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Author: Olaguer, Eduardo P.

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Overview of the Benzene and Other Toxics Exposure (BEE-TEX) Field Study

Eduardo P. Olaguer

Houston Advanced Research Center (HARC), The Woodlands, TX, USA.

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ABSTRACT: The Benzene and other Toxics Exposure (BEE-TEX) field study was an experimental campaign designed to demonstrate novel methods for measuring ambient concentrations of hazardous air pollutants (HAPs) in real time and to attribute these concentrations to quantified releases from specific emission points in industrial facilities while operating outside facility fence lines. BEE-TEX was conducted in February 2015 at three neighboring communities in the Houston Ship Channel of Texas, where a large number of petrochemical facilities are concentrated. The novel technologies deployed during BEE-TEX included: (1) tomographic remote sensing based on differential optical absorption spectroscopy; (2) real-time broadcasting of ambient air monitoring data over the World Wide Web; (3) real-time source attribution and quantification of HAP emissions based on either tomographic or mobile measurement platforms; and (4) the use of cultured human lung cells *in vitro* as portable indicators of HAP exposure.

KEYWORDS: hazardous air pollutants, air toxics, human exposure, source attribution, remote sensing, real-time monitoring, microscale modeling

SUPPLEMENT: Ambient Air Quality (B)

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CORRESPONDENCE: eolaguer@harcresearch.org

Introduction

In the United States, economically or racially disadvantaged communities frequently coexist side by side with a major industry, such as in the Houston Ship Channel region of Texas, where there is a large concentration of refineries, olefin plants, and other petrochemical facilities in the midst of African-American or Hispanic neighborhoods. Addressing human health risk within such communities has become a major priority of the U.S. Environmental Protection Agency (USEPA). The USEPA's 2005 National-Scale Air Toxics Assessment found that cancer risk in Ship Channel census tracts could exceed 100 per million due to emissions of hazardous air pollutants (HAPs).[1] Earlier, Mayor Bill White of Houston had formed a Task Force that evaluated 179 HAPs and criteria pollutants and concluded that a dozen of these substances posed a definite risk to human health, including the carcinogen, benzene.[2]

The protection of human health due to ambient exposure to HAPs is made difficult by the state of monitoring and source attribution technologies for air toxics used by regulatory and enforcement agencies. As discussed by Olaguer et al.[3], conventional ambient air monitoring methods, such as Summa canisters, adsorption cartridges, and automated gas chromatographs suffer from severe flaws, such as poor temporal and spatial coverage, especially when winds are highly variable. Direct sampling of point source emissions with USEPA Method 21,[4] or more sophisticated short-range remote sensing using Differential Absorption Lidar[5] or open path Fourier Transform Infrared spectroscopy,[6] requires access within the fence line of facilities, and may thus be influenced by operator behavior.

In recent years, sophisticated real-time in situ measurement techniques using either optical methods (eg, quantum cascade lasers [QCLs]) or chemical ionization–mass spectrometry (CIMS) have become increasingly available to measure greenhouse gases, ozone precursors, and air toxics.[7] Such instruments typically have sub-ppb detection limits and response times less than one second and can be mounted on mobile platforms to more effectively target and measure pollution plumes. Mobile monitoring can be complemented by long-path remote sensing techniques, including various flavors of differential optical absorption spectroscopy (DOAS).[8] For example, an imaging DOAS can measure emission fluxes from individual combustion units, such as flares and catalytic crackers, using the sun as a light source and data from nearby wind monitors.[9] Moreover, there has been greater use of mathematically intensive inverse modeling techniques to pinpoint and quantify emissions from industrial facilities based on research-grade ambient measurements. The 2009 Study of Houston Atmospheric Radical Precursors (SHARP) conducted in the

Houston Ship Channel region demonstrated the marriage of all these techniques simultaneously in quantifying refinery and other petrochemical emissions, and attributing them to individual emission points, all while operating exclusively outside facility fence lines.[7]

More recently, contemporary real-time methods were applied to urban air toxics monitoring during the 2015 Benzene and other Toxics Exposure (BEE-TEX) field study. Like SHARP, BEE-TEX was conceived and implemented by the Houston Advanced Research Center (HARC) with the help of a number of institutions that were involved in both campaigns. The main scientific participants included the University of California at Los Angeles, the University of North Carolina (UNC), and Aerodyne Research, Inc. (Aerodyne), with ancillary participation by the University of Houston (UH) and Rice University (Rice). Logistical support was provided by the Houston Regional Monitoring Corporation, the Port of Houston Authority, the Port Terminal Railroad Association, the Texas Department of Transportation, the City of Houston, and the City of Galena Park.

The goal of BEE-TEX was to facilitate the establishment of cost-effective, state-of-the-art, neighborhood-scale monitoring networks in the Houston Ship Channel and other areas of high toxic emissions around the country, the improvement of emission inventories, the conduct of epidemiological studies for air toxics, and ultimately, the enforcement of air quality regulations. BEE-TEX went beyond SHARP in demonstrating the feasibility of (1) intensive monitoring of an entire community using tomographic scanning techniques; (2) real-time data broadcasting and source attribution; and (3) *in vitro* monitoring of human exposure using cultured human lung cells. Olaguer et al.[10] have already described the application of mobile monitoring techniques to detect pipeline leaks of benzene in the first published study resulting from BEE-TEX. Additional major findings from the field study will be documented in the other publications currently in preparation or in review. This paper provides a review of the design and implementation of BEE-TEX, while other papers in this special supplement provide greater detail on particular aspects of the field study.

Study Design

The BEE-TEX study area consisted of three neighboring industrial fenceline communities: Galena Park on the north shore of the Houston Ship Channel, Manchester on the south shore, and Milby Park further south of the Ship Channel (Fig. 1). Galena Park and Manchester both have port terminals frequented by marine, rail, and truck traffic. Manchester is home to a major refinery, while Milby Park is the site of a large rubber processing plant. Petrochemical facilities in these communities are connected by a network of underground chemical pipelines carrying feedstock and refined products, which are also stored in arrays of above ground storage tanks. The large number of industrial point

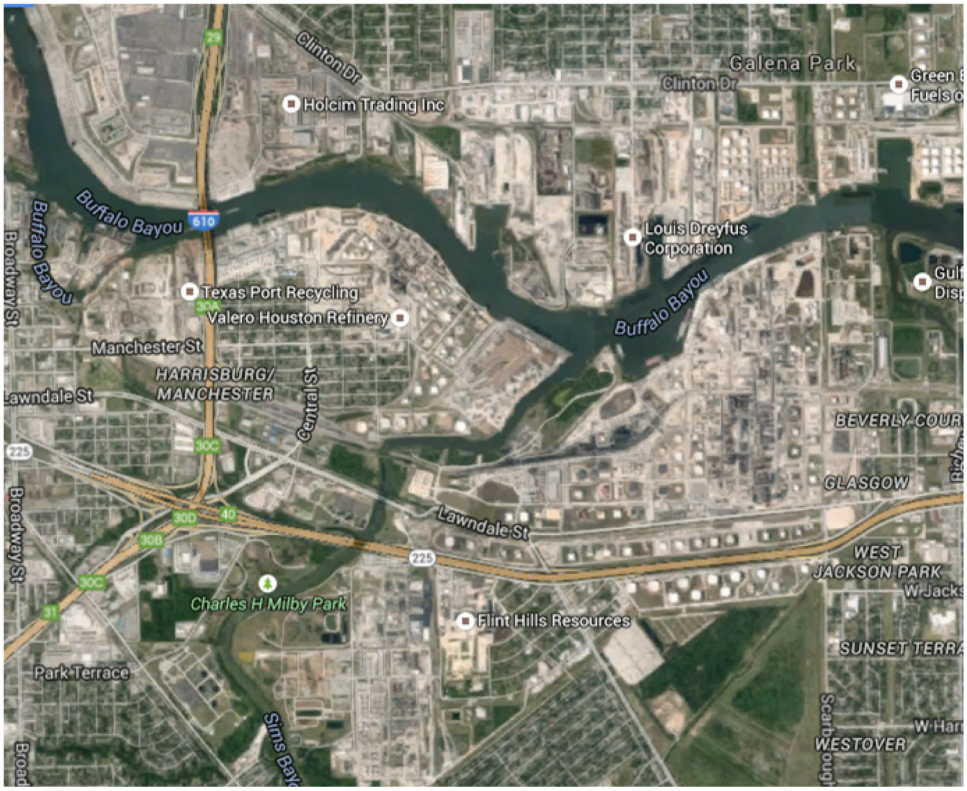

Figure 1. Google maps image of the BEE-TEX study area. The top of the figure points north.

sources colocated with residential communities made the BEE-TEX study area an ideal test bed for several innovative technologies and approaches for monitoring and attributing toxic air pollution.

One of the most important technologies demonstrated during BEE-TEX was the combination of remote sensing based on the long-path DOAS technique and computer-aided tomography (CAT). The particular implementation of DOAS for the BEE-TEX field study integrated several narrow band light-emitting diodes (LEDs) in the spectral regions where toxic aromatics, including the so-called BTEX compounds (benzene, toluene, ethyl benzene, and xylenes), absorb radiation. (Overlaps with other species are easily accounted for due to foreknowledge of the corresponding spectra.) Two geographically separated sets of LED sources were used to generate overlapping light beams that were reflected back by an array of mirrors encircling the community of interest toward detectors colocated with the light sources. The multiple overlapping light paths made it possible to mathematically infer a two-dimensional concentration map on the plane of the DOAS measurements from the measured spectral absorption and associated average concentration over each light path using the Algebraic Reconstruction Technique (ART)[11,12] or other tomographic reconstruction methods.

During BEE-TEX, a CAT-DOAS network was set up and operated continuously for the entire month of February 2015 in Manchester, as shown in Figure 2. Three retroreflector mirrors were mounted on mobile towers beside a rail yard along the southern end of Manchester, while two others were mounted on a bridge across the Ship Channel. The light sources and detectors were mounted on two 45′ scaffolding towers, the first at a local park and the second near the main entrance of a major refinery. Unfortunately, tall trees blocked some key sight lines and prevented at least two light paths from being activated in the network. However, the most interesting emission events occurred away from the degraded areas of the network, allowing the experiment to proceed successfully despite the problem. Approximately half an hour was required to complete one cycle of DOAS light path measurements.

To complement the area-wide CAT scans, three mobile laboratories equipped with real-time instrumentation were deployed by Aerodyne, HARC, and UH within the BEE-TEX study area. Each mobile laboratory carried a proton transfer reaction–mass spectrometer[13] (PTR-MS), which is a type of CIMS instrument, in addition to a global positioning system and a portable weather station. The main air toxics monitored by PTR-MS were BTEX compounds and styrene. The Aerodyne mobile laboratory[14] was equipped with additional devices, including a QCL, and an alternative NO^+ ion source for the PTR-MS, which normally uses hydronium (H_3O^+) as a reagent ion. The former made it possible to conduct real-time measurements of formaldehyde (HCHO), which normally cannot be measured by PTR-MS without substantial drying of air samples,[15] while the latter enabled

Figure 2. Tomographic network configuration in the Manchester neighborhood of Houston during the BEE-TEX study.
Note: The top of the figure points north. Red lines indicate active light paths (figure courtesy of Jochen Stutz).

the Aerodyne PTR-MS to measure 1,3-butadiene using NO^+ as a reagent ion. Both HCHO and 1,3-butadiene are among the most important HAPs identified by Mayor White's Task Force as posing a significant risk to the health of Houston residents.[2] The HARC mobile laboratory also attempted to measure HCHO by coupling a simple dry ice cryogenic trap to the PTR-MS in addition to a Nafion dehumidifier, which not only limits unwanted reactions with water vapor but also allows the PTR-MS to be operated at lower ion kinetic energies, thus limiting interferences caused by molecular fragmentation. In the case of HCHO, however, the instrument sensitivity was only sufficient to measure ambient concentrations in excess of ~10 ppb, and so the HCHO measurements were largely disregarded. Further details of the HARC mobile laboratory operation during BEE-TEX, including the calibration and intercomparison of PTR-MS measurements, are provided by Olaguer et al.[10]

The Aerodyne and UH mobile laboratories both carried a much larger suite of instrumentation than the HARC mobile laboratory, in order to conduct measurements of several non-HAP pollutants, such as carbon monoxide, carbon dioxide, methane, sulfur dioxide, nitrogen oxides, and particulate matter. These compounds were useful in fingerprinting sources, such as distinguishing between combustion and fugitive emissions. The Aerodyne and HARC mobile laboratories roamed freely through the BEE-TEX study area, often guided by the DOAS CAT scans. The UH laboratory, on the other hand, operated in stationary mode at one of the DOAS tower locations, specifically near the entrance of the major refinery in Manchester.

The location of the UH laboratory coincided with the site of an experiment by UNC, in which cultured human pulmonary type II epithelial cells[16] were exposed *in vitro* to captured ambient air for four hours after noon (the most active period for photochemistry) using a novel exposure instrument.[17–24] The enzymes and proteins released by the lung cells, as well as the cell genetic response, were used to distinguish ambient exposure to various classes of air pollutant, including aldehyde and aromatic HAPs. While the Aerodyne and HARC mobile laboratories could be used to test the validity of ambient concentrations inferred from CAT-DOAS measurements throughout the Manchester neighborhood, the UH laboratory provided a continuous stream of ambient concentration measurements at a single site that could be compared with lung cell responses to ambient air pollution. In addition to PTR-MS and other chemical measurements, the UH team operated an aerosol Lidar to assess vertical mixing in the planetary boundary layer, which can influence the transport of initially elevated pollution plumes. The aerosol Lidar was likewise colocated with the UNC lung cell experiment.

In addition to demonstrating the potential of various new technologies, the scientific assets deployed during BEE-TEX enabled the following research questions to be investigated:

- How well does the current network of regulatory monitoring stations, five of which are located in the BEE-TEX study area (Galena Park, Clinton Drive, Manchester, Milby Park, and Cesar Chavez), characterize ambient HAP concentrations?
- Which toxic species create the greatest burden in the selected fenceline communities?
- Where do hot spots of ambient HAP exposure tend to occur, and how large are they?
- Which emission sources tend to be responsible for observed hot spots of HAPs?
- How well do current emission inventories characterize point source emissions in the BEE-TEX study area?
- How do marine, railroad, auto, and truck mobile sources compare to stationary sources?
- How do human lung cells respond to ambient peak concentrations of specific chemicals (eg, benzene) or compound classes (eg, aldehydes vs aromatics)?
- How does non-HAP pollution affect human lung cell response to air toxics?

Real-Time Data Broadcasting and Modeling Analyses

The HARC mobile laboratory played a unique role in the BEE-TEX field study by broadcasting PTR-MS, wind, and GPS data to the World Wide Web approximately every six seconds. The details of the real-time broadcast and Geographical Information System methodology were described by Olaguer et al.[3] in the context of oil and gas site measurements in the Eagle Ford Shale of South Texas prior to the BEE-TEX campaign. An Internet site visible only to a selected number of field study participants was used to display the past and current mobile laboratory location and attendant chemical and wind measurements, so that study participants could see when and where large transient pollution plumes were encountered by the HARC mobile laboratory. An example of a real-time broadcast is shown in Figure 3. The data broadcasting capability enabled adaptive monitoring of emission events to take place, such as when the HARC and Aerodyne mobile laboratories collaborated in mapping different sections of the same pollution plume. Thus, the HARC mobile laboratory often functioned like a "pathfinder" for the Aerodyne laboratory.

In addition to displaying mobile laboratory data, the HARC real-time broadcasting system also displayed contours of BTEX ambient concentrations derived from mathematical plume reconstructions based on DOAS CAT scan data and the ART technique. The ART technique was complemented by more computationally intensive and accurate CAT plume reconstructions based on the 4D variational data assimilation (4Dvar) method as described by Olaguer.[25] These more elaborate reconstructions were based on the HARC 3D microscale forward and adjoint air quality model that has been successfully documented

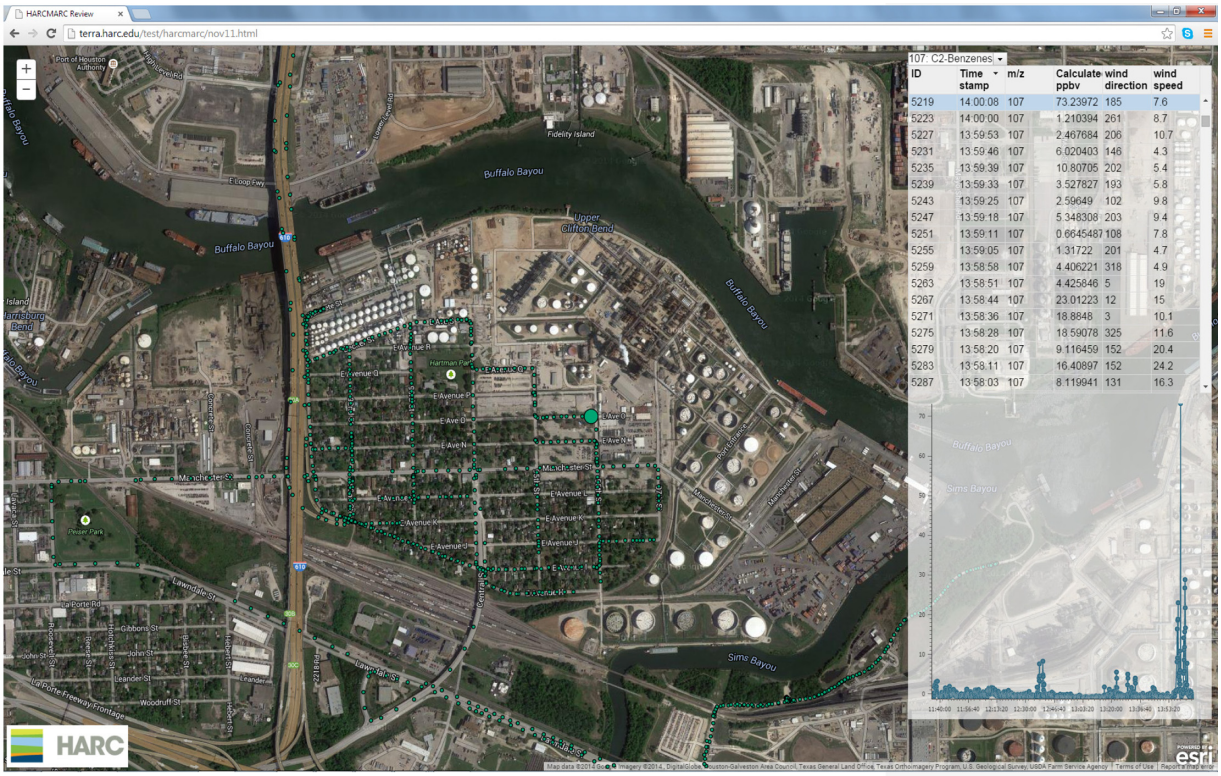

Figure 3. Web screenshot of the HARC real-time broadcasting system displaying mobile laboratory locations (green dots) and attendant measurements in both tabular and graphical form (right-hand side of screenshot). PTR-MS data are displayed for C2-benzenes (ethyl benzene plus xylenes).

and evaluated based on field measurements in several peer-reviewed publications.[26–30]

The HARC model simulates the air quality impacts of stationary and/or mobile sources within horizontal domains of <10 km. This usually requires spatial resolutions of the order of 100 m and computational time steps of the order of five seconds. The HARC model accounts for advection and turbulent diffusion of air pollutants by the local wind field, as well as the chemistry of highly reactive species close to their emission sources. Because the focus of BEE-TEX was on relatively unreactive species such as benzene, atmospheric chemistry was ignored in order to speed up model execution for real-time applications.

The HARC air quality model makes use of a spatially varying wind field extrapolated from either mobile laboratory measurements or wind observations at local monitoring stations. Meteorological extrapolations are performed using the computationally efficient Quick Urban and Industrial Complex wind model[31] based on 3D Lidar measurements of urban morphology to account for the effects of built structures on wind flow at the neighborhood scale. Olaguer et al.[10] provide more detail and a specific example of how meteorological extrapolation was performed during BEE-TEX.

The HARC model can not only function in the traditional forward mode, in which emissions are specified as inputs and the model is run forward in time to generate corresponding ambient pollutant concentrations around sources, but can also be run in adjoint mode to apportion and quantify emissions based on ambient air observations and the 4Dvar method. This inverse modeling technique optimizes model parameters by minimizing a cost function that measures the disagreement between ambient measurements and forward model-predicted concentrations, either at specific locations or averaged over DOAS light paths. The adjoint model propagates an influence function backward in time from the location of measurements to individual sources, favoring those most likely to have contributed to observed peaks due to the prevailing wind. Emissions from sources are then adjusted upward or downward based on their degree of influence on the cost function as well as the uncertainty in their magnitude as measured by an error covariance. Successive forward and adjoint model runs are conducted until the cost function converges. In this way, one can optimize emissions starting from initial guess estimates, which during BEE-TEX were obtained from the USEPA's 2011 National Emissions Inventory. The optimized emissions are then used to reconstruct pollution plumes using the forward model, thus extrapolating the spatially limited ambient concentration measurements throughout the domain of interest.

During BEE-TEX, source attribution and plume reconstruction were performed based on either DOAS CAT scans or mobile laboratory measurements. Real-time modeling analyses were conducted in some cases to yield immediate clues as to which sources were involved in observed emission events. These were typically based on incomplete information and were merely the first of several iterations of data analyses.

Later modeling runs consolidated the initial impressions obtained from real-time observations and analyses.

Summary

The BEE-TEX study was a pioneering experiment in which real-time methods for tomographic remote sensing, community-scale monitoring, data broadcasting, and emission source attribution and quantification were successfully combined for the very first time. In addition, BEE-TEX incorporated the first attempt in an urban setting to use cultured human lung cells in vitro as a gage of human exposure to toxic air pollution. These techniques can provide the basis for new and incisive studies on the impacts of toxic air pollution on human health, especially in industrial fenceline communities as well as the timely and transparent enforcement of environmental regulations governing industrial pollution sources.

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Author Contributions

Eduardo P. Olaguer was the lead scientist who conceived and designed the BEE-TEX experiment. The author reviewed and approved of the final manuscript.

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