Available online at www.sciencedirect.com
SCIENCE DIRECT®
Atmospheric Environment 39 (2005) 2421–2431
ATMOSPHERIC ENVIRONMENT
www.elsevier.com/locate/atmosenv

Fine particulate (PM2.5–PM1) at urban sites with different traffic exposure

Michele Giugliano, Giovanni Lonati*, Paola Butelli, Laura Romele, Ruggero Tardivo, Mario Grosso

Dipartimento di Ingegneria Idraulica, Ambientale, Infrastrutture viarie, Rilevamento, Sezione Ambientale, Politecnico di Milano, P.zza L. da Vinci, 32–20133 Milano, Italy

Received 19 January 2004; received in revised form 8 April 2004; accepted 9 June 2004

Abstract

Fine particulate concentration data resulting from several monitoring campaigns performed in the city of Milan at urban sites with different exposure to the emission sources are presented. Low volume PM2.5 and PM1 samplers are utilised together with a low volume optical analyser, enabling the intercomparison between the measurements obtained by the gravimetrical and the optical method. The concentration levels observed at the different sites are compared in order to point out intra-site seasonal differences and inter-site differences for corresponding seasons of the year. These different concentration levels are analysed and explained considering the exposure to the primary emissions and accounting for the role of meteorology. PM10, PM2.5 and PM1 are described in terms of the distribution of 1-h concentration data and their relative mass fractions are determined. In order to assess the significance of secondary sources of fine particulate, a PM2.5 high volume sampler is utilised for the collection of dust-loaded filters to be analysed for chemical characterisation. The composition of PM2.5 emission from traffic is investigated by analysing 24-h samples from an urban tunnel site (TU): data on carbonaceous species, organic carbon (OC) and elemental carbon (EC), are obtained and the ratio between these species is evaluated for real traffic emissions. Secondary organic aerosol (SOA) contribution to PM2.5 mass in ambient air is assessed by means of the primary OC/EC ratio approach, based on chemical data of the filters from the TU. Organic and inorganic secondary production in the outdoor atmosphere is contributing for about 75% of PM2.5 mass in winter and 40% in summer: as a consequence, effective long-term actions, still controlling the emissions of primary pollutants too, are required for air quality standards attainment and the potentiality of short-term interventions, as traffic restriction, appears quite limited.

Keywords: Gravimetric measurements; Optical measurements; Chemical characterisation; Urban area; Real traffic; OC/EC method; Secondary organic aerosol

1. Introduction

Atmospheric pollution due to airborne fine particles i s an environmental issue of increasing concern in Lombardy, Italy's most industrialised region, and in particular in the metropolitan area of Milan. With

*Corresponding author. Tel.: +39 02 23 99 64 30; fax: +39 02 23 99 64 99.
E-mail address: giovanni.lonati@polimi.it (G. Lonati).

1352-2310/$ - see front matter © 2005 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2004.06.050

almost 3 millions inhabitants and about 1.5 million passenger cars and utility vehicles circulating daily, the area is representative of the largest Central and Western European cities.

The long-term ($40\,\mu g\,m^{-3}$ as annual average concentration) and short-term (no more than 35 exceedances per year of $50\,\mu g\,m^{-3}$ for the daily average) standards for PM10 established by the European Union Directive (EU, 1999) are both largely not attained: out of the 40 stations of the regional air quality monitoring network operating in Lombardy during 2001, the long-term standard was not respected at 19 sites and the short-term standard at 37 sites. Warning episodes, as defined by the regional authorities (daily average concentration simultaneously exceeding $50\,\mu g\,m^{-3}$ at 2 or more monitoring sites of the area for seven consecutive days), frequently occur during the winter period. Moreover, the processes of secondary photochemical production of fine particles seem to play a significant role even during summertime, when peak concentration events are observed too. The apportionment of the contribution of the stationary sources, mainly domestic heating, and of the mobile sources has a very important role for the definition of the interventions to be taken when the warning episodes occur. These latter almost exclusively consist in the restriction of the circulation for some categories of vehicles during weekdays and eventually in the prohibition of the circulation extended to all vehicles on Sundays. For long-term control strategies implementation, however, information from emission inventory and source apportionment have to be completed by the assessment of the role of secondary formation in the atmosphere from primary precursors.

In order to assess the contribution of traffic to the observed concentration levels of fine particulate, several monitoring campaigns have been performed at 4 urban sites characterised by different exposure to the traffic source: an urban background site, 2 kerbside sites and a tunnel site (TU), all located in the city of Milan. In the TU it is possible to isolate the actual role of the traffic sources under typical driving conditions (Swietlicki et al., 1999). A PM2.5 and a PM1 low-volume sampler, together with a low-volume optical analyser, have been simultaneously utilised during all the campaigns, which took place in winter, spring, autumn and in the summer holiday period in August. Particulate matter samples obtained by the PM2.5 and PM1 samplers have been analysed for mass concentrations, allowing the intercomparison between the gravimetrical measurements and those performed by the optical analyser. The observed intra-site seasonal differences in concentration levels, as well as the inter-site differences for corresponding seasons of the year, have been explained considering the exposure to the primary emissions and accounting for the role of meteorology. The mass contribution of both PM2.5 and PM1 to PM10 concentration levels are evaluated and the distributions of PM10, PM2.5 and PM1 concentration data are analysed.

Sampling campaigns for the production of PM2.5-loaded filters, to be speciated for the major chemical components, have been performed at the urban background site and the TU site by means of a high-volume sampler. At both sites, PM2.5 mass composition is analysed in terms of the ionic components (Cl^-, NO_3^-, SO_4^{2-}, NH_4^+) and of the carbon species, elemental carbon (EC) and organic carbon (OC). Data from the TU, where the secondary production is significantly limited, give a useful information on primary contribution of ionic compounds and carbon species to PM2.5 mass for traffic emissions. For the carbon species in particular, the chemical characterisation suggests a reliable estimate of the average primary OC/EC, representative of the real traffic emissions in the area. Based on the analytical results at the TU, the primary OC/EC ratio approach is applied to assess the contribution of secondary organic aerosol (SOA) to the PM2.5 mass at the urban background site, where the higher values observed for the OC/EC ratio indicate a relevant role for the secondary source.

2. Methods

2.1. Monitoring sites

The monitoring campaigns have been performed between April 2001 and March 2003 at 4 sites in the city of Milan characterised by a different exposure to the mobile sources. A short description of each site and of its exposure to the traffic emissions, the duration and the period when the campaigns took place are reported in Table 1.

At the 2 kerbside sites (KR and KS) the instruments were located immediately beside the pavement, while at the TU they were located in the traffic divider half-way the 250 m long tunnel. At both ends of the tunnel there are traffic lights that frequently cause queues of idling vehicles.

2.2. Instruments

2.2.1. Gravimetric samplers

The sampling of PM2.5 has been carried out by means of both a low-volume automatic sequential sampler (Manufacturer: TCR Tecora) and a high-volume one (Manufacturer: DIGITEL).

The low-volume sampler ($1\,m^3\,h^{-1}$ flow rate) is equipped with PM2.5 cut-off inlet and 47 mm PTFE filter; the cartridge of filters are manually changed every 2 days without interruption of sampling. The weighing operation follows an internal procedure based on blank filters pre-conditioning at controlled relative humidity