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Seasonal variability of size segregated particulate matter ratios: the use of a novel approach to its representation and study

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Abstract

The present study deals with a novel approach to represent the seasonal variability of the ratios between the mass concentrations 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). To this aim, studies presenting simultaneous measurements of PM_{10}, $PM_{2.5}$ and PM_1 mass concentrations were considered and the corresponding ratios (i.e., PM_1/PM_{10}, $PM_{2.5}/PM_{10}$, $PM_1/PM_{2.5}$ and $PM_{2.5}$-PM_1/PM_{10}-PM_1) were calculated and displayed by a triangular diagram. Results point out that the data collected during winter season are mostly characterized by higher values of the $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} ratios and are displayed toward the upper left region of the triangular diagram, such that in some cases the intermodal size fraction can contribute substantially to fine fraction and in most cases that the intermodal size fraction can be of comparable magnitude with coarse size fraction. Moreover, the most of the data referring to winter season accumulate into a cluster. Instead, the data collected during summer season are mostly displayed toward lower values of $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} ratios with some exceptions for summer wildfire episodes. Finally, during the warm season no clusters are observed.

Keywords: PM, seasonal variability, triangular diagram.

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1. Introduction

It is well know that human activities and natural processes contribute to the formation and emission in the environment of the particulate matter (PM), which turns out to be a very complex and heterogeneous mixture of solid as well as liquid particles suspended in air. The sources of PM usually differ and they include a wide range of types. Typically, the fine and submicron particles (i.e., $PM_{2.5}$ and PM_1, aerosol particles with aerodynamic diameters less than 2.5 and 1 μm, respectively) are produced from combustion processes, transformations of gaseous species (Trippetta et al., 2013; Marawska et al., 2008), whereas coarser ones (i.e., PM_{10} aerosol particles with aerodynamic diameters less than 10 μm) originate from natural sources such as re-suspension of local soil by winds, desert dust, forest fire, volcanic eruptions (Aleksandropoulou & 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). According to the particle size, PM can pose risks to human health because of its adverse effects both on the respiratory and cardiovascular systems (Pope & Dockery, 2006). In fact, coarse particles are likely to be deposited in the extra-thoracic and upper bronchial region. Instead, fine and submicron particles may travel deeply into the lungs and may be deposited in the lower bronchial and alveolar regions.

In the light of this, simultaneous measurements of different PM fractions were performed to identify the PM formation and emission sources as well as to plan possible actions in order to mitigate the potential PM adverse effects (Russell & Brunekreef, 2009).

Moreover, the ratios between the mass concentrations of different PM size fractions have been studied in order to compare the PM levels among several sites (Speranza et al., 2014; Shahsavani et al., 2012; Claiborn et al., 2000), to achieve preliminary indication about PM emission sources (Perez et al., 2010; Vecchi et al., 2004) as well as to highlight the seasonal variability of the PM mass concentrations (Pérez et al., 2008; Giugliano et al., 2005; Artiñano et al., 2004).

In this context, the present study deals with a novel approach to represent the seasonal variability of the ratios between the mass concentrations of size segregated PM (i.e., PM_1/PM_{10}, $PM_{2.5}/PM_{10}$, $PM_1/PM_{2.5}$ and $PM_{2.5}$-PM_1/PM_{10}-PM_1). To this aim, simultaneous measurements of PM mass concentrations reported in literature have been considered and the corresponding ratios have been calculated and displayed by means of a dedicated triangular diagram.

2. Methodology

The triangular diagram, based on Sneed and Folk's original idea (Sneed and Folk, 1958) has been opportunely arranged both to represent and study the seasonal characteristic of the ratios between the mass concentrations of the size segregated PM. This approach is based on the calculation of the ratios between $PM_{2.5}$ and PM_{10} mass 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} mass concentrations (i.e., PM_1/PM_{10} that is the submicron fraction contribution to the PM_{10}). Moreover, the ratios between PM_1 and $PM_{2.5}$ mass concentrations (i.e., $PM_1/PM_{2.5}$ that is the submicron fraction contribution to the fine fraction) and the ratios between

$PM_{2.5}$-PM_1/PM_{10}-PM_1 (that is the proportion between the intermodal and the coarse fractions) are represented.

Studies presenting simultaneous measurements of PM_1, $PM_{2.5}$ and PM_{10} mass concentrations have been considered and the corresponding ratios have been calculated and displayed on the triangular diagram by means of dedicated software (Graham & Midgley, 2000).

3. Results and discussion

In order to represent the seasonal variability of simultaneous size-segregated PM measurements reported in literature, both $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios were utilized. The triangular diagram (Fig. 1) points out that the data collected during the winter season are displayed toward the upper region of the triangular diagram, with some exceptions for arid site (Haller et al., 1999). This means that the data collected during the winter season may be mostly characterized by higher values of the $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} ratios. The data referring to the winter season accumulate into a cluster whose limits are about 60% < $PM_{2.5}/PM_{10}$, 45% < PM_1/PM_{10}.

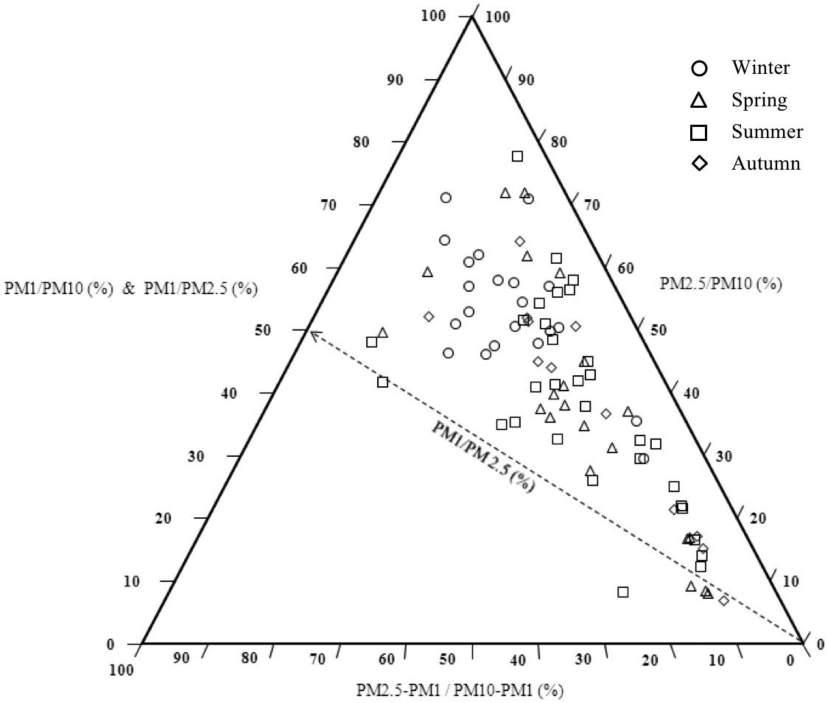

Fig. 1. Triangular diagram for PM10, PM2.5 and PM1of published data categorized according to the season of sampling. Alastuey et al. (2004); Alastuey et al. (2005); Cheng et al. (2006); Chiari et al. (2005); Colbeck et al. (2011); Giugliano et al. (2005); Gomišček et al. (2004); Haller et al. (1999); Hieu and Lee (2010); Keywood et al. (1999); Klejnowski et al. (2012); Laakso et al. (2003); Li and Lin (2002); Lundgren et al. (1996); Makkonen et al. (2006); Massey et al. (2012); Putaud et al. (2002); Rodríguez et al. (2008); Shahsavani et al. (2012); Spindler et al. (2004); Trippetta et al. (2013) ; Vallius et al. (2000); Vecchi et al. (2004).

Furthermore, these data are displayed toward large values of the intermodal size fraction ($PM_{2.5}$-PM_1/PM_{10}-PM_1 ratio above 25%) such that in some cases intermodal size fraction can be of comparable magnitude with the coarse size fraction ($PM_{2.5}$-PM_1/PM_{10}-PM_1 ≥ 50%). Moreover, the intermodal size fraction can contribute substantially to the determination of the fine size fraction (low values of PM_1/$PM_{2.5}$, about 70-60%). Likewise during winter season, the data collected during the autumn season is partially included in the above cluster, exception for arid site (Haller et al., 1999), dusty roads (Colbeck et al. 2011) and sites interested by monsoon precipitations (Li & Lin, 2002). The increase of $PM_{2.5}$/PM_{10}, and PM_1/PM_{10} concentration ratios during the colder seasons could be attributed to the increase in the emission of fine particles and submicron particles due to an intensification of the anthropogenic activities such as domestic/industrial heating and fuel consumption activities (e.g., wood-burning) (Klejnowski et al. 2012; Theodosi et al. 2011; Spindler et al. 2007), to the low mixing height that allows the accumulation of secondary organic aerosol (SOA) precursors with the effect of increasing their formation rate (Strader et al., 1999; Han et al. 2010) and to wet weather condition, which determine a suppression of coarse particles.

On the contrary, most of the data collected during the summer and spring seasons are displayed toward low values of the $PM_{2.5}$/PM_{10} < 60% and PM_1/PM_{10} < 45% and toward small values of the intermodal size fraction ($PM_{2.5}$-PM_1/PM_{10}-PM_1 ratio below 25%) where the contribution of intermodal size fraction to PM may be rather limited. However, some exceptions have been recorded for wildfire episodes (Makkonen et al., 2006) and for a south boreal climate (Laakso et al., 2003) in summer and spring seasons, respectively. A prevalence of the coarse size fraction on the fine/submicron size fractions observed during the warmer seasons may be due to the dry weather conditions which could facilitate the dust resuspension due to vehicles travelling, fugitive dusts and dust long-range transport, determining an increase of the emission of coarse particles associated with natural as well as to anthropogenic mechanical processes (e.g. material grinding and crushing) (Vecchi et al. 2004; Haller et al., 1999).

4. Conclusions

The study presented a novel approach to show the seasonal variability of the ratios between the mass concentrations of PM_{10}, $PM_{2.5}$ and PM_1 based on the use of a triangular diagram. The use of this approach allowed us to point out that the PM mass concentration ratios, plotted on the triangular diagram, are segregated in distinguishable regions related to colder and warmer seasons. In particular, the triangular diagram highlights that during the colder seasons the intermodal size fraction may not be negligible, both with respect to coarse and fine size fractions. Instead, during the warmer seasons clusters are not observed. The proposed triangular diagram, applied to PM mass concentration ratios, represents a suitable approach to compare a large environmental data set using only one diagram.

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