

The use of real-time monitoring data to evaluate major sources of airborne particulate matter

Stig Hellebust*, Arnaud Allanic, Ian P. O'Connor, John C. Wenger, John R. Sodeau**

Department of Chemistry and Environmental Research Institute, University College Cork, Cork, Ireland

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ABSTRACT

Real-time chemical measurements have been made as part of a field study of air quality in the city and harbour of Cork, Ireland. The data relate to the year 2008, with particular attention paid to the period between May and August. Eight air quality parameters were measured: NO, O₃, NO₂, SO₂, EC, OC, particulate SO₄²⁻ and PM_{2.5}. The data have been used in a novel way involving wind and temporal averaging, along with Principal Component Analysis (PCA) and Positive Matrix Factorisation (PMF) methodologies to extrapolate major source contributions for PM_{2.5}. It is demonstrated that continuous monitoring of standard air quality parameters, such as NO, NO₂, SO₂, along with EC, OC and particulate SO₄²⁻, can be used to provide relevant, cost-effective initial estimates of source contributions to ambient PM_{2.5} levels. It is also shown that the benefit of including OC and particulate SO₄²⁻ in the monitoring protocol is considerable. Three major source groups of ambient PM_{2.5} mass in Cork were identified and quantified using this combined monitoring and modelling approach; road transport (19%), domestic solid fuel burning (14%) and oil-fired domestic and industrial boilers, including power generation plants (31%).

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1. Introduction

A recently published study describes how historical SO₂ pollution data can be used to estimate public exposure to black smoke and particulates (Hodgson et al., 2009). Such quantitative information is now commonly available in most urban centres along with other routine measurements of air quality such as PM_{2.5}, PM₁₀, NO_x, CO and O₃. These measurements are generally made to monitor compliance with European and National Air Quality standards and attract little attention unless threshold exceedances are observed. Such extensive datasets also have, potentially, an important use with regard to the quantification of localised, homogeneous and heterogeneous air pollution sources. The use of standard air quality data in this manner has been little explored, although a small number of studies have been reported both for gaseous pollutants and particulates (Bruno et al., 2001; Guo et al., 2004; Grover and Eatough, 2008).

A meaningful reduction in population exposure to ambient PM can only be achieved cost-effectively if the sources of ambient PM are identified. Identifying source contributions, also known as

source apportionment, often employs multivariate data analysis and matrix factorisation in receptor modelling (Hopke, 1991, 1997, 2003b). Factor analysis strategies attempt to determine simple patterns in the relationships between the measured variables *i.e.* if a group of chemicals or species possess a common origin, they should show similar variation with time. The objective is to discover latent features of the independent variables that are not measured directly. In other words, factor analysis seeks to determine whether observed variables can be explained in terms of a reduced number of variables, called factors. After identifying proper factors, the temporal trend of each can be related to historical events or human activities.

The fundamental relationship between an emission source and receptor (the sampling location) can be expressed as follows (Hopke, 2003b):

$$\mathbf{X} = \mathbf{GF}^T + \mathbf{E} \quad (1)$$

where \mathbf{G} is an $n \times p$ matrix representing source contributions to the samples, \mathbf{F}^T is the transpose of an $m \times p$ matrix of source profiles, \mathbf{E} is the matrix of residuals, n refers to the number of samples, p is the number of variables or species and m is the number of extracted components or source categories. Each sample is an observation in time, and so \mathbf{G} describes the temporal variation of the source contributions. The overall dataset matrix is made up of samples

* Corresponding author. Tel.: +353 21 4903526; fax: +353 21 4903014.

** Corresponding author. Tel.: +353 21 4902680.

E-mail addresses: s.hellebust@ucc.ie (S. Hellebust), j.sodeau@ucc.ie (J.R. Sodeau).

(rows) and species (columns). A viable statistical solution to the expression is sought, the aim of which is to minimise the residuals in E .

A basic and common factorisation technique employed is Principal Component Analysis, or PCA (Nordtvedt et al., 1996; Hopke, 2003a), which attempts to express two or more variables by principal components, which are extracted as solutions to equation (1) above. PCA has been employed in a number of receptor modelling studies (Hosiokangas et al., 1999; Garcia et al., 2006; Viana et al., 2006) in Europe (Harrison et al., 1996, 1997; Fernandez Espinosa et al., 2004; Salvador et al., 2004; Almeida et al., 2005; Pires et al., 2008) and Asia (Fung and Wong, 1995; Harrison et al., 1997; Fang et al., 2006; Ho et al., 2006). The relative contribution of each source can be quantified using absolute principal component scores (Thurston and Spengler, 1985; Artaxo et al., 1999; Vallius et al., 2003, 2005) and the contribution of each component (*i.e.* source) to the total ambient aerosol mass can be obtained by regression against measured aerosol mass.

A more advanced method is termed Positive Matrix Factorisation, or PMF (Hopke, 2003a; Ramadan et al., 2003). It applies constrained maximisation of a weighted object function, related to equation (1) above which takes into account the uncertainties associated with each measurement. Hence the variables are weighted by a measure of trust in the individual measurements and adjusted to their detection limits; “bad” samples can be down-weighted or excluded and the selected solution is based on goodness of fit. Results are also constrained to the non-negative, because the result is not meaningful if the sources turn out to have negative contributions to measured concentrations at the receptor sites. This method has been successfully employed in many studies (Huang et al., 1999; Ramadan et al., 2000; Kim et al., 2003, 2007; Zhao and Hopke, 2004; Chung et al., 2005; Ogulei et al., 2005, 2006; Zabalza et al., 2006; Zhao and Hopke, 2006; Hwang and Hopke, 2007; Pere-Trepat et al., 2007; Shrivastava et al., 2007; Viana et al., 2007), including one carried out in Ireland (Huang et al., 2001). However, it is important to evaluate different receptor models on the same dataset because the strengths of the different techniques combine to provide a more robust solution (Viana et al., 2008).

One issue common to factor analysis techniques is that large datasets of pollutant measurement are required. Obtaining a dataset often involves prolonged collection periods followed by highly labour-intensive laboratory analyses for either organic species, inorganic species (metals and ions), or both. Model output improves for data collected at high temporal resolution, which also impacts on the levels of analytical labour required. For example, a high temporal resolution of the data (*e.g.* half-hourly or hourly records) allows for a resolution of sources of volatile organic compounds and organic aerosol such as local and transported vehicle exhaust emissions, biogenic emissions, photochemical reaction products, which typically exhibit different temporal variation within any given 24 h period, depending on factors such as human activity and sunlight (Jorquera and Rappenglück, 2004; Grover and Eatough, 2008). A temporal resolution in the dataset corresponding to one record for every 24 h period would not allow for separation of emissions with higher frequency variation. Studies therefore have to strike a balance between intensive sampling campaigns (with high temporal resolution of collection that sacrifice seasonal information) and long-term sampling campaigns with, say, daily resolution that can incorporate seasonal information at the expense of temporal resolution. Naturally, if sufficient financial and human resources are available, high temporal resolution can also be obtained over the long term, but these constraints mean that such studies focus on a limited number of well-supported and established research “super-sites” (Huang et al., 2001; Hwang and Hopke, 2007; Saarikoski et al., 2008). Many

other important, but less resourced, locations throughout the world do not have the benefit of such long-term and extensive data bases.

The aim of this work is to show that better, more efficient and predictive use can be made of routinely obtained air quality data, if the right parameters are included in the monitoring network and appropriate source apportionment models are applied to real-time measurements. Two models are presented here, based on PCA and PMF of real-time data records of NO, O₃, NO₂, SO₂, EC, OC, particulate SO₄²⁻ and PM_{2.5} obtained in Cork, Ireland.

2. Methodology

2.1. Sampling site

The sampling site was located at Tivoli Docks in the Port of Cork (51°54'5 N, 8°24'38 W), approximately 3 km east of the city centre. The Port of Cork is part of a large natural harbour, an outline of which is shown in Fig. 1. A berth for liquid bulk ships is located approximately 150 m to the south-west and berths for container ships are situated 400–600 m to the west-south-west (Healy et al., 2009). The prevailing winds are south-westerly.

2.2. Real-time measurements

The sampling station was a customised commercial mobile unit (Securi-Cabin, Ireland) containing a range of real-time monitoring and PM collection equipment. This included a NO_x analyser (model 42i TL), an SO₂ analyser (model 43i), a particulate sulfate analyser (model 5020), an O₃ analyser (model 49i), all from Thermo Inc., USA. A tapered element oscillating microbalance (TEOM, RP 1400a) was employed for continuous measurement of PM_{2.5} mass and a carbon aerosol analysis field instrument (Sunset Laboratory Inc., USA) was used for semi-continuous measurements of Elemental and Organic Carbon (EC/OC). Values for NO_x, SO₂ and O₃ were recorded at a temporal resolution of 1 min. Sulfate was recorded at a temporal resolution of 10 min, PM_{2.5} was recorded at half-hourly intervals and the temporal resolution of EC and OC was one average value for every two-hour period, out of which 12 min were spent analysing particulate matter collected over the previous 108 min, hence the name semi-continuous analyser. The majority of the above instrumentation was deployed on site throughout the whole of 2008 and within

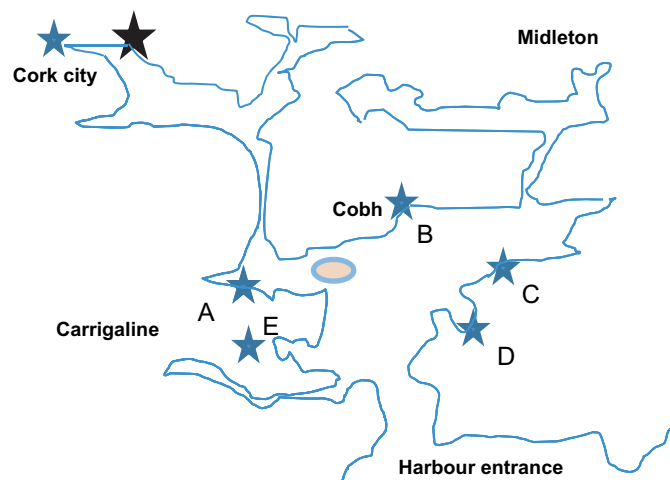


Fig. 1. Outline of Cork harbour. Sampling location marked with black star in figure (top left). The site is located almost due east of the City centre. Other points marked are A) Ringaskiddy deep-water berth operated by Port of Cork, B) Cobh cruise ship terminal, C) power generating station, D) oil refinery and E) pharmaceutical industries.

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