Journal of Hazardous Materials 265 (2014) 30–40

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Locating sources of hazardous gas emissions using dual pollution rose plots and open path Fourier transform infrared spectroscopy

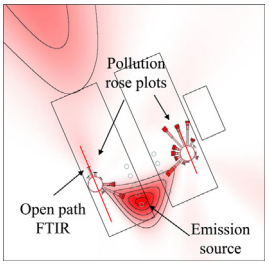
Lung-Yu Sung[a,b], Ruei-Hou Shie[b], Chia-Jung Lu[a,*]

[a] Department of Chemistry, National Taiwan Normal University, Taipei 11677, Taiwan
[b] Industrial Technology Research Institute, Hsin-Chu 30011, Taiwan

HIGHLIGHTS

- Field measurements of gas emission from a semiconductor plant.
- Two open path FTIRs and wind direction data were used to plot pollution roses.
- Locating emission sources via probability-product from two pollution roses.
- Emission sources of multiple compounds were simultaneously identified.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 12 May 2013
Received in revised form
20 September 2013
Accepted 5 November 2013
Available online 13 November 2013

Keywords:
Hazardous gases
Open-path FTIR
Emission source
Pollution rose plot

ABSTRACT

A new approach employing two pollution rose plots to locate the sources of multiple hazardous gas emissions was proposed and tested in an industrial area. The data used for constructing the pollution rose plots were obtained from two side-by-side measurements of open-path Fourier Transform Infrared (OP-FTIR) spectrometers during one week of continuous analysis on the rooftop of a semiconductor plant. Hazardous gases such as CF_4, C_2F_6, CH_3OH, NH_3, NO_2, and SF_6 were found and quantified at the ppb level by both OP-FTIR measurement sites. The data of the top 20% highest concentrations and associated wind directions were used to construct the pollution rose plots. Pollution source probability contours for each compound were constructed using the probability-product of directional probability from two pollution rose plots. Hot spots for SF_6, CF_4, NO_2, and C_2F_6 pointed to the stack area of the plant, but the sources of CH_3OH and NH_3 were found outside of this plant. The influences of parameters for this approach such as the variation in wind direction, lower limit concentration threshold and the nearby buildings were discussed.

1. Introduction

The emissions of hazardous gases from industrial or other sources (i.e., such as landfill or agricultural applications) have profound impact on human health and environment [1,2]. Identifying the emission source is the first step toward the control or reduction of hazardous gases/vapors in the environment. However, identification can be difficult by area analysis because of variations in the dispersion and transportation of pollutants. Over the years, many techniques have been developed to locate airborne pollutant emissions sources by employing industrial layout and weather conditions [3]. These previously developed methods included: emission analyses, trajectory analyses, receptor models, and dispersion models [4]. Various types of area source model for studying the emission of hazardous substances have also been statistically evaluated recently [5].

Conventionally, the concentrations of ambient pollutants are measured by point sampling methods (i.e., Canister with GC–MS analysis). However, in order to locate the emission source, multipoint sampling measurements are usually required. The number of sampling points is often limited due to the cost per analysis; therefore, the spatial concentrations between actual sampling points

* Corresponding author. Tel.: +886 277346132; fax: +886 229324249.
E-mail address: cjlu@ntnu.edu.tw (C.-J. Lu).

0304-3894/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jhazmat.2013.11.006

are usually estimated by interpolation. The most common interpolation approaches include: spatial averaging, nearest neighbor, inverse distance weighting (IDW), and the Kriging method [6,7]. Chen et al. used multiple canisters to collect air samples in a petrochemical plant and were analyzed by gas chromatograph–mass spectrometer. The Kriging method was used in this study to interpolate the concentrations between actual measurement points, and a concentration contour was successfully established to locate the possible emission source [8].

In the past two decades, numerous researches have studied on constructing a concentration contour via a combination of the path-integrated concentration from OP-FTIR and a Computed Tomography (CT) algorithm [9–16]. Similarly, the measurement data from differential optical absorption spectroscopy (DOAS) have also been used in conjunction with CT algorithm [17]. There are two different methods used to reconstruct the spatial distribution of concentrations: the pixel-based method and the model-based method. A pixel-based algebraic reconstruction technique (ART) was an earlier approach for two-dimensional reconstruction [18,19]. Drescher et al. later developed a model-based CT algorithm, which requires the assumption of certain basis functions (e.g., bivariate Gaussian) to describe a pollutant's distribution [20,21]. That method is also known as smooth basis function minimization (SBFM), which shows a significant improvement in prediction.

One limitation to using CT algorithm in the prediction of a concentration profile is time consumption due to the requirement of multiple OP-FTIR light paths and interweaving beam geometry. Therefore, some researchers have developed a method of non-overlapped radial scanning beam geometry using an SBFM algorithm (radial-SBFM) to reduce the number of OP-FTIR measurement paths [22–24]. This approach later evolved into a horizontal radial plume mapping (HRPM) method [25–27]. The HRPM described in the US EPA Other Test Method 10 (OTM-10) was designed to locate the hot spots of contaminants. The SBMF algorithm was also used in one-dimensional concentration profile to find high-concentration areas by fence-line monitoring [28–31].

Most of the source-locating methods mentioned above are based on the assumption of steady-state wind conditions. It is obvious that the distribution of contaminants in an environment will change with wind direction over time. Therefore, some studies have established a "pollution rose plot" based on time-dependent wind direction and concentration data [32–34]. A pollution rose plot can be used to identify the possible direction of an emission source. However, unlike CT or radial-SBFM methods, a pollution rose plot is not able to find the hot spot of contamination. In the present study, therefore, we proposed a method that uses dual OP-FTIR to establish two pollution rose plots. Through triangulation using these two pollution rose plots and probability-product calculations for every spatial point in the area, this method can locate the possible emission sources of various contaminants. The OP-FTIR field data measured on the rooftop of a semiconductor plant was used to test this method. The parameters in the field that may affect the prediction of an emission source are discussed.

2. Methods and experiments

2.1. Measuring setup and site description

The OP-FTIR spectrometers were obtained from ETG (Air Sentry, Baltimore, Maryland, USA) and Bomen (ABB Bomen Inc., St-Laurent, Canada). The instrument was equipped with a mid-band mercury cadmium telluride (MCT) detector that was cooled to 77 K with liquid nitrogen. The infrared light beam was projected through a telescope and aligned to a retro-reflector consisting of gold-plated corner cubes. The infrared spectra of airborne pollutants that passed through the light path between the telescope and the retro-reflector were measured. The sampling rate of continuous analysis was one spectrum per 5 min. The concentration of individual compounds was calculated using classical least square (CLS) fitting with the reference spectra provided by U.S. EPA and Infrared Analysis, Inc. (Anaheim, CA, USA).

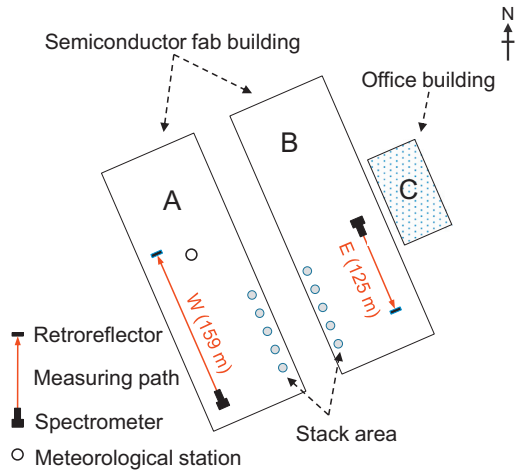


Fig. 1. Layout of buildings and OP-FTIR measurement sites.

Two measurement OP-FTIR light paths were located on the rooftop of a semiconductor plant. There were three buildings in this area, as indicated in Fig. 1. The main processes of this semiconductor plant (e.g., lithography, etching and thin film deposition) were conducted inside Buildings A and B. The light path at the west side (Path W) was on the top of Building A with a length of 159 m. The light path at the east side (Path E) was on the top of building B with a length of 125 m. The height of the buildings is 40 m, the distance between buildings is 15 m, and the distance between sample lines is approximately 250 m. The stacks between Buildings A and B are indicated by the gray circles. To the northeast of the OP-FTIR light path of Building B is the office building for this company. The office building was about 10 m taller than both Buildings A and B.

2.2. Pollution rose plot

A cumulative distribution function (CDF) curve of concentration was used for the scale in the pollution rose. The lower- and upper-limit percentiles were chosen and test at different level and then define the scale of concentration. The software for the CDF, rose plot and probability-product contour were programmed by Microsoft Visual Studio 2010.

2.3. Pollution probability-product and emission source locating

The pollution rose plot provides the probability of observing concentrations for a given direction. The method proposed in the present study employed the triangulation of direction-dependent possibilities from two pollution rose plots. As illustrated in Fig. 2a, the observers P and Q represent the two measurement sites for the OP-FTIR. At any given location in this area, the observation angle is θ_p and the probability of observing a high concentration is $P(\theta_p)$ for observer P. The same location has an observation angle of θ_q and a concentration observing probability of $Q(\theta_q)$ for observer Q. Thus, the product of the concentration observing probability is $P(\theta_p) \times Q(\theta_q)$ for this location. We calculated the probability-product for every point in this area to construct a probability-product contour. The location with the