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Using a source–receptor approach to characterise VOC behaviour in a French urban area influenced by industrial emissions Part I: Study area description, data set acquisition and qualitative data analysis of the data set

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ABSTRACT

The global objective of this two part study was (1) to conduct VOC measurements in order to further understand VOC behaviour in an urban area influenced by industrial emissions and (2) to evaluate the role of these specific sources relative to urban sources. In this first paper a thorough descriptive and qualitative analysis is performed. A second article will be devoted to the quantitative analysis using Chemical Mass Balance (CMB) modelling. In the Dunkerque (France) area most industrial sources are situated in the north and the west of the receptor site whereas urban and traffic sources are located in the south and the east. A data set constituted of nearly 330,000 VOC data has been developed from the hourly measurements of 53 VOCs for 1 year from September 2002 to August 2003. It also contains

meteorological parameters such as temperature, wind direction and wind speed. Using different graphical methods, the influence of the different sources on the ambient VOC concentrations has been highlighted at different time scales. In this work, the analysis of daily time series for the 53 VOCs shows the influence of traffic exhaust emissions because of the increases at traffic rush hours. Besides, the seasonal evolution of the VOC/acetylene ratio points out the influence of evaporative sources on ambient VOC concentration. Concerning other point sources, the variations of measured VOC concentrations for different wind directions and scatter plots of VOC hourly concentrations highlight the influence of some industrial sources.

1. Introduction

Various human activities emit large amounts of anthropogenic hydrocarbons in the atmosphere (traffic via vehicle exhaust and fuel evaporation, industries, energy production, solvent use...). Most ambient hydrocarbon species are strongly involved in the formation of photochemical oxidants and elevated surface ozone levels in urban areas (Seinfeld and Pandis, 1997). Some of them are toxic or carcinogenic for humans and animals above certain doses (Ashford and Miller, 1998).

A large number of studies or monitoring programs have been devoted to VOC measurements in urban areas (Singh et al., 1992; Hansen and Palmgren, 1996; Moschonas and Glavas, 1996; Cheng et al., 1997; Brocco et al., 1997; Derwent et al., 2000; Liu et al., 2000; Na and Kim, 2001; Colon et al., 2001; Barletta et al., 2002; Lai et al., 2004; Barletta et al., 2005; Latella et al., 2005) and thus concentration levels, behaviours and sources of VOCs in this type of site are now well-known. Indeed, a lot of studies have been conducted to show the importance of traffic emissions on VOC ambient concentrations and all have demonstrated the increase

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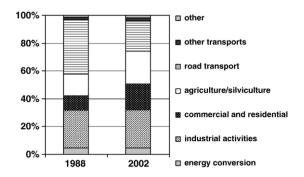


Fig. 1-Evolution of VOC emissions between 1998 and 2002 in France.

of VOC concentrations with increased traffic flow densities in urban areas (Kourtidis et al., 1999; Muezzinoglu et al., 2001; Na et al., 2002; Lai et al., 2004). Some other studies have shown that both quantity and nature of VOCs emitted into the atmosphere are related to the fuel types used, vehicle types and ages, traffic flow rates and speeds as well as roads and environmental conditions in the city (Fontaine, 2000; Fontaine and Galloo, 2002; Paul, 1997). Considering this information, effective methods to reduce atmospheric VOC levels have been developed. In France, recent important efforts have been made to reduce the amounts of VOCs emitted by traffic. Fig. 1 illustrates VOC emissions in France between 1998 and 2002 depending on source types. On the one hand, for 4 years, the total quantity of VOCs decreased by more than 40%. On the other hand, the decrease of traffic emissions implied the increase of the relative part of VOCs emitted by industrial activities. So industries now constitute the first emitter of VOCs in France reaching 27%. Considering this fact, it seems particularly important to evaluate the role of specific sources like industrial areas.

Some recent studies deal with industrial source influence on VOC levels measured in urban areas close to industrial activities and show the importance of industrial emissions for some VOCs (Na et al., 2001; Jobson et al., 2004; Lai et al., 2005).

Because their processes are associated with the emission of a large number of VOCs into the atmosphere, the study areas are very often petrochemical complexes with petroleum refineries and petrochemical plants which are generally large industrial installations (Cetin et al., 2003; Lin et al., 2004). Most of these studies were performed punctually with canister campaigns and only the study of Cetin et al. (2003) permits to appreciate seasonal variations of concentrations.

The chosen area for our study, located around the city of Dunkerque (northern part of France), gathers about 225 industrial plants emitting more than 4000 tons/year of VOCs. 75% of these emissions are distributed between only 6 major emitters. In this study, hourly concentrations of 53 different VOCs (mainly from C_2 to C_9) have been measured from September 2002 to August 2003, providing a year of measurements. Considering the long observation duration, seasonal variations of source contributions can be evaluated and considering the hourly frequency of measurements daily variations of these contributions can be investigated as well.

The global objective of this study is to conduct VOC measurements in order to further understand VOC behaviour in an urban area influenced by industrial emissions and to evaluate the role of specific sources (industrial plants) relative to urban sources. Several steps have been necessary to reach this objective: (1) the measured VOC concentrations in this urban site close to a large industrial area have been compared to the values measured in other urban and suburban sites both in France and around the world; (2) temporal and spatial behaviour of VOC concentrations has been investigated to identify precisely the sources playing an important role in ambient levels; (3) industrial source profiles have been established to provide the CMB model with input data; (4) source contribution assessment and the relative importance of industrial activities have been evaluated using the CMB model. Steps 3 and 4 will be described in Part II of the study (Badol et al., 2007).

Part I of this work deals with the description of the study area and the data set comprising hourly VOC concentration

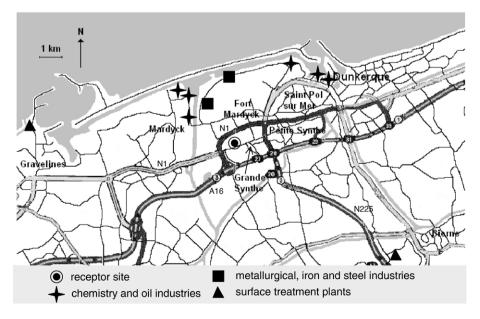


Fig. 2-Receptor site and source locations.

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