
Modeling for VOCs Source Apportionment

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Outline

- Introduction
- Sampling and Analysis of VOCs
- Source Apportionment
 - Profiles Known
 - Profiles Unknown
- Summary



Introduction

- Why do VOCs matter?
 - Direct health effects
 - Acute – carbonyls and irritation responses
 - Chronic – PAHs and cancer
 - Drive atmospheric chemistry
 - Ozone formation
 - SOA formation



Introduction

- What VOCs matter?
 - PAHs
 - Reactive hydrocarbon compounds
 - Olefins including terpenes and isoprene
 - Aromatics like benzene, toluene, xylenes
 - Aliphatics to a lesser extent



Sampling and Analysis

- In the US, we have sites to look at the hydrocarbon compounds most related to ozone production.
- These are **Photochemical Assessment Monitoring Stations (PAMS)**
- They are described at <http://www3.epa.gov/ttnamti1/pamsmain.html>



Sampling and Analysis

- Sampling
 - Semicontinuous in-situ sampling and analysis
 - Auto-GC
 - Integral samples
 - Canisters returned to the laboratory for sampling.



Sampling and Analysis

VOC Measurement Technologies

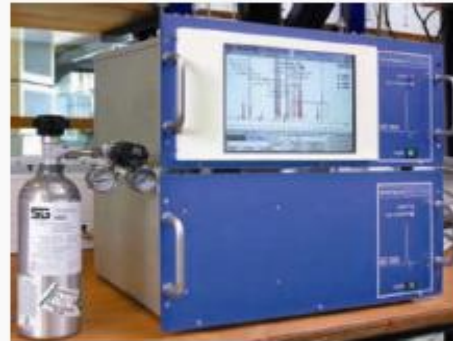
Canisters

vs

Auto-GCs



- Data averaged over sampling period
- Low capital cost
- Continuing lab/shipping costs
- Manually intensive
- Canister “artifacts”



- Hourly data
- Higher capital cost
- Higher skill level required to run and analyze data
- Difficulty resolving some compounds



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Sampling and Analysis

Hydrocarbons		
Ethylene	3-Methylpentane	Styrene
Acetylene	2-Methyl-1-Pentene	o-Xylene
Ethane	n-hexane	n-Nonane
Propylene	Methylcyclopentane	Isopropylbenzene
Propane	2,4-dimethylpentane	n-Propylbenzene
Isobutane	Benzene	m-Ethyltoluene
1-Butene	Cyclohexane	p-Ethyltoluene
n-Butane	2-methylhexane	1,3,5-Trimethylbenzene
t-2-Butene	2,3-dimethylpentane	o-Ethyltoluene
c-2-Butene	3-methylhexane	1,2,4-trimethylbenzene
Isopentane	2,2,4-trimethylpentane	n-Decane
1-Pentene	n-Heptane	1,2,3-trimethylbenzene
n-Pentane	Methylcyclohexane	m-Diethylbenzene
Isoprene	2,3,4-trimethylpentane	p-Diethylbenzene
t-2-pentene	Toluene	n-Undecane
c-2-pentene	2-methylheptane	
2,2-Dimethylbutane	3-methylheptane	Carbonyls
Cyclopentane	n-Octane	Formaldehyde
2,3-dimethylbutane	Ethylbenzene	Acetone
2-methylpentane	m&p-Xylenes	Acetaldehyde



Which system is best for source apportionment?

- The auto-GC provides hourly data so 24 samples a day
- Canisters provide up to 8 3-hour samples a day, but most canister sites simply collect 2 per day (morning and afternoon) and miss the overnight hours.

In general, auto-GCs are highly preferred for source apportionment studies.



Apportionment Methods

- All of the apportionment methods work on the basis of a mass balance approach
- That is the concentrations we observe are additive contributions from a set of independent source types.
- Thus, we will do a mass balance analysis



Mass Balance

A mass balance equation can be written to account for all m chemical species in the n samples as contributions from p independent sources

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

Where $i = 1, \dots, n$ samples, $j = 1, \dots, m$ species and $k = 1, \dots, p$ sources



Receptor Modeling

- The question is then what is known *a priori* to solve this equation.
- Divide the problem into two classes
 - Source Profiles Known
 - Source Profiles Unknown



Receptor Modeling

- **SOURCES PROFILES KNOWN**
 - **Chemical Mass Balance**
 - **Multivariate Calibration Methods**
 - Partial Least Squares
 - Artificial Neural Networks
 - Simulated Annealing
 - Genetic Algorithm



Chemical Mass Balance Model

The mass balance equation can be rewritten as a regression problem where the profiles and the ambient concentrations are known.

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

Where the equation is now written for one sample at a time.



Chemical Mass Balance

- The CMB model is then a regression problem. However, there are errors in both the dependent and independent variables so it is necessary to solve the problem using error models.
- The EPA has adopted the effective variance least-squares approach and incorporated it in CMB 8 that is available at www.epa.gov/ttn/SCRAM



Chemical Mass Balance

- The CMB model has been widely used for PM_{10} apportionment in the western US and for fine particle organic carbon apportionment based on specific organic species (“molecular markers”) that have been identified in a series of source emissions by the late Glen Cass and his former students.



Chemical Mass Balance

- The key issue in the application of the CMB are knowing the profiles
- It is difficult and expensive to perform emissions sampling and very few sources have been examined. Thus, profiles may not be available for the specific source types needed.
- Very little is known with respect to the variability in composition in the profiles for a given source type.



Receptor Modeling

- SOURCES PROFILES UNKNOWN
 - Factor Analysis
 - Principal Components Analysis
 - Absolute Principal Components Analysis
 - SAFER/UNMIX
 - Positive Matrix Factorization



Factor Analysis

- We do not have time today to get into the details of these various approaches.
- One method has become the most widely used method because it has the ability to make more complete use of the data and what is known regarding the related measurement uncertainties.
- That method is Positive Matrix Factorization (PMF)



Positive Matrix Factorization

- Explicit least-squares approach to solving the factor analysis problem
- Individual data point weights
- Imposition of natural and other constraints, and
- Flexibility to build more complicated models



Positive Matrix Factorization

- The Objective Function, Q , is defined by

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

where σ_{ij} is an estimate of the uncertainty in x_{ij}



Positive Matrix Factorization

- The US EPA version of PMF (version 5) is available at <https://www.epa.gov/air-research/positive-matrix-factorization-model-environmental-data-analyses>



Positive Matrix Factorization

- There have been a number of application of PMF to VOC data. A good illustrative example is:
- 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.



Sampling and Analysis

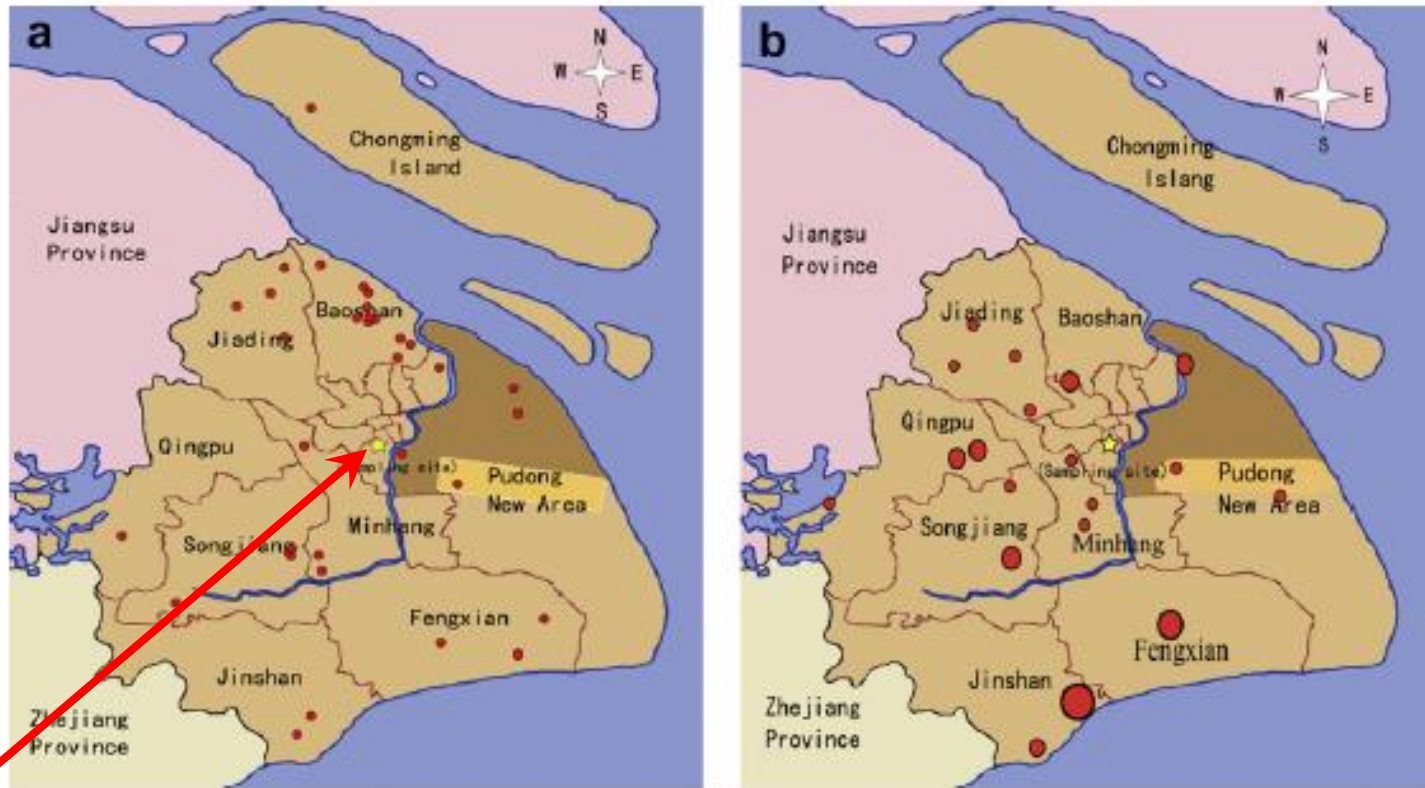


Fig. 1. Location of sampling site (yellow star) and the distribution of large smelter and steel factories (a) and large chemical industrial complex (b) in Shanghai.



Sampling and Analysis

- The sampling site is located at Xujiahui (XJH) commercial center of Shanghai
- VOCs were sampled at 6:00-9:00 for 3 h using a 6 L silonite canister with silonite valve (model 29-10622, Entech Instruments Inc., USA) from Jan. 2007 to Mar. 2010.
- To study the diurnal variations, VOCs were intensively measured (8 samples a day with a 3 h interval) from August 25 to September 20, 2009



Sampling and Analysis

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Sampling and Analysis

- Gas samples were pre-processed using an Entech Model 7100 VOC pre-concentrator.
- Analyzed by gas chromatography with a mass-selective detector.
- Thirty-two compounds were available for the source apportionment study



Wind Data

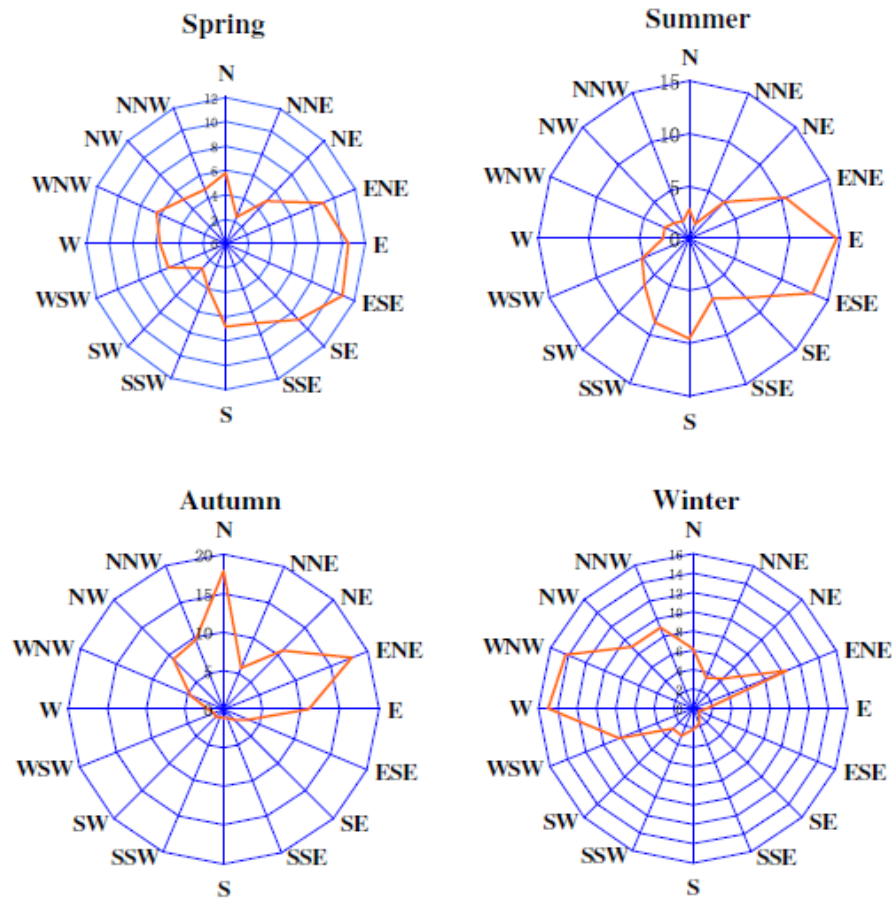


Fig. 2. The prevailing winds during different seasons in Shanghai.



Wind Data

- There are winds from different directions over the course of the year.
- This is helpful since now there are sources that are upwind and downwind at different times
- This variation provides a stronger basis for the PMF analysis.



PMF Details

- Using PMF, users need to choose a number of factors (p).
- The value of p is chosen based on several parameters, including:
 - the normalized sum of error squares in individual VOC concentrations (Q-value),
 - the normalized residual distributions for the individual VOC compounds, and
 - the physical interpretability of the derived source profiles



PMF Details

- Uncertainties need to assigned to each data value.
 - $U = \sqrt{(EF \times conc)^2 + (MDL)^2}$ Conc > MDL
 - where EF represents an error fraction (EF = the percent uncertainty/100), and MDL represents the method detection limit.
- If the concentration is less than or equal to the MDL, the calculation is:
 - $U = (5/6) \times MDL$ Conc < MDL



PMF Results

- Eight factors were selected according to the resulted stable Q values and interpretability of the resulting profiles.
- In order to better classify the automobile sources, three important components associated with vehicular emissions of VOCs were identified in this study, including vehicular exhaust (about 40%), fuel evaporation from tank (about 40%), and internal engine burned and unburned emissions from crankcase ventilation (about 20%).

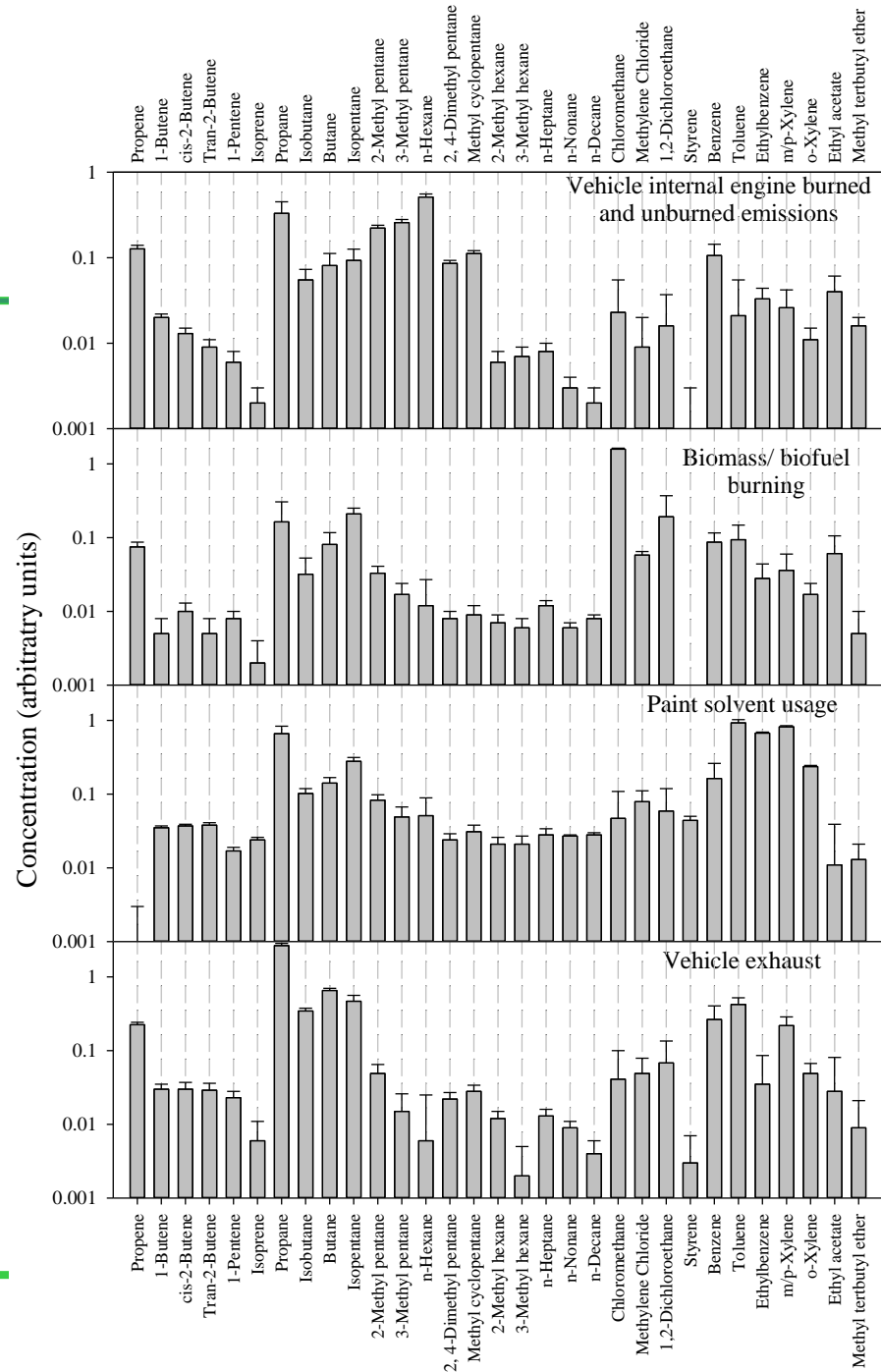


PMF Results

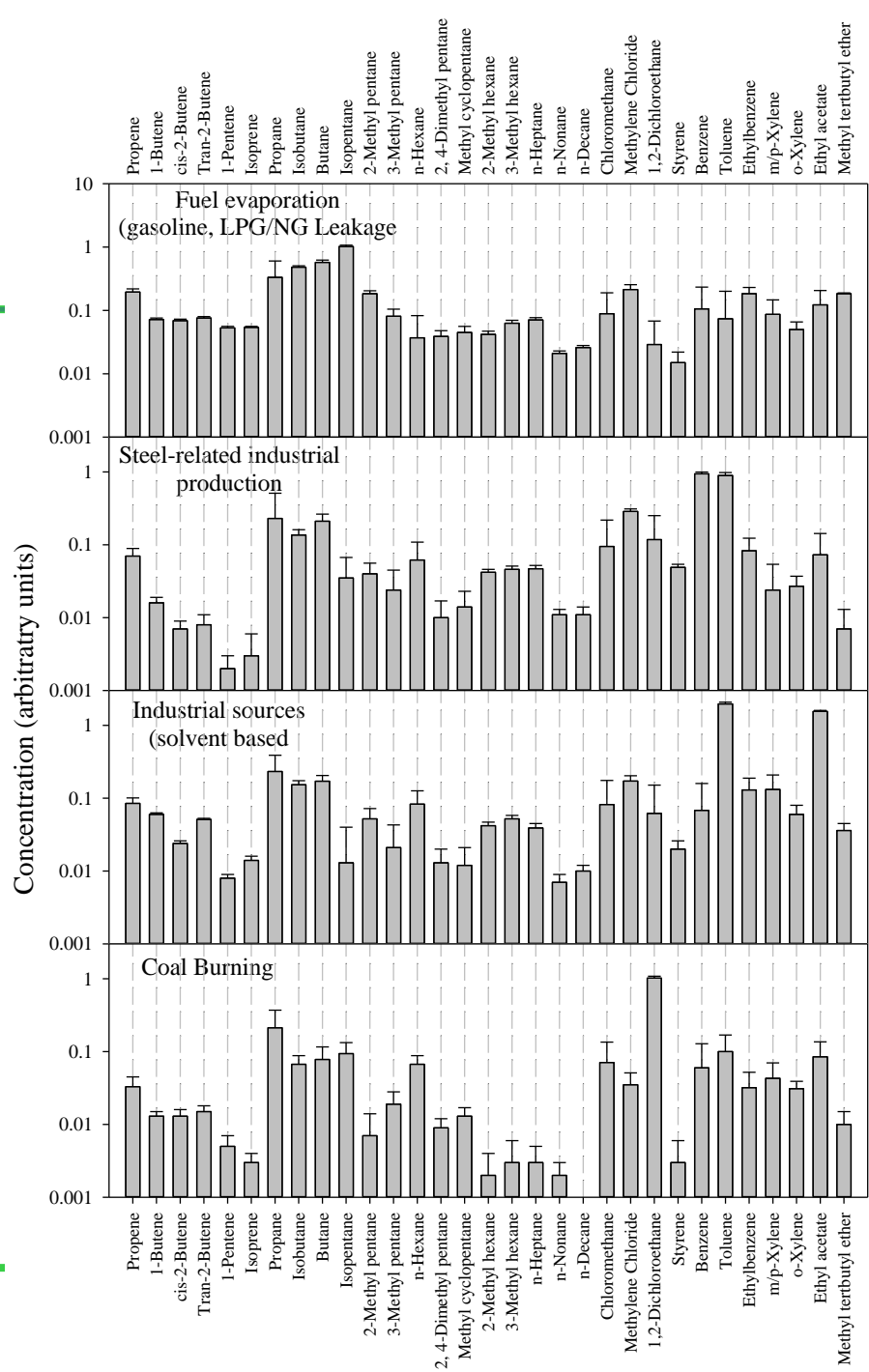
- The details of how the profiles were assigned to the various source types is described by Cai et al.
- There is not enough time now to discuss how each profile is attributed to a source type.



PMF Results



PMF Results



PMF Results

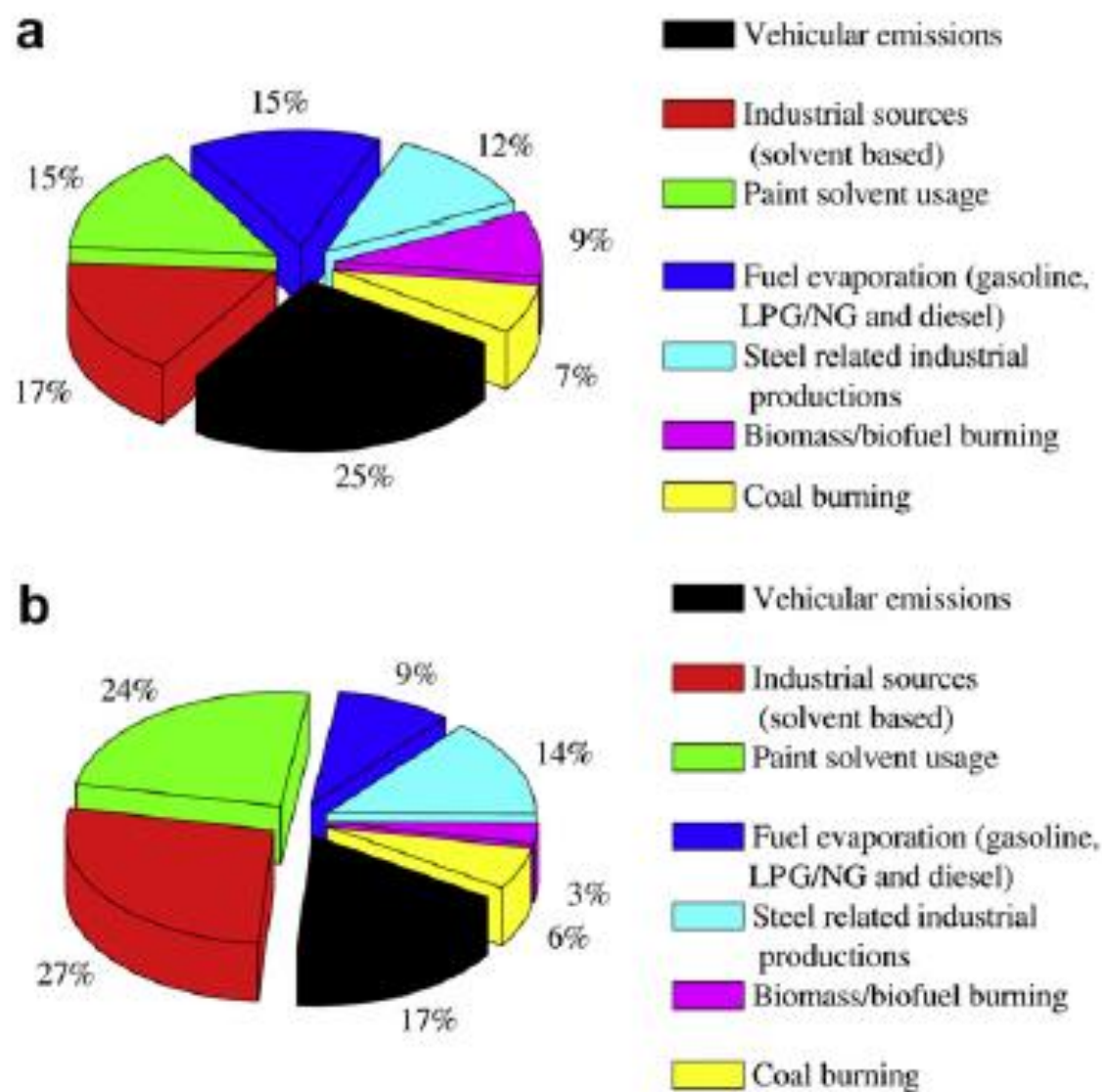


Fig 6. a, Individual contributions of VOC sources to the measured VOC concentrations. b, Same to a, except for ozone formation potential.

Additional Methods

- With the higher time resolved data available from an auto-GC system, it is possible to use other methods to identify and apportion the VOCs to their sources.
- An example is the use of non-parametric regression by Henry et al. (Atmospheric Environment 36 (2002) 2237–2244) to identify several major sources of VOCs in Houston, TX.



Non-Parametric Regression

- This approach combines the concentration data for any given compound with the wind direction during the hour during which the concentration was measured.



Non-Parametric Regression

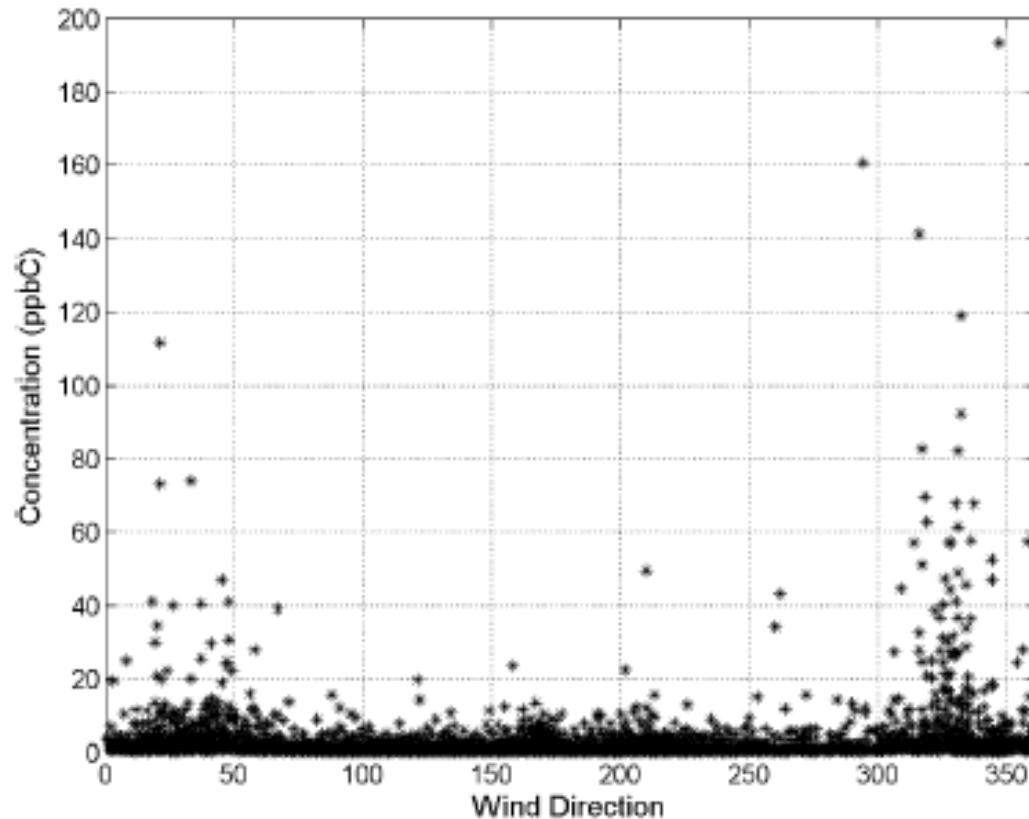


Fig. 1. Hourly cyclohexane measured at Deer Park during 1997 versus the azimuth of the wind direction.



Nonparametric regression (NPR)

- Analyzing source contribution vs wind direction.
- A regression model without parameters since it estimates expected value of concentration given wind direction.
- The average concentration over a sliding window of width $\Delta\theta$ centered at θ

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

W_i : measured wind direction

C_i : measured concentration for the i th sample

n : total number of samples

- To give different weights to the measurements, a Gaussian kernel function, $K(x)$, is used and defined as

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



Nonparametric regression (NPR)

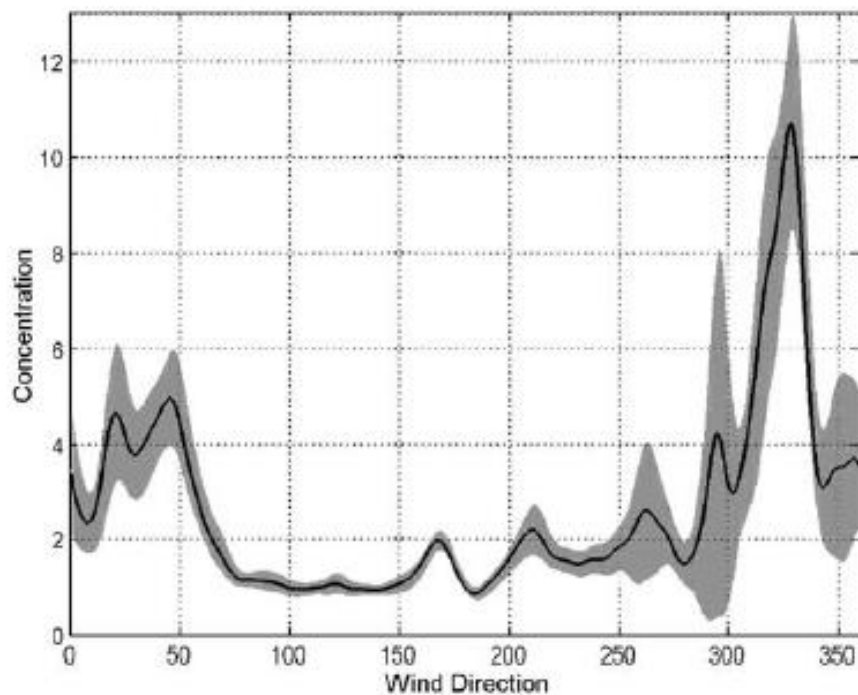


Fig. 4. Nonparametric regression of cyclohexane versus wind direction using a Gaussian kernel with a 10° FWHM. Data with wind speed < 1 mile/h are excluded. The gray region is the 95% confidence interval.



Nonparametric regression (NPR)

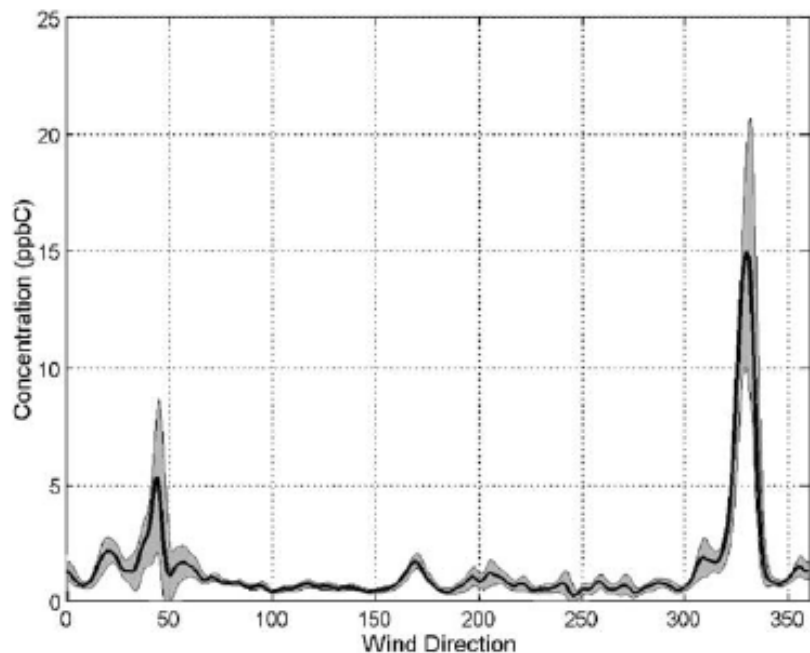


Fig. 6. Nonparametric regression of cyclohexane at Deer Park using a Gaussian kernel with a FWHM of 5. Data are restricted to periods with wind speed > 6 miles/h (about 1 h travel time from the largest source to the site). The gray region is the 95% confidence interval.

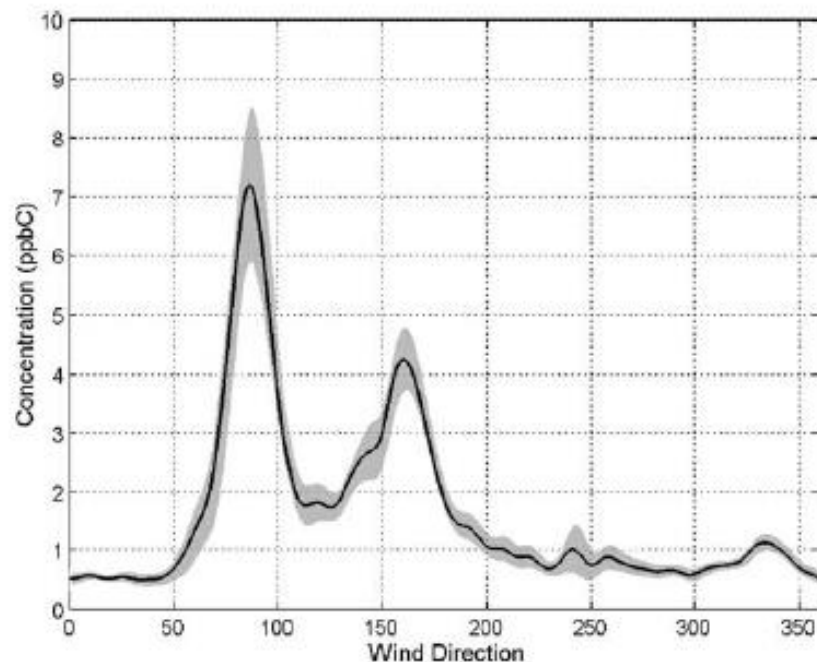


Fig. 7. Nonparametric regression of cyclohexane at Clinton Drive using a Gaussian kernel with a FWHM of 10. Data are restricted to periods with wind speed > 5 miles/h (about 1 h travel time from the largest source to the site). The gray region is the 95% confidence interval.



Nonparametric regression (NPR)

Table 1
Emissions of cyclohexane for 1997 in Harris Co., TX

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								

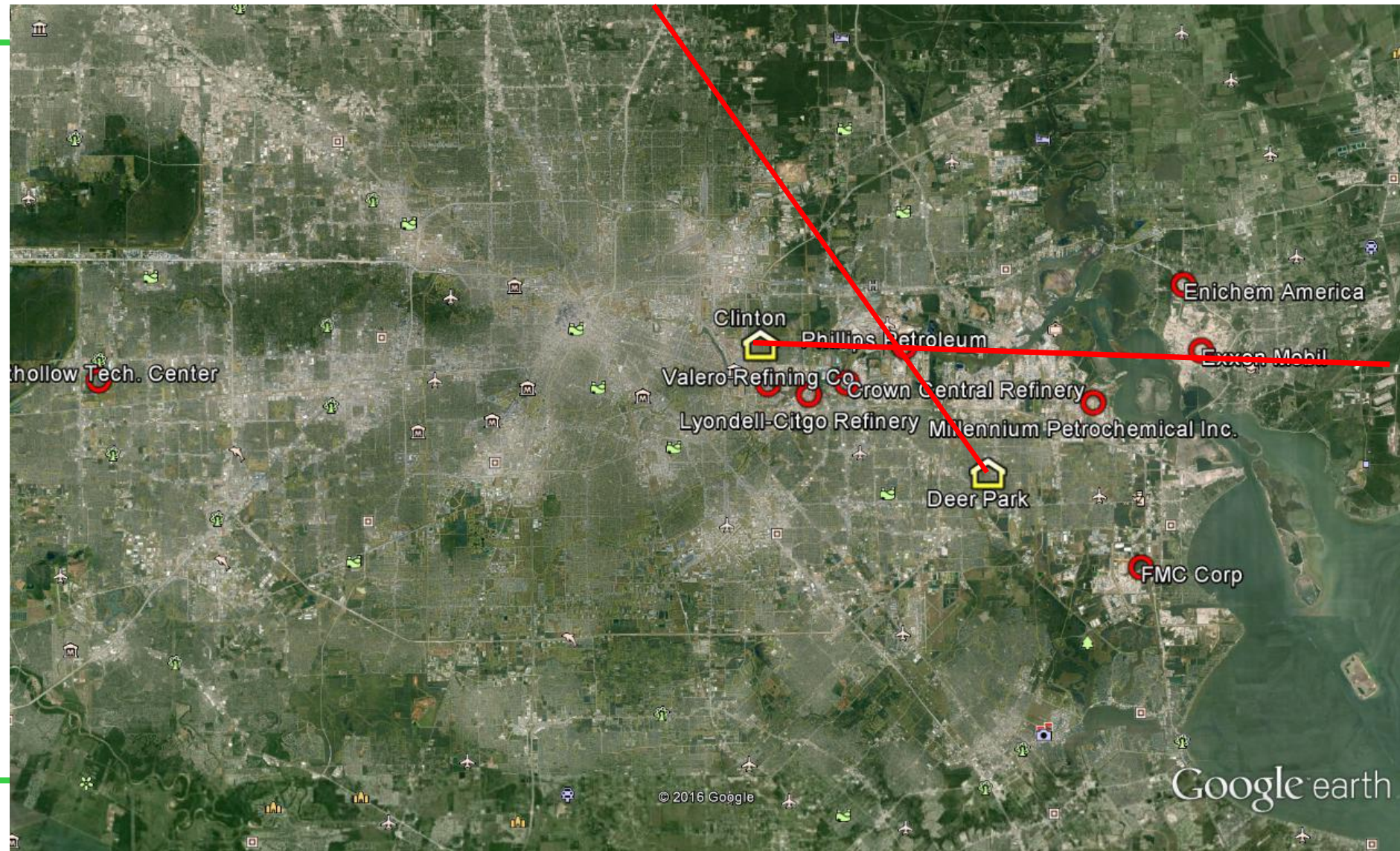


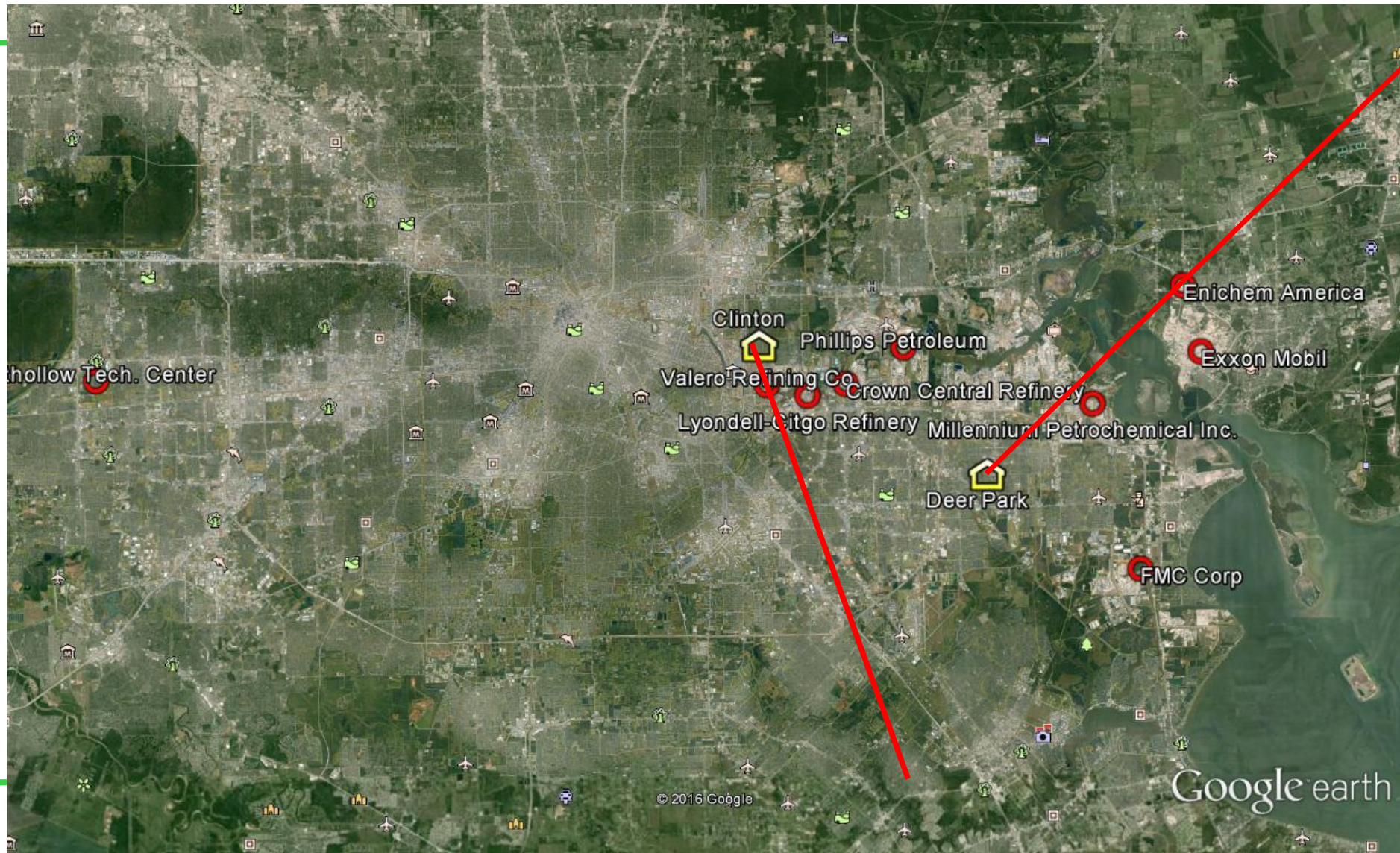
Nonparametric regression (NPR)

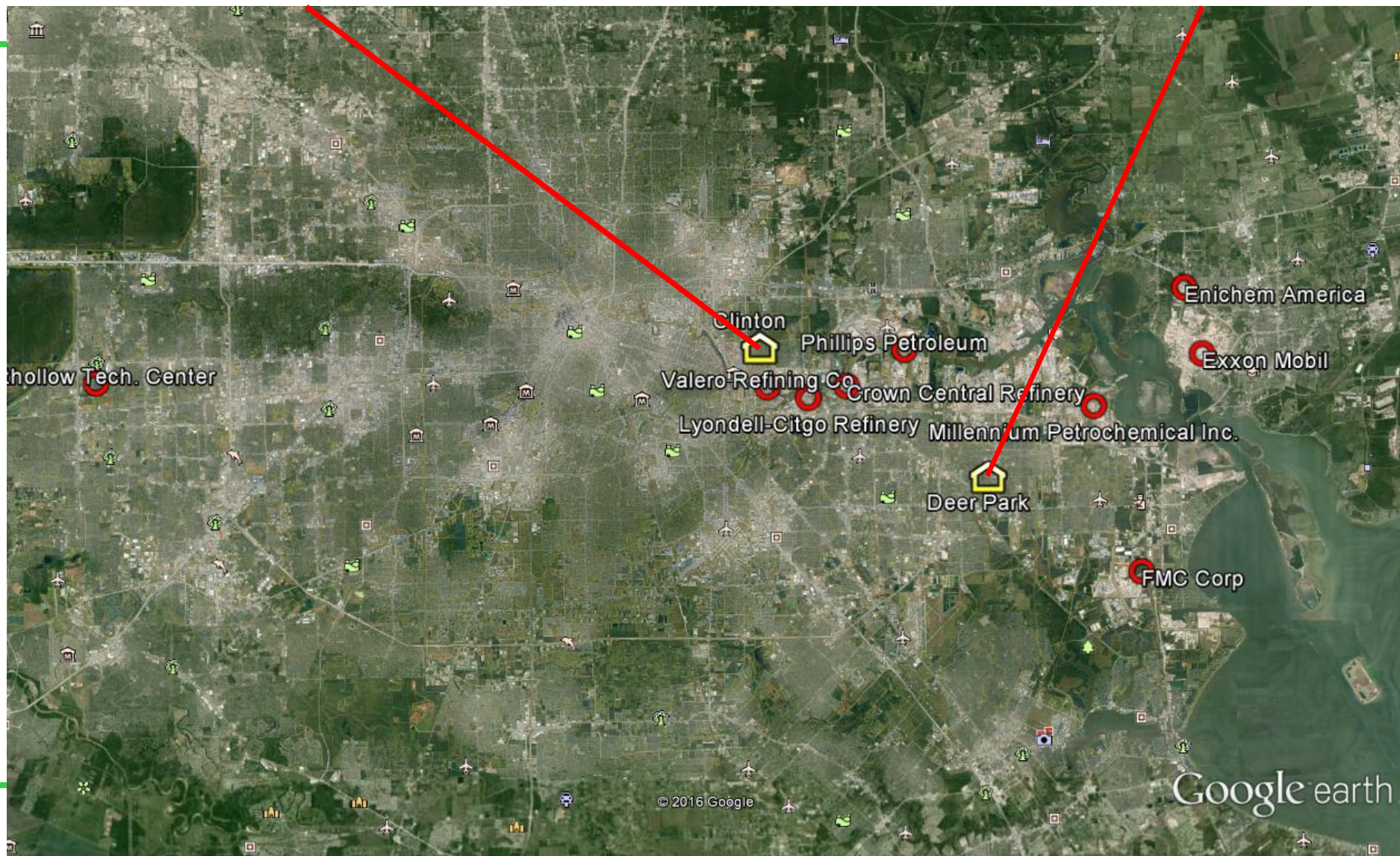
Table 2
Largest peaks in the nonparametric regression of cyclohexane on wind direction, Figs. 6 and 7

	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



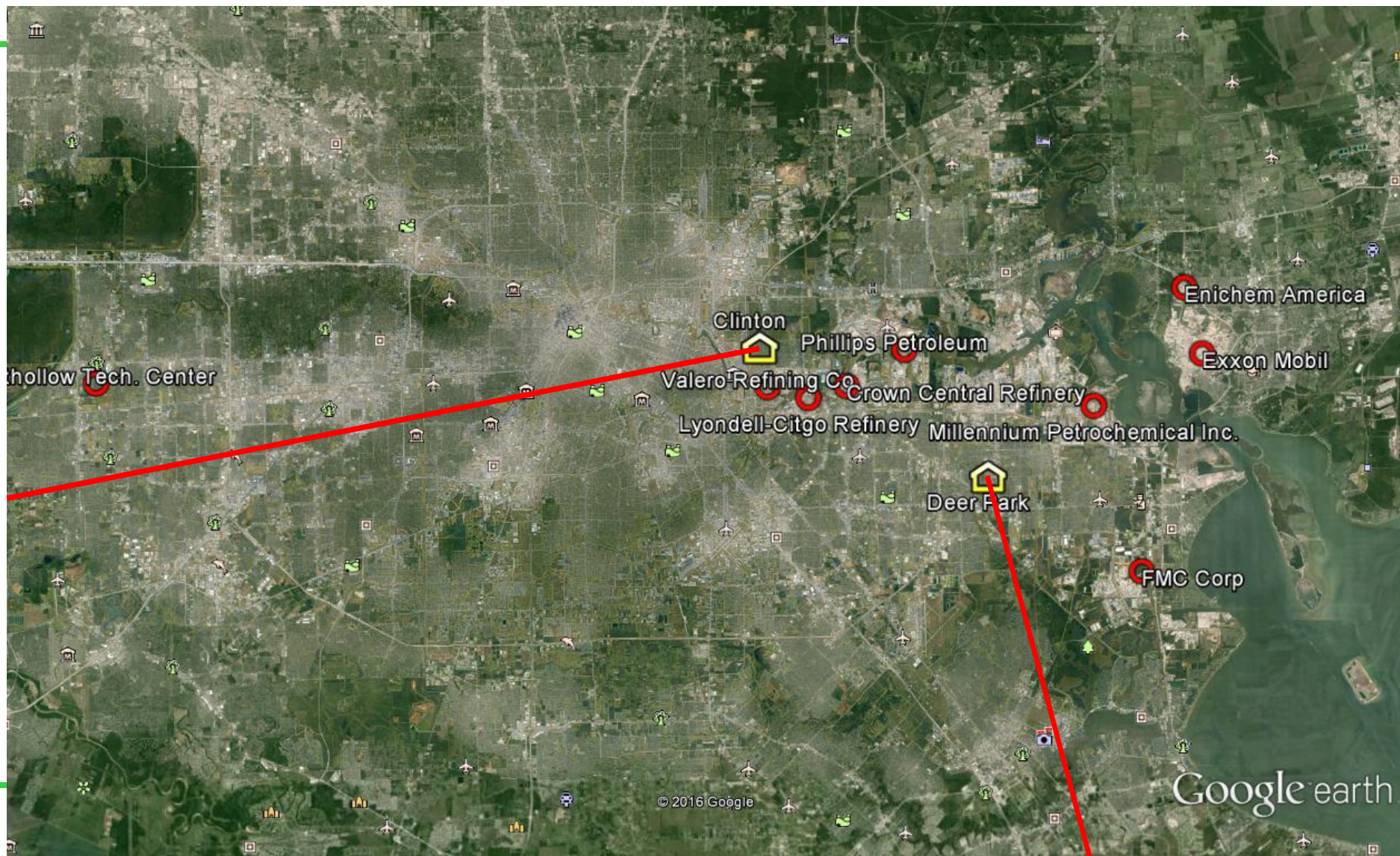






Google earth

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Nonparametric regression (NPR)

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Total emissions		284288								



Other Related Methods

- There are other related methods that could be similarly applied.
- Simplest is Conditional Probability Function (CPF) analysis (see 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.)



Other Related Methods

- Henry, R., G.A. Norris, R. Vedantham, and J.R. Turner. 2009. Environ. Sci. Technol. 43:4090–4097 introduce an extension of NPR to include both wind direction and wind speed that they call Nonparametric Wind Regression Methodology.



Other Related Methods

- Vedantham, R., G. Norris, S.G. Brown, and P. Roberts. 2012. Atmos. Pollut. Res. 3:105–111 describe Sustained Wind Incidence Method (SWIM) further extends NPR and NWR.
- However, no publically available software is currently available to perform these analyses.
- The mathematics are provided in the papers, but it would need to be programmed.



Highly Time Resolved Data

- It can be seen that having hourly data as one gets from an auto-GC has significant advantages for both types of analyses.
 - It will provide better input to PMF
 - It will permit these wind analysis approaches that will provide both apportionment and directionality of the sources.



VOC Source Apportionment in China

- There have been a number of journal papers on apportionment of VOCs.
- A list is appended to this presentation.



Thank you for your attention!

Questions?



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Papers on Source Apportionment

Ambient volatile organic compounds and their effect on ozone production in Wuhan, central China By: Lyu, X. P.; Chen, N.; Guo, H.; et al. SCIENCE OF THE TOTAL ENVIRONMENT Volume: 541 Pages: 200-209 Published: JAN 15 2016

On-road emission characteristics of VOCs from light-duty gasoline vehicles in Beijing, China By: Cao, Xinyue; Yao, Zhiliang; Shen, Xianbao; et al. ATMOSPHERIC ENVIRONMENT Volume: 124 Pages: 146-155 Part: B Published: JAN 2016

Process-specific emission characteristics of volatile organic compounds (VOCs) from petrochemical facilities in the Yangtze River Delta, China By: Mo, Ziwei; Shao, Min; Lu, Sihua; et al. SCIENCE OF THE TOTAL ENVIRONMENT Volume: 533 Pages: 422-431 Published: NOV 15 2015

Characterization of VOC sources in an urban area based on PTR-MS measurements and receptor modelling By: Stojic, A.; Stojic, S. Stanisic; Sostaric, A.; et al. ENVIRONMENTAL SCIENCE AND POLLUTION RESEARCH Volume: 22 Issue: 17 Pages: 13137-13152 Published: SEP 2015

Vehicular volatile organic compounds losses due to refueling and diurnal process in China: 2010-2050 By: Yang, Xiaofan; Liu, Huan; Cue, Hongyang; et al. JOURNAL OF ENVIRONMENTAL SCIENCES Volume: 33 Pages: 88-96 Published: JUL 1 2015

Emission and profile characteristic of volatile organic compounds emitted from coke production, iron smelt, heating station and power plant in Liaoning Province, China By: Shi, Jianwu; Deng, Hao; Bai, Zhipeng; et al. SCIENCE OF THE TOTAL ENVIRONMENT Volume: 515 Pages: 101-108 Published: MAY 15 2015



Papers on Source Apportionment

Characterization and source apportionment of volatile organic compounds in urban and suburban Tianjin, China
By: Han Meng; Lu Xueqiang; Zhao Chunsheng; et al. ADVANCES IN ATMOSPHERIC SCIENCES Volume: 32 Issue: 3 Pages: 439-444 Published: MAR 2015

Characterization of ambient volatile organic compounds and their sources in Beijing, before, during, and after Asia-Pacific Economic Cooperation China 2014
By: Li, J.; Xie, S. D.; Zeng, L. M.; et al. ATMOSPHERIC CHEMISTRY AND PHYSICS Volume: 15 Issue: 14 Pages: 7945-7959 Published: 2015

Do vehicular emissions dominate the source of C6-C8 aromatics in the megacity Shanghai of eastern China? By: Wang, Hongli; Wang, Qian; Chen, Jianmin; et al. JOURNAL OF ENVIRONMENTAL SCIENCES-CHINA Volume: 27 Pages: 290-297 Published: JAN 1 2015

Sources of C-2-C-4 alkenes, the most important ozone nonmethane hydrocarbon precursors in the Pearl River Delta region
By: Zhang, Yanli; Wang, Xinming; Zhang, Zhou; et al. SCIENCE OF THE TOTAL ENVIRONMENT Volume: 502 Pages: 236-245 Published: JAN 1 2015

Characteristics and source apportionment of VOCs measured in an industrial area of Nanjing, Yangtze River Delta, China
By: An, Junlin; Zhu, Bin; Wang, Honglei; et al. ATMOSPHERIC ENVIRONMENT Volume: 97 Special Issue: SI Pages: 206-214 Published: NOV 2014

Source apportionment of VOCs in a suburb of Nanjing, China, in autumn and winter
By: Xia, Li; Cai, Changjie; Zhu, Bin; et al. JOURNAL OF ATMOSPHERIC CHEMISTRY Volume: 71 Issue: 3 Pages: 175-193 Published: SEP 2014

The characteristics, seasonal variation and source apportionment of VOCs at Gongga Mountain, China
By: Zhang, Junke; Sun, Yang; Wu, Fangkun; et al. ATMOSPHERIC ENVIRONMENT Volume: 88 Pages: 297-305 Published: MAY 201



Papers on Source Apportionment

Contribution of VOC sources to photochemical ozone formation and its control policy implication in Hong Kong By: Ling, Z. H.; Guo, H. ENVIRONMENTAL SCIENCE & POLICY Volume: 38 Pages: 180-191 Published: APR 2014

Source Profiles and Chemical Reactivity of Volatile Organic Compounds from Solvent Use in Shanghai, China By: Wang, Hongli; Qiao, Yuezhen; Chen, Changhong; et al. AEROSOL AND AIR QUALITY RESEARCH Volume: 14 Issue: 1 Pages: 301-310 Published: FEB 2014

A temporally and spatially resolved validation of emission inventories by measurements of ambient volatile organic compounds in Beijing, China By: Wang, M.; Shao, M.; Chen, W.; et al. ATMOSPHERIC CHEMISTRY AND PHYSICS Volume: 14 Issue: 12 Pages: 5871-5891 Published: 2014

Species profiles and normalized reactivity of volatile organic compounds from gasoline evaporation in China By: Zhang, Yanli; Wang, Xinming; Zhang, Zhou; et al. ATMOSPHERIC ENVIRONMENT Volume: 79 Pages: 110-118 Published: NOV 2013

A new monitoring-simulation-source apportionment approach for investigating the vehicular emission contribution to the PM_{2.5} pollution in Beijing, China By: Cheng, Shuiyuan; Lang, Jianlei; Zhou, Ying; et al. ATMOSPHERIC ENVIRONMENT Volume: 79 Pages: 308-316 Published: NOV 2013

Evidence of coal combustion contribution to ambient VOCs during winter in Beijing By: Wang, Ming; Shao, Min; Lu, Si-Hua; et al. CHINESE CHEMICAL LETTERS Volume: 24 Issue: 9 Pages: 829-832 Published: SEP 2013



Papers on Source Apportionment

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