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A novel approach to compare simultaneous size-segregated particulate matter (PM) concentration ratios by means of a dedicated triangular diagram using the Agri Valley PM measurements as an example

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This work presents a novel approach to compare and graphically represent simultaneous concentration measurements of PM_{10} , $PM_{2.5}$ and PM_1 (i.e., aerosol particles with aerodynamic diameters less than 10, 2.5 and 1 μm , respectively) with similar data reported in literature using $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios. To this aim, a dedicated triangular diagram was used. The proposed approach was applied to size-segregated PM concentrations recorded in Agri Valley (Basilicata Region – southern Italy). Results shows that the PM_{10} , $PM_{2.5}$ and PM_1 concentrations recorded in the Agri Valley are comparable both in terms of PM concentration ratios and PM levels to an urban site.

1 Introduction

The growing interest aroused by aerosol particles (also referred to as particulate matter, PM) is due to their impact on human health, air quality and global climate change (Colette et al., 2008; IPCC, 2007; WHO, 2006). These particles can differ in size, shape and chemical composition and can be generated both from natural and/or anthropic sources. The particle size fraction is an important physical parameter since it can provide information relating to the particle origin, their formation process and effects on human health. In fact, the PM coarse fraction (i.e., PM_{10} , aerosol particles with aerodynamic diameters less than 10 μm) mainly originates from natural sources such as re-suspension of local soil by winds, desert dust, forest fire, volcanic eruptions (Alekandropoulou and Lazaridis, 2013; Campos-Ramos et al., 2011) as well as anthropogenic sources such as re-suspension of road dust, material grinding and crushing (Colbeck, 2008; Van Dingenen et al., 2004). Regarding fine and ultrafine fractions (i.e., $PM_{2.5}$ and PM_1 , aerosol particles with aerodynamic diameters less than 2.5 and 1 μm , respectively), they mainly originate from anthropic sources such as industrial activities, traffic, domestic heating (Caggiano et al., 2010; Lin and Lee, 2004). According to

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the particle size, PM can pose risks to human health because of its adverse effects both on the respiratory and cardiovascular systems (Pope and Dockery, 2006). In fact, coarse particles are likely to be deposited in the extra-thoracic and upper bronchial region. Instead, fine and ultrafine particles may travel deeply into the lungs and may be deposited in the lower bronchial and alveolar regions.

In the light of this, several efforts have been made in order to obtain simultaneous concentration measurements of different PM fractions (Massey et al., 2012; Cabada et al., 2004) and their ratios have been used to obtain preliminary indication about the sources contributing to the presence of the PM in the local atmosphere (Pérez et al., 2010; Cheng et al., 2006; Vecchi et al., 2004) and/or to compare the PM levels among different sites (Shahsavani et al., 2012; Gomišček et al., 2004; Li and Lin, 2002; Claiborn et al., 2000).

This work presents a novel approach to compare and graphically represent simultaneous measurements of PM_{10} , $PM_{2.5}$ and PM_1 concentrations with similar data reported in literature using both $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios. To this aim, a dedicated triangular diagram was used. The proposed approach was applied using PM concentration measurements (Trippetta et al., 2013) recorded in Agri Valley (Basilicata Region – southern Italy). This area was chosen since it is an area of great environmental concern. In fact, the Agri Valley houses the largest European on-shore reservoir (crude oil and gas) and the biggest oil/gas pre-treatment plant (identified as Centro Olio Val d'Agri – COVA) in a rural/anthropized context (Fig. 1). The COVA plant implies continuous gaseous and particulate atmospheric emissions that could give rise to a wide range of environmental and human health impacts due to the large presence of cultivated and grazing areas and of several small towns (from 1700 to 5400 inhabitants) in its surroundings.

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2 Methodology

The triangular diagram, based on Sneed and Folk's original idea (Sneed and Folk, 1958) and generally applied in pebbles and fabric shapes (Benn, 1994; Illenberger, 1991), has been opportunely arranged both to compare and graphically represent simultaneous PM_{10} , $PM_{2.5}$ and PM_1 concentration ratios measured in a reference site with similar measurements reported in literature.

This approach is based on the calculation of the ratios between $PM_{2.5}$ and PM_{10} concentrations (i.e., $PM_{2.5}/PM_{10}$ that is the fine fraction contribution to the PM_{10}) and between PM_1 and PM_{10} concentrations (i.e., PM_1/PM_{10} that is the ultrafine fraction contribution to the PM_{10}) and their representation by means of a dedicated software (Graham and Midgley, 2000).

The peculiar features of this diagram also allows the representation of $PM_1/PM_{2.5}$ and the $(PM_{2.5} - PM_1)/(PM_{10} - PM_1)$ concentration ratios. In particular, the former identifies the contribution of the ultrafine fraction to the fine fraction. The latter represents the proportion between the intermodal and the coarse fractions.

Each site will be identified on the diagram with a point representing the corresponding size-segregated PM concentration ratios. Two data points on the triangular diagram (e.g. P_a and P_b) can be considered closed if they have got $PM_{1a}/PM_{10a} \approx PM_{1b}/PM_{10b}$ as well as $PM_{2.5a}/PM_{10a} \approx PM_{2.5b}/PM_{10b}$ within an acceptable tolerances. As consequence, it is also valid that $(PM_{2.5a} - PM_{1a})/(PM_{10a} - PM_{1a}) \approx (PM_{2.5b} - PM_{1b})/(PM_{10b} - PM_{1b})$ and that $PM_{1a}/PM_{2.5a} \approx PM_{1b}/PM_{2.5b}$. Therefore, to compare simultaneous sampling of PM on the triangular diagram enables of comparing the ratios among the mass concentrations of PM (i.e. PM_1/PM_{10} , $PM_{2.5}/PM_{10}$, $(PM_{2.5} - PM_1)/(PM_{10} - PM_1)$ and $PM_1/PM_{2.5}$) altogether.

A site is considered similar to the reference one in terms of PM ratios if the corresponding $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios differ no more than $\pm 5\%$. It is important to observe that, for comparison purposes, close values of PM_1/PM_{10} , $PM_{2.5}/PM_{10}$, $PM_1/PM_{2.5}$ and $PM_{2.5} - PM_1/PM_{10} - PM_1$ do not necessarily mean close

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values of PM_1 , $PM_{2.5}$ and PM_{10} concentration. Indeed, if two close points (e.g. P_a and P_b) displayed on the triangular diagrams are considered, they have got similar values of the respective ratios $PM_{1a}/PM_{10a} \approx PM_{1b}/PM_{10b}$ and $PM_{2.5a}/PM_{10a} \approx PM_{2.5b}/PM_{10b}$, etc. Nevertheless, the concentration value of PM_{1a} can be different from that of PM_{1b} as well as the concentration value of $PM_{2.5a}$ can be different from that of $PM_{2.5b}$. Hence, only if close points have got PM_{10a} concentration $\approx PM_{10b}$ concentration then it is PM_{1a} concentration $\approx PM_{1b}$ concentration and $PM_{2.5a}$ concentration $\approx PM_{2.5b}$ concentration.

In order to refine the comparison and identify all the sites which are also characterised by PM_{10} , $PM_{2.5}$ and PM_1 concentrations similar to those measured in the reference site, the points previously identified on the diagram are further categorised according to the corresponding PM_{10} concentration values. In particular, the PM_{10} concentration values are grouped into twelve classes. The first eleven classes vary from 0 to $50 \mu\text{g m}^{-3}$ using a step of $5 \mu\text{g m}^{-3}$. The last class includes all the PM_{10} concentration values higher than $50 \mu\text{g m}^{-3}$.

3 Results and discussion

In order to compare the Agri Valley data with respect to simultaneous PM measurement reported in literature, both $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios were utilized.

The triangular diagram (Fig. 2) points out that PM concentration ratios calculated for Agri Valley, i.e. $PM_{2.5}/PM_{10} = 0.64$ and $PM_1/PM_{10} = 0.52$, fall close to the PM concentration ratios calculated for residential environment in Spokane (Haller et al., 1999), urban environment in Vienna (Gomišček et al., 2004), industrial environment in Linz (Gomišček et al., 2004), roadside environments in Hong Kong (Cheng et al., 2006), urban background environments in Birmingham (Yin and Harrison, 2008) and urban environment in Barcelona (Pey et al., 2010). In fact, all these sampling sites show $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios included in a range of 0.64 ± 0.032 and 0.52 ± 0.026 , respectively. Moreover, regarding the ultrafine fraction, the

$PM_1/PM_{2.5} \geq 0.5$ highlights that its contribution to the fine fraction is not negligible. This last result is shown for almost all the $PM_1/PM_{2.5}$ concentration ratios considered.

Taking again into account the $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios found in the $\pm 5\%$ range and also considering the sampling season, the results show that the Agri Valley PM concentrations ratios are comparable with those calculated by Gomišček et al. (2004) and Cheng et al., (2006) during the Summer season, accordingly with the Agri Valley measurements collected mainly during the warm period.

The $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios observed for the Agri Valley are plotted in a segment of the triangular diagram where it is possible to find most of the $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios calculated for urban sites referring to large cities such as Barcelona, Vienna, Athens, Birmingham, Milan, Madrid, Helsinki among others (Theodosi et al., 2011; Pérez et al., 2010, 2008; Pey et al., 2010; Amato et al., 2009; Rodríguez et al., 2008; Yin and Harrison, 2008; Cheng et al., 2006; Giugliano et al., 2005; Artiñano et al., 2004; Vecchi et al., 2004; Gomišček et al., 2004; Li and Lin, 2002; Vallius et al., 2000) as well as to sites characterized by vehicular traffic and construction/industrial emissions (Klejnowski, 2012; Massey et al., 2012; Hieu and Lee, 2010; Pey et al., 2009; Gomišček et al., 2004; Li and Lin, 2002; Wu et al., 2002; Querol et al., 2001) despite their not all being included in the 5% range. Furthermore, in this segment of the triangular diagram, the fine fraction is predominant with respect to the coarse fraction, with $PM_{2.5}/PM_{10}$ concentration ratios ranging from about 0.5 to 0.7. Instead, PM_1/PM_{10} can range between about 0.6 and 0.3. Moreover, the intermodal size fraction is always lower than the coarse fraction, i.e., $PM_{2.5} - PM_1/PM_{10} - PM_1$ ratio below 0.5.

The triangular diagram also shows that both the $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios recorded for the Agri Valley are different from those calculated for rural and semirural sites (Spindler et al., 2010, 2004; Yin and Harrison, 2008; Laakso et al., 2003; Putaud et al., 2002). The PM concentration ratios calculated for these sites fall in a segment of the diagram characterized by values of the PM_1/PM_{10} above 0.5 as well as high values of the $PM_{2.5}/PM_{10}$ concentration ratio, which are above 0.7 with

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predominance of a size fraction with another one. Moreover, in the future, the diagram could be used to compare the PM concentration ratios refining the data with respect to the climate conditions and specific pollution events.

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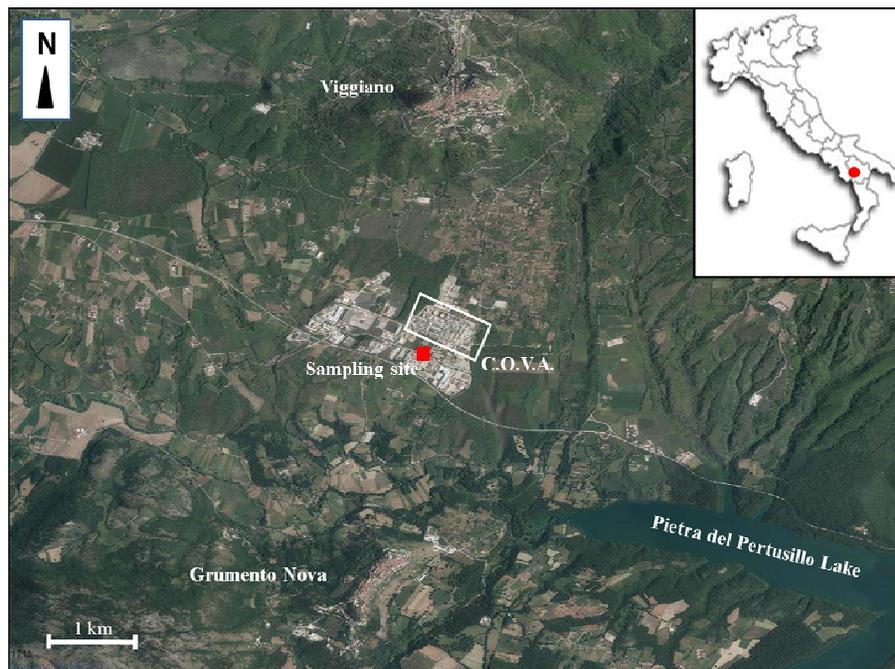


Figure 1. COVA plant and PM sampling site locations. Courtesy of orthophotograph AGEA 2011 <http://rsdi.regione.basilicata.it/>.

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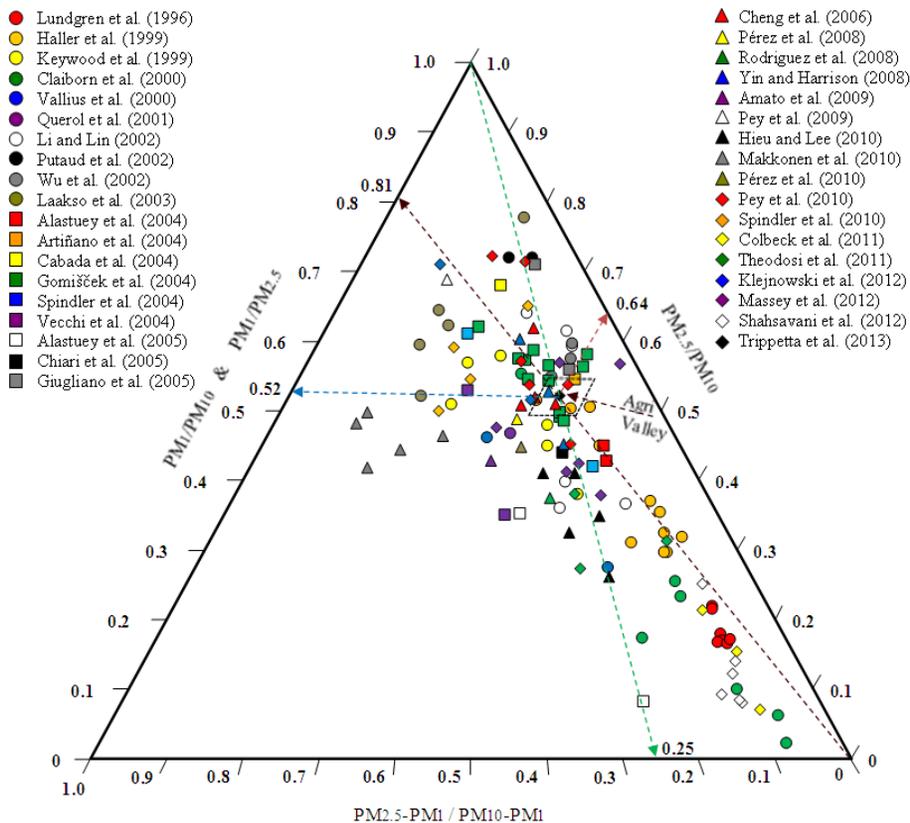


Figure 2. Triangular diagram for PM_1/PM_{10} , $PM_{2.5}/PM_{10}$, $PM_1/PM_{2.5}$ and $(PM_{2.5} - PM_1)/(PM_{10} - PM_1)$ concentration ratios.

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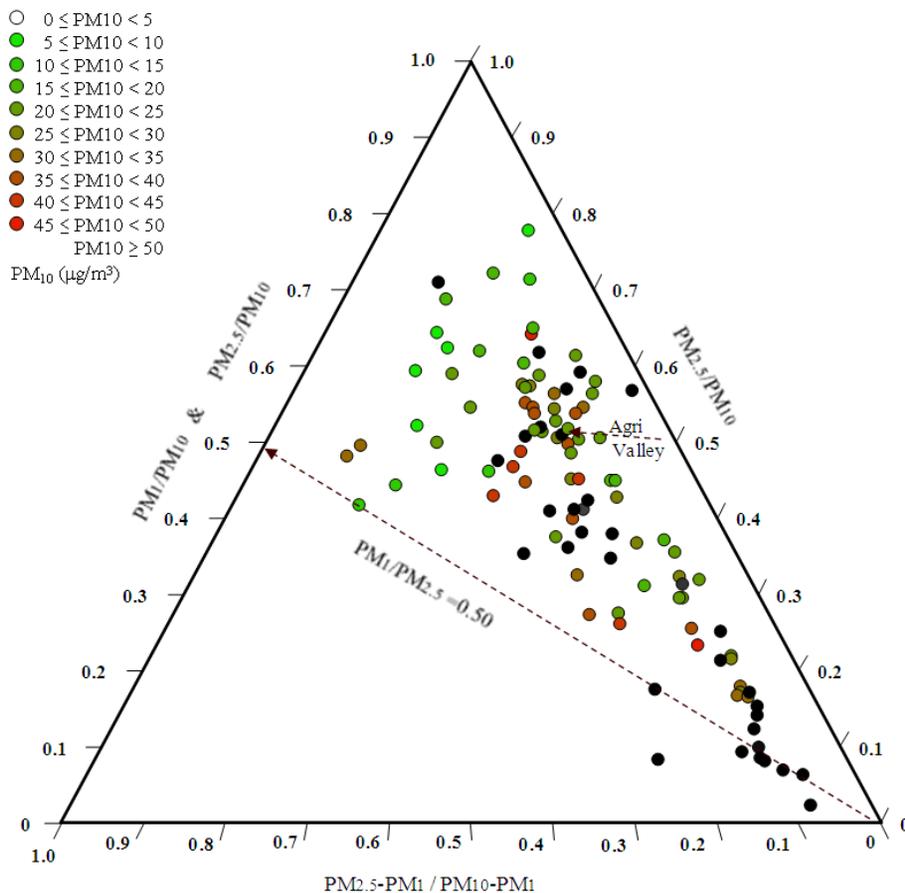


Figure 3. The triangular diagram of PM concentration ratios further categorized according to the PM_{10} concentrations. The line in the centre of the triangular diagram represents $PM_1/PM_{2.5}$ concentration ratio = 0.5.

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