



Source attribution of air pollution by spatial scale separation using high spatial density networks of low cost air quality sensors



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HIGHLIGHTS

- Fast response, high spatial density measurements from a low cost sensor network.
- Technique developed to extract underlying pollution levels from high resolution data.
- Regional and local contributions to total pollution levels are quantified.
- Generally applicable technique for sensor networks (gases and particulates).

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ABSTRACT

To carry out detailed source attribution for air quality assessment it is necessary to distinguish pollutant contributions that arise from local emissions from those attributable to non-local or regional emission sources. Frequently this requires the use of complex models and inversion methods, prior knowledge or assumptions regarding the pollution environment. In this paper we demonstrate how high spatial density and fast response measurements from low-cost sensor networks may facilitate this separation. A purely measurement-based approach to extract underlying pollution levels (baselines) from the measurements is presented exploiting the different relative frequencies of local and background pollution variations. This paper shows that if high spatial and temporal coverage of air quality measurements are available, the different contributions to the total pollution levels, namely the regional signal as well as near and far field local sources, can be quantified. The advantage of using high spatial resolution observations, as can be provided by low-cost sensor networks, lies in the fact that no prior assumptions about pollution levels at individual deployment sites are required. The methodology we present here, utilising measurements of carbon monoxide (CO), has wide applicability, including additional gas phase species and measurements obtained using reference networks. While similar studies have been performed, this is the first study using networks at this density, or using low cost sensor networks.

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1. Introduction

Numerous studies have demonstrated that certain gas-phase pollutants, such as nitrogen oxides (NO_x), ozone (O₃) and carbon monoxide (CO) can be physiologically toxic and may have adverse health effects even at low-level concentrations (e.g. Morris, 2000; Vreman et al., 2000; Wayne, 2000). As such, mitigating urban air pollution has gained considerable importance. Monitoring air quality within urban areas is vital in providing the necessary information to carry out detailed source attribution, and to inform

policy that allows the effective reduction of pollution levels as well as providing more detailed information for epidemiology.

A number of methods exist to carry out source apportionment to investigate the influence of emissions from varying sources that include methods based on emission inventories and dispersion models, and receptor models based on the statistical evaluation of chemical data acquired at measurement sites (Viana et al., 2008). Such methods however may be restricted by the accuracy, availability of emission inventories and pollution source information (Hopke et al., 2006).

The evaluation of monitoring data is a suitable alternative to those methods outlined above with the main advantage being the simplicity of the mathematical methods applied and the reduced effect of mathematical artefacts due to data processing or prior

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assumptions (e.g. Viana et al., 2008).

Of themselves, atmospheric measurements of pollutants provide only the total pollution levels and thus combine contributions from the underlying regional background sources as well as those from local emissions (e.g. Lenschow et al., 2001; Ketzel et al., 2003). In order to undertake more effective source attribution, measurements must be separated into several components, distinguishing between local plume events on short temporal and small spatial scales and underlying trends including the long range transport and variations in natural background pollution sources (Lenschow et al., 2001).

Within urban areas, a number of additional emission sources (such as vehicle exhaust and background heating) exist that contribute to total pollution levels when compared to the wider sub-urban and rural environments. These local source emissions can accumulate and thus increase pollution over longer time scales. Quantifying these contributions to the total pollution levels is useful for pollution mitigation.

A number of air quality monitoring networks exist that provide atmospheric measurements of key pollutants such as NO_x , O_3 , CO and particulate matter (PM). The Automatic Urban and Rural Network (AURN) is one such network and is deployed in the UK in order to comply with national and European air quality regulations (Defra, 2014). However, its temporal (1 h) and spatial (~100 sites in the entire UK) resolution is far too sparse to allow the contributions of different sources to total pollution levels across the UK to be quantified without significant additional constraints and assumptions such as the use of physical or statistical models. As outlined in Ketzel et al. (2003) measurements at both polluted and unpolluted sites in close proximity are necessary to effectively estimate the amounts that local plume events, emission build-up and background levels contribute to the overall pollution levels. This is particularly important in terms of advanced source attribution for relatively short-lived species that are chemically converted within close proximity (several hundred metres) of their sources (e.g. NO_x with a lifetime in the order of one day (Seinfeld and Pandis, 1998)). Low-cost electrochemical air quality sensors can be deployed in denser networks, potentially alleviating this problem (see for example Mead et al., 2013).

In this paper, we combine high temporal and spatial resolution data generated by a low-cost high density air quality network with a novel approach to data analysis to illustrate how, combined, source attribution can be achieved. The focus of this paper is CO, a moderately long lived chemical species with a lifetime in the order of weeks to months (Holloway et al., 2000; Zellweger et al., 2009). CO is thus subject to longer range transport and may be used as tracer molecule to investigate the influence of larger-scale meteorological events on tropospheric pollution. Because of its long lifetime, CO generally is a useful indicator of local pollutant emissions. CO is the main (70%) loss mechanism for the hydroxyl radical, OH, (Novelli et al., 1998) with increased CO levels enhancing the rate of OH removal, subsequently reducing the scavenging mechanism for other pollutants as well as augmenting tropospheric ozone production. Knowledge of CO emission sources may therefore be indirectly important in terms of reducing adverse health effects related to atmospheric pollutants. It also contributes to carbon emissions making it important in terms of climate change mitigation.

In the present work we will show that we are able to define a regional CO signal through a purely data-based approach (section 4.1), subsequently allowing detailed source attribution to be carried out based on the measurements alone. This new technique is used to separate the different contributing scales of air pollution namely regional, far field and near field (section 4.2). The methods presented may be applied to other pollutant species when differences

in abundance due to their chemistry and lifetimes are considered.

2. The Cambridge air quality monitoring network

In spring 2010, a network of 45 low-cost electrochemical sensor nodes was deployed in and around the city of Cambridge, UK during a period covering 2.5 months (11 March 2010 to 30 May 2010). The network provided measurements of CO, nitric oxide (NO) and nitrogen dioxide (NO_2) as well as temperature and relative humidity at a high temporal (10 s) resolution. To account for stabilisation of the electrochemical cells within the surrounding ambient conditions, the initial 14 days of the full measurement campaign were excluded from further analysis. Thus, the study covers an eight week period from 28 March 2010 to 23 May 2010. Of the 45 sensor nodes deployed, 9 were discarded as a result of reduced data coverage (<1 month) due to battery issues or physical damage. An additional 4 nodes had technical problems or were clearly biased by external factors (Alphasense, 2005) thus 32 (71%) of the sensor nodes deployed were included in this study.

2.1. Accuracy of the electrochemical sensors to ambient concentrations

Mead et al. (2013) have reported on characterisation of electrochemical sensors, determining an instrumental detection limit (IDL) of 4 ppb (parts-per-billion) for CO and their sensitivity to ambient pollution levels. The electrochemical sensors' long-term stability allowed observed differences in the measured absolute mixing ratios for individual sensors to be corrected for during operation and data processing.

We reference the data of the sensor network to gas-chromatographic measurements, averaged to a 30 min resolution and calibrated daily against several NOAA standards. These data were obtained from the Greenhouse Gas Laboratory at Royal Holloway, University of London (RHUL), 75 miles south-west of Cambridge in Egham, Surrey. Every sensor node is referenced to this one station. To remove local influences on the measurements, a meteorological filter is applied. The individual sensor offset to ambient pollution levels is therefore defined as the difference between the node's minimum CO concentration during those nights (01:00–04:00, all times in BST) with a wind speed (U) greater than 2 ms^{-1} and the corresponding value of the RHUL data set.

2.2. Deployment details of the electrochemical sensor network

The sensor nodes were mounted on lamp posts, 3 m above street level. A higher density of sensor nodes were deployed within the urban environment (Fig. 1) where higher variability of pollution levels were expected compared to rural areas. Sensor nodes were approximately evenly distributed in the sub-urban area of Cambridge in order to investigate the influence of varying wind direction on atmospheric pollution and to inter-compare rural environments.

The current of each electrochemical sensor was measured every 10 s, and then converted to counts (via a resistor to generate a voltage and an Analogue-To-Digital-Converter) and stored on-board the sensor. The collected (10 s time resolution) raw data were then transmitted in packets to a central computer server at two hour intervals, in order to reduce power consumption by the GPRS in each node.

The transmission process induces electrical interference on the sensor signals; thus, the initial 65 recordings, that is, ~11 min of data after each transmission were filtered out prior to further analysis. The data were then converted into mixing ratios using pre-defined, sensor-specific sensitivity factors.

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