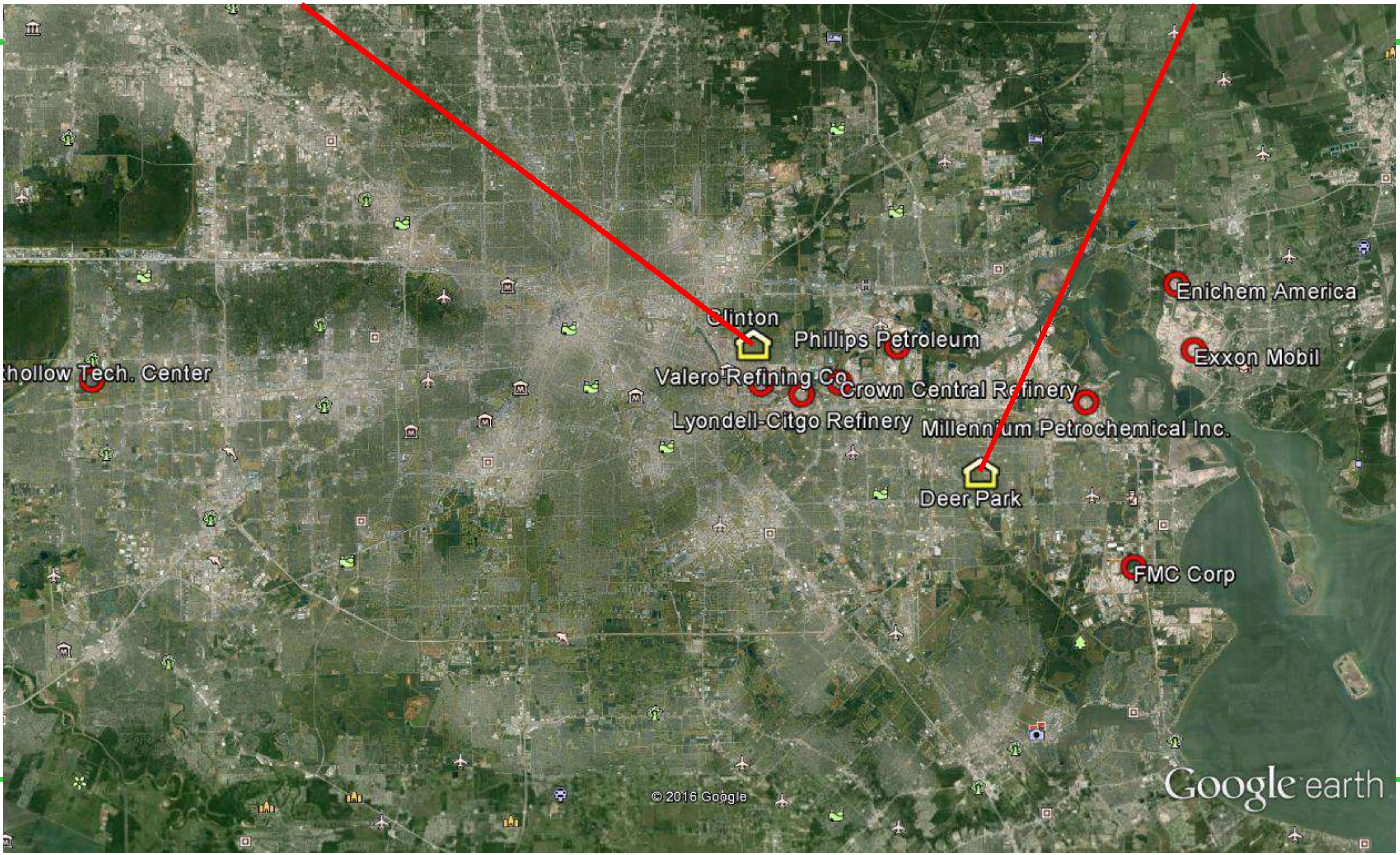
Clinton
Phillips Petroleum
Valero Refining Co
Crown Central Refinery
Lyondell-Citgo Refinery
Millennium Petrochemical Inc.
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Exxon Mobil
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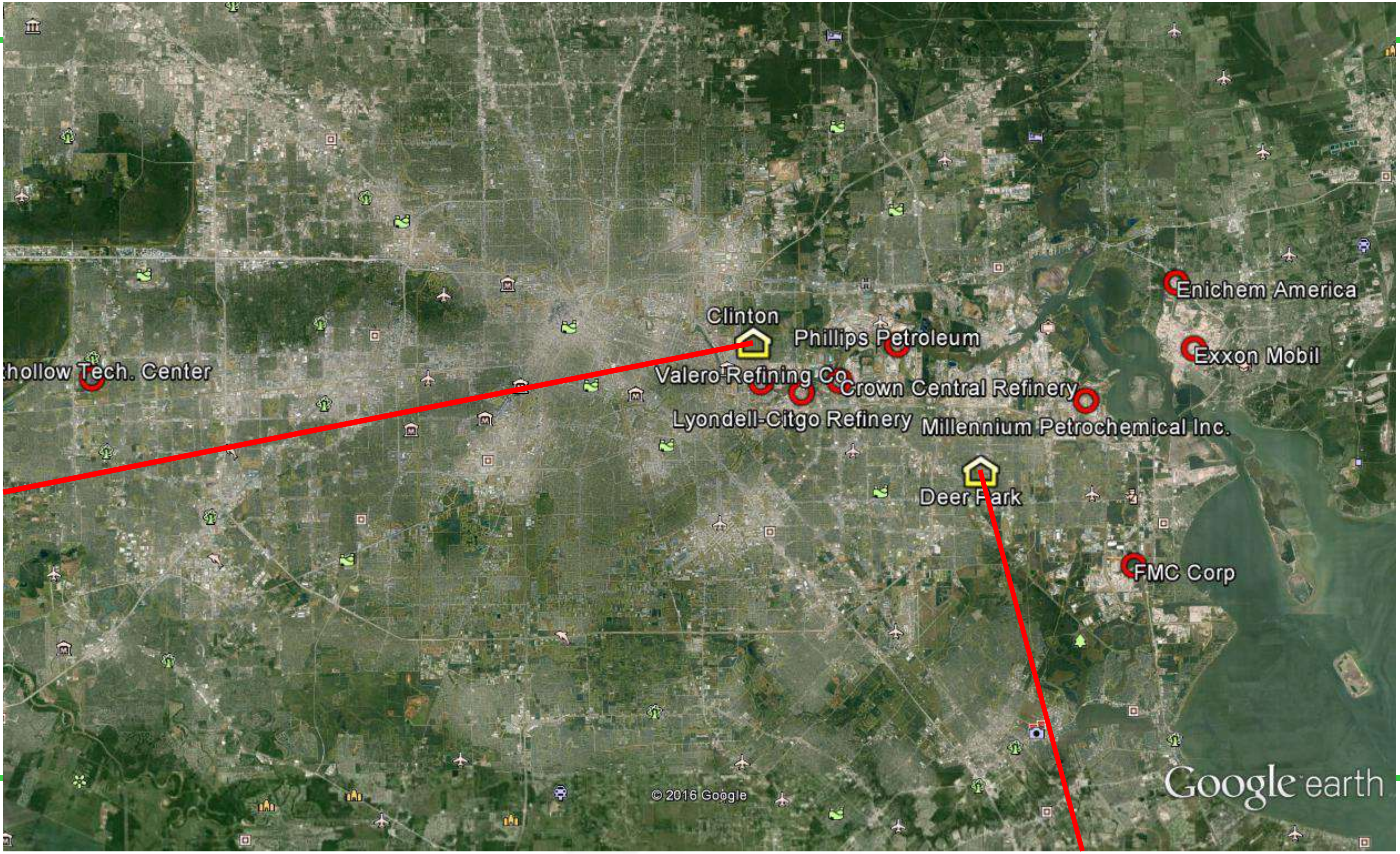
Clinton
Phillips Petroleum
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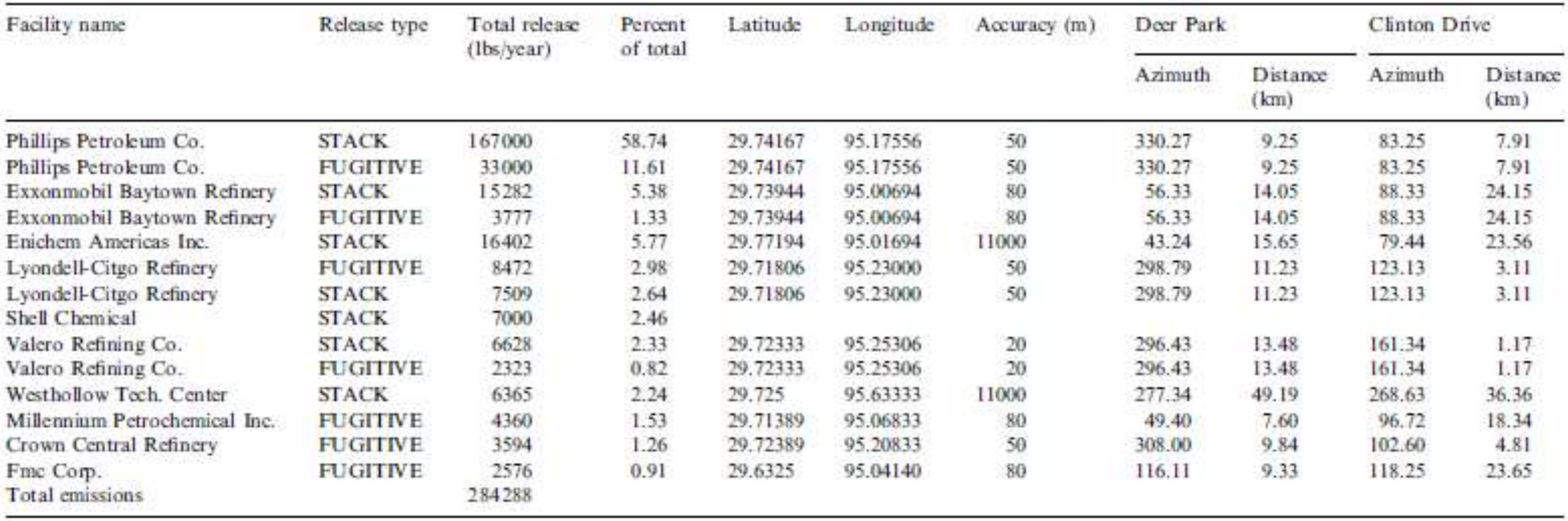
Nonparametric regression (NPR)

表格1
德克萨斯州Harris公司1997年环己烷排放量



Facility name	Release type	Total release (lbs/year)	Percent of total	Latitude	Longitude	Accuracy (m)	Deer Park		Clinton Drive	
							Azimuth	Distance (km)	Azimuth	Distance (km)
Phillips Petroleum Co.	STACK	167000	58.74	29.74167	95.17556	50	330.27	9.25	83.25	7.91
Phillips Petroleum Co.	FUGITIVE	33000	11.61	29.74167	95.17556	50	330.27	9.25	83.25	7.91
Exxonmobil Baytown Refinery	STACK	15282	5.38	29.73944	95.00694	80	56.33	14.05	88.33	24.15
Exxonmobil Baytown Refinery	FUGITIVE	3777	1.33	29.73944	95.00694	80	56.33	14.05	88.33	24.15
Enichem Americas Inc.	STACK	16402	5.77	29.77194	95.01694	11000	43.24	15.65	79.44	23.56
Lyondell-Citgo Refinery	FUGITIVE	8472	2.98	29.71806	95.23000	50	298.79	11.23	123.13	3.11
Lyondell-Citgo Refinery	STACK	7509	2.64	29.71806	95.23000	50	298.79	11.23	123.13	3.11
Shell Chemical	STACK	7000	2.46							
Valero Refining Co.	STACK	6628	2.33	29.72333	95.25306	20	296.43	13.48	161.34	1.17
Valero Refining Co.	FUGITIVE	2323	0.82	29.72333	95.25306	20	296.43	13.48	161.34	1.17
Westhollow Tech. Center	STACK	6365	2.24	29.725	95.63333	11000	277.34	49.19	268.63	36.36
Millennium Petrochemical Inc.	FUGITIVE	4360	1.53	29.71389	95.06833	80	49.40	7.60	96.72	18.34
Crown Central Refinery	FUGITIVE	3594	1.26	29.72389	95.20833	50	308.00	9.84	102.60	4.81
Fmc Corp.	FUGITIVE	2576	0.91	29.6325	95.04140	80	116.11	9.33	118.25	23.65
Total emissions		284288								

其他相关方法

- 还可以类似地应用其它相关方法。
- 最简单的是条件概率函数 (CPF)分析，可参照这篇发表于《大气环境》的文献，对比了条件概率函数与非参数回归方法。(Kim, E., and Hopke, P. K., 2004. Comparison between conditional probability function and nonparametric regression for fine particle source directions. *Atmospheric Environ.*, **38**: 4667-4673.)

其他相关方法

- 发表于《环境科学技术》的一篇文献（Henry, R., G.A. Norris, R. Vedantham, and J.R. Turner. 2009. Environ. Sci. Technol. 43:4090–4097 ）介绍了NPR模型的扩展用法，包括风向和风速，该方法称为非参数风回归方法（Nonparametric Wind Regression Methodology） 。

其他相关方法

- 发表于《大气污染研究》的一篇文献（Vedantham, R., G. Norris, S.G. Brown, and P. Roberts. 2012. Atmos. Pollut. Res. 3:105–111）描述了持续风关联法（Sustained Wind Incidence Method ， SWIM），是NPR和NWR方法的扩展。
- 但是，这些分析方法目前都没有公开可得的操作软件。
- 文献中给出了数学方法，然而需要进行编程。

高时间分辨率数据

- 可以看出，自动气相色谱仪得出的小时数据对于上述分析都发挥了重要作用。
  - 可为PMF分析提供更好的数据。
  - 也可用于结合风数据的分析方法，提供来源和方向解析。

中国的VOC源解析

- 已经有一些VOCs源解析研究成果发表了。报告附件中包括了这些已发表文献。

谢谢您的倾听!

提问?

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