Science of the Total Environment 450–451 (2013) 22–30

Contents lists available at SciVerse ScienceDirect

Science of the Total Environment

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

A methodology for elemental and organic carbon emission inventory and results for Lombardy region, Italy

Stefano Caserini [a], Silvia Galante [a,*], Senem Ozgen [a], Sara Cucco [a], Katia de Gregorio [a], Marco Moretti [b]

[a] Politecnico di Milano, DICA Environmental Engineering Section, Piazza Leonardo da Vinci 32, 20133 Milano, Italy
[b] Environmental Protection Agency of Lombardia Region, ARPA, 20124 Milano, Italy

HIGHLIGHTS

► Diesel and wood combustion contribute to more than 80% of total EC and OC.
► More than 50% of EC emissions come from road transport.
► Monte Carlo method is used to assess the uncertainty of wood combustion emissions.
► Residential wood combustion is the main source of uncertainty of EC OC inventory.
► In terms of CO_2eq, EC and OC correspond to 3% of CO_2 emission in Lombardy.

ARTICLE INFO

Article history:
Received 27 July 2012
Received in revised form 14 December 2012
Accepted 24 January 2013
Available online 1 March 2013

Keywords:
Emission inventory
Organic carbon
Elemental carbon
Black carbon
Diesel
Wood combustion

ABSTRACT

This paper presents a methodology and its application for the compilation of elemental carbon (EC) and organic carbon (OC) emission inventories. The methodology consists of the estimation of EC and OC emissions from available total suspended particulate matter (TSP) emission inventory data using EC and OC abundances in TSP derived from an extensive literature review, by taking into account the local technological context. In particular, the method is applied to the 2008 emissions of Lombardy region, Italy, considering 148 different activities and 30 types of fuels, typical of Western Europe. The abundances estimated in this study may provide a useful basis to assess the emissions also in other emission contexts with similar prevailing sources and technologies.

The dominant sources of EC and OC in Lombardy are diesel vehicles for EC and the residential wood combustion (RWC) for OC which together account for about 83% of the total emissions of both pollutants. The EC and OC emissions from industrial processes and other fuel (e.g., gasoline, kerosene and LPG) combustion are significantly lower, while non-combustion sources give an almost negligible contribution. Total EC + OC contribution to regional greenhouse gas emissions is positive for every sector assuming whichever GWP100 value within the range proposed in literature. An uncertainty assessment is performed through a Monte Carlo simulation for RWC, showing a large uncertainty range (280% of the mean value for EC and 70% for OC), whereas for road transport a qualitative analysis identified a narrower range of uncertainty.

1. Introduction

There is an increasing interest in black carbon (BC) and organic carbon (OC) emissions, due not only to their health effects typical of fine particles, but also to their impact on climate, given their potential to alter the Earth's energy balance through a complex net of processes (Ramanathan and Carmichael, 2008). Reducing BC emissions is thought to be a potential powerful tool to combat global warming (Chung et al., 2012; UNEP-WMO, 2011).

Black carbon in soot is the dominant absorber of visible solar radiation in the atmosphere and is estimated to be the second largest contributor to global warming after carbon dioxide (Ramanathan and Carmichael, 2008). There is a range of quantitative estimates in the literature for global average radiative forcing due to BC. According to a recent report by the United Nations Environment Programme and World Meteorological Organization, global average net radiative forcing for BC is likely to be positive with a best estimate of 0.6 W m^{-2} (UNEP-WMO, 2011). The same assessment reports for OC particles, which generally tend to produce a cooling influence on climate, central values in the range of -0.08 to -0.30 W m^{-2}, with a best estimate of -0.19 W m^{-2}.

The terms black carbon and elemental carbon are frequently used to specify non-organic, non-carbonate part of the carbonaceous aerosol fraction; the term BC is usually employed when pure optical methods are used in the quantification. The term elemental carbon

* Corresponding author.
E-mail address: silvia1.galante@polimi.it (S. Galante).

0048-9697/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.scitotenv.2013.01.073

(EC) is frequently employed to refer to refractory carbon determined with physical and chemical analyses (e.g. thermo-optical methods). Thus BC and EC are correlated but not coincident. BC inventories nearly always use EC from particulate matter (PM) source emission samples (Chow et al., 2010); for this reason the following analyses use the term EC in reference to the emission values.

The present paper assesses the elemental and organic carbon emissions for the year 2008 in Lombardy, a 9 million-inhabitant-region in northern Italy, through an extensive literature review of EC and OC abundances (%) in particulate matter emissions. The study pays particular attention to the main sources of EC and OC in Lombardy, namely residential wood combustion (RWC) and on-road diesel vehicle emissions; it also considers minor sources such as gasoline vehicles, off-road vehicles, fires, and stationary combustion (i.e., coal, fuel oil, natural gas).

2. Methodology

Following the approach of Kupiainen and Klimont (2007) and Winther and Nielsen (2011), EC and OC emissions are estimated by the product of particulate emissions and the related EC and OC abundances in PM for each activity sector and fuel, according to Eq. (1):

$$E_{y,i,j} = p_{y,k,i,j} \cdot E_{k,i,j} \quad (1)$$

where:

- y,i,j,k chemical species, sector, fuel, PM size fraction;
- $E_{y,i,j}$ emissions for the chemical species y for sector i and fuel j;
- $E_{k,i,j}$ emissions for the PM size fraction k for sector i and fuel j;
- $p_{y,k,i,j}$ abundance (%) of chemical species y in PM size fraction k for sector i and fuel j.

This methodology allows taking advantage of the available information on PM emission inventories developed in the last years in many European regions. The main drawback concerns sources in which PM emission is governed by the inorganic content of the fuel, and therefore a correlation with BC and OC, whose formation depends on the parameters of the combustion process, may not exist (Streets et al., 2001). However, as will be shown in Section 3, such activities (e.g., coal combustion, use of diesel oil with high sulfur content) are very limited in Lombardy.

The values for particulate emissions are taken from the 2008 regional emission inventory INEMAR of Lombardy region (ARPA Lombardia, 2011). Three size fractions are considered, namely total suspended particulate matter (TSP), particles with aerodynamic diameters less than 10 μm (PM10) and those with aerodynamic diameters less than 2.5 μm (PM2.5). The INEMAR emission inventory is compiled according to EMEP/EEA Atmospheric Emission Inventory Guidebook (AEIG) and using specific regional data (Caserini et al., 2012; EEA, 2010); as an example, detailed traffic data and COPERT IV (Ntziachristos et al., 2009) algorithms are used for road transport emissions, whereas results from a detailed survey on biomass consumption in domestic appliances and AEIG emission factor are used for RWC (Pastorello et al., 2011).

EC and OC abundances came from a detailed analysis of available scientific literature, as described in Section 3. In order to make the comparison possible, the abundances found in the literature data are expressed in terms of the same PM size fraction, namely TSP. The particulate size fractions (i.e., PM2.5/TSP, PM10/TSP) are taken from the INEMAR inventory.

In order to have a first level assessment of the climate impact of the carbonaceous aerosols considered in this paper, CO_2eq emissions from EC and OC sources are calculated considering the average, minimum and maximum global warming potential values for a time horizon of 100 years (GWP100) suggested in the UNEP-WMO report (UNEP-WMO, 2011): 680, 210, and 1500 for BC (or EC), and −69, −129, and −25 for OC.

3. EC and OC abundances from various emission sources

An extensive literature review is performed, to evaluate EC and OC share for all the PM sources considered by the regional inventory; the attention is particularly focused on the most important emission sources, as discussed in the following paragraphs.

The plain area of the Lombardy region is characterized by the combination of unfavorable atmospheric dispersion conditions with a high population density and intensity of economic activities, which makes it one of the most polluted areas in Western Europe. Therefore in the last decades many regulations have tried to limit emissions: for instance, coal is now of very limited use, the use of coal and fuel oil in the residential sector is banned, methane is largely the dominant fuel in the power sector.

In 2008, more than 70% of PM10 and PM2.5 emissions are derived from two sources, namely on road diesel vehicles and RWC, that are also known from literature to have a high OC and EC share in PM (Bond et al., 2004; Kupiainen and Klimont, 2007). The attention of this work is particularly focused on these sources, in order to identify how local information could improve the reliability of the EC and OC inventory.

3.1. Diesel vehicles — on-road transport

Diesel exhaust is one of the key emission sources for gaseous pollutants as well as particulate matter. Particulate emissions from diesel engines consist of soot particles surrounded by condensed organic compounds and trace metals from lubricating oil and fuel additives (Kittelson, 1998), with the composition of the particles being predominantly EC and OC (Sharma et al., 2005). Nanoparticles (diameter<0.05 μm) constitute the 90% of the number emissions. Most of the particle mass, on the other hand, exists in the accumulation mode (i.e., 0.1–0.3 μm) where the carbonaceous agglomerates and associated adsorbed materials reside (Kittelson, 1998).

EC and OC emissions are different from engine to engine and are largely dependent on the driving mode, engine operating history and maintenance (Shah et al., 2004). OC fraction increases during modes with low vehicle speed and in particular at idling conditions (Cocker et al., 2004; Sharma et al., 2005) and EC fraction increases at higher speeds and higher engine load (Cocker et al., 2004; Lim et al., 2008). Engine age influences the EC/OC ratios with values much lower than unity reflecting high OC emissions of the older engines (Shah et al., 2004). Diesel particulate filter (DPF) reduces significantly the EC fraction but does not have much effect on the OC fraction because of the condensable fraction in the gas phase (Geller et al., 2006) which is effectively reduced by catalytic devices (Biswas et al., 2009).

The available data doesn't allow the definition of the detailed variation of EC and OC emissions for different driving modes or vehicle maintenance conditions; therefore, in the following, emission factors are divided into categories in function of vehicle category only.

Fig. 1 (Table SM1 in the Supplementary Material) lists the EC and OC abundances reported in various studies in literature. Where needed, the vehicles are classified in Euro emission categories based on the production year of the investigated vehicles. Literature values are divided in two main groups, namely light duty vehicles (LDV, comprehensive of passenger cars) and heavy duty vehicles (HDV). Vehicles equipped with exhaust after-treatment units (i.e., diesel particulate filter — DPF) are assessed separately. Average values are considered for the determination of the EC and OC abundances to be used in this study. As proposed by COPERT IV methodology (EEA, 2010), particles with diameters less than 2.5 μm are assumed to be the totality of TSP. Where it is necessary, an OM/OC ratio of 1.3 is assumed to convert into organic carbon the organic material (OM)