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A novel approach to comparing 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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Abstract. This work presents a novel approach to comparing and graphically representing 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 the literature using $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios. With this aim, a dedicated triangular diagram was used. The proposed approach was applied to size-segregated particulate matter (PM) concentrations recorded in the Agri Valley (Basilicata region – southern Italy). Results show 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, its formation process and its 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 (Aleksandropoulou 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, and domestic heating (Caggiano et al., 2010; Lin and Lee, 2004). According to the particle size, PM can pose risks to human health because of its adverse effects on both the respiratory and cardiovascular systems (Pope III and Dockery, 2006). In fact, coarse particles are likely to be deposited in the extra-thoracic and upper bronchial regions. 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 indications of 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 between 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 comparing and graphically representing simultaneous measurements of PM_{10}, $PM_{2.5}$ and PM_1 concentrations with similar data

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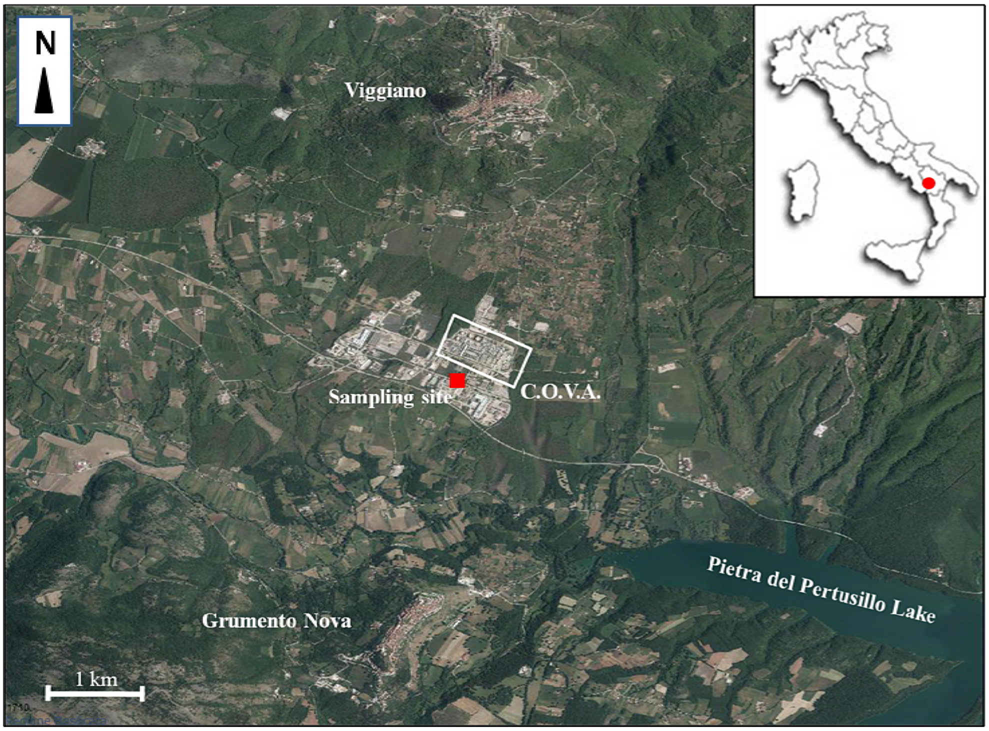

Figure 1. COVA plant and PM sampling site location. Copyright AGEA – Orthophoto 2011.

reported in the literature using both $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios. With this aim, a dedicated triangular diagram was used. The proposed approach was applied using PM concentration measurements (Trippetta et al., 2013) recorded in the 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 onshore reservoir (crude oil and gas) and the biggest oil/gas pre-treatment plant, identified as the Centro Olio Val d'Agri (COVA) in a rural/anthropized context (Fig. 1). The COVA plant produces 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 several small towns (from 1700 to 5400 inhabitants) in its surroundings.

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 at a reference site with similar measurements reported in the 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}), 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 package (Graham and Midgley, 2000).

The particular features of this diagram also allow 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 coarse fractions.

Each site will be identified in the diagram with a point representing the corresponding size-segregated PM concentration ratios. Two data points in the triangular diagram (e.g., P_a and P_b) can be considered closed if they have $PM_{1a}/PM_{10a} \approx PM_{1b}/PM_{10b}$ as well as $PM_{2.5a}/PM_{10a} \approx PM_{2.5b}/PM_{10b}$ within an acceptable tolerance. As a 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, comparing simultaneous sampling of PM in the triangular diagram enables a comparison of the ratios between 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 $PM_1/PM_{2.5}$) all together.

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 by no more than $\pm 5\,\%$. The

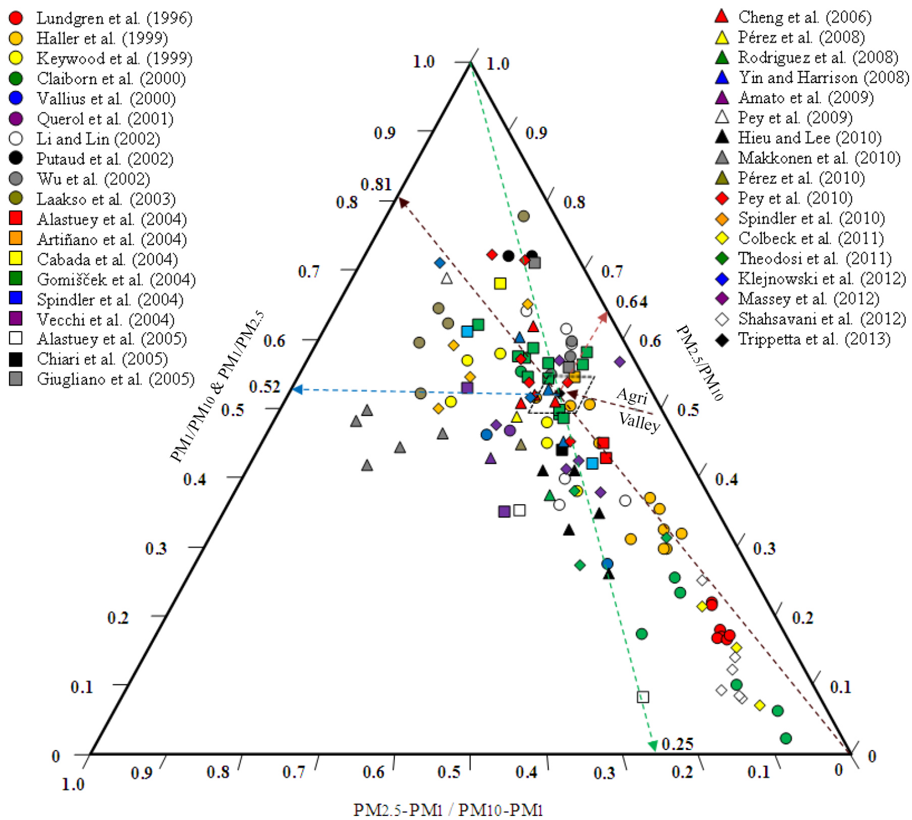

Figure 2. Triangular diagram for the PM_1/PM_{10}, $PM_{2.5}/PM_{10}$, $PM_1/PM_{2.5}$ and $(PM_{2.5} - PM_1)/(PM_{10} - PM_1)$ concentration ratios.

uncertainty for the PM_1/PM_{10} and of $PM_{2.5}/PM_{10}$ concentration ratios has been evaluated from the data reported in Trippetta et al. (2013). The combined uncertainty has been taken into account for $A = B/C$:

$$\frac{\delta_a}{A} = \sqrt{\left(\frac{\delta_b}{B}\right)^2 + \left(\frac{\delta_c}{C}\right)^2}, \tag{1}$$

$$\delta_x = \frac{s_x}{\sqrt{n}} (x = a, b, c), \tag{2}$$

(Bell, 1999). The uncertainty is within 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 values of PM_1, $PM_{2.5}$ and PM_{10} concentrations. Indeed, if two close points (e.g., P_a and P_b) displayed in the triangular diagrams are considered, they have 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}, and the concentration value of $PM_{2.5a}$ can be different from that of $PM_{2.5b}$. Hence, only if close points have PM_{10a} concentration $\approx PM_{10b}$ concentration, then PM_{1a} concentration $\approx PM_{1b}$ concentration and $PM_{2.5a}$ concentration $\approx PM_{2.5b}$ concentration.

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

3 Results and discussion

In order to compare the Agri Valley data with respect to simultaneous PM measurements reported in the 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 the Agri Valley, i.e., $PM_{2.5}/PM_{10} = 0.64$ and $PM_1/PM_{10} = 0.52$, are close to the PM concentration ratios calculated for residential environments in Spokane (Haller et al., 1999), urban environments in Vienna (Gomiščck et al., 2004), industrial environments in Linz (Gomiščck et al., 2004), roadside environments in Hong Kong (Cheng et al., 2006), urban background environments in Birmingham (Yin and Harrison, 2008), and urban environments 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 ranges of 0.64 ± 0.032 and 0.52 ± 0.026, respectively. Moreover, regarding the ultrafine fraction, $PM_1/PM_{2.5} \geq 0.5$ highlights the fact 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 ± 5 % range, and also considering the sampling season, the results show that the Agri Valley PM concentrations ratios are comparable with those calculated by Gomiščcek et al. (2004) and Cheng et al. (2006) during the summer season, and 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, and Helsinki, among others (Theodosi et al., 2011; Pérez et al., 2008, 2010; 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; Gomiščcek 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 et al., 2012; Massey et al., 2012; Hieu and Lee, 2010; Pey et al., 2009; Gomiščcek et al., 2004; Li and Lin, 2002; Wu et al., 2002; Querol et al., 2001), despite them 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 from about 0.6 to 0.3. Moreover, the intermodal size fraction is always lower than the coarse fraction, i.e., the $PM_{2.5} - PM_1/PM_{10} - PM_1$ ratio is below 0.5.

In addition, the comparison of the Agri Valley data with respect to simultaneous PM measurements reported in the literature can be further extended to all those sites whose $PM_1/PM_{2.5}$ and PM_1/PM_{10} concentration ratios plus their uncertainties partially overlap the ranges $PM_1/PM_{2.5} = 0.64 \pm 0.032$ and $PM_1/PM_{10} = 0.52 \pm 0.026$ in the triangular diagram. Within this framework, as a first step, the sites to be compared, considering also the season of sampling (warmer period) in the Agri Valley, are the industrial environment in Linz, the urban environment in Graz (Gomiščcek et al., 2004), and the residential site in Spokane (Haller et al., 1999).

This comparison considers a larger data set. Again, this shows that the Agri Valley PM concentration ratios are comparable with PM concentration ratios calculated for urban sites.

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., 2004, 2010; Yin and Harrison, 2008; Laakso et al., 2003; Putaud et al., 2002). The PM concentration ratios calculated for these sites fall into a segment of the diagram characterized by values of the PM_1/PM_{10} ratio above 0.5 as well as high values of the $PM_{2.5}/PM_{10}$ concentration ratio, which are above 0.7, with some exceptions for Gomišcek et al. (2004) and Spindler et al. (2004). Therefore, in this segment, the fine fraction and the ultrafine fraction are predominant over the coarse fraction, while the intermodal fraction is comparable with the coarse fraction. In fact, the $PM_{2.5} - PM_1/PM_{10} - PM_1$ concentration ratio observed can reach values above 0.5.

As Fig. 2 shows, the Agri Valley $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios are also quite different from those calculated for arid sites (Shahsavani et al., 2012; Lundgren et al., 1996), for sites affected both by dust storm originating in Asia (Claiborn et al., 2000) and strong African dust outbreak episodes (Alastuey et al., 2005), and dusty roads (Colbeck et al., 2011). In fact, the PM concentration ratios calculated for these sites are plotted towards the right lower vertex of the triangular diagram, where $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} are below 0.5. Therefore, in this segment, the coarse fraction is predominant with respect to the fine fraction and the ultrafine fraction. The intermodal fraction compared to the coarse fraction is found to be very low. In fact, the $PM_{2.5} - PM_1/PM_{10} - PM_1$ ratios are in a range between about 0.1 and 0.3.

In order to identify all the sites with similar PM_1, $PM_{2.5}$ and PM_{10} concentrations, the PM_{10} concentrations reported by the selected studies were grouped into eleven ranges. By considering the PM concentration of the sites whose $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios differ by no more than ± 5 % from the corresponding ratios calculated for the Agri Valley, the values of PM_1, $PM_{2.5}$ and PM_{10} concentrations recorded in the Agri Valley (11.0, 13.6 and 21.2 $\mu g\,m^{-3}$, respectively) are comparable with the PM_1, $PM_{2.5}$ and PM_{10} concentrations measured in Vienna (14.2, 17.5 and 26.1 $\mu g\,m^{-3}$, respectively), but they are quite different from those measured in Hong Kong (56.3, 71.0 and 110.3 $\mu g\,m^{-3}$, respectively) (Fig. 3). In conclusion, by considering $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios, the PM concentrations measured in the Agri Valley are comparable with those reported in Gomiščcek et al. (2004).

Finally, the Agri Valley data (i.e., the PM_1/PM_{10} and $PM_{2.5}/PM_{10}$ concentration ratios) are placed toward the upper part of the segment, where most of the data from urban sites can be found, next to the segment where most of the rural measurements are plotted, and far away from the data recorded at arid sites. Nevertheless, the contributions of the anthropogenic emissions are such that the data recorded are

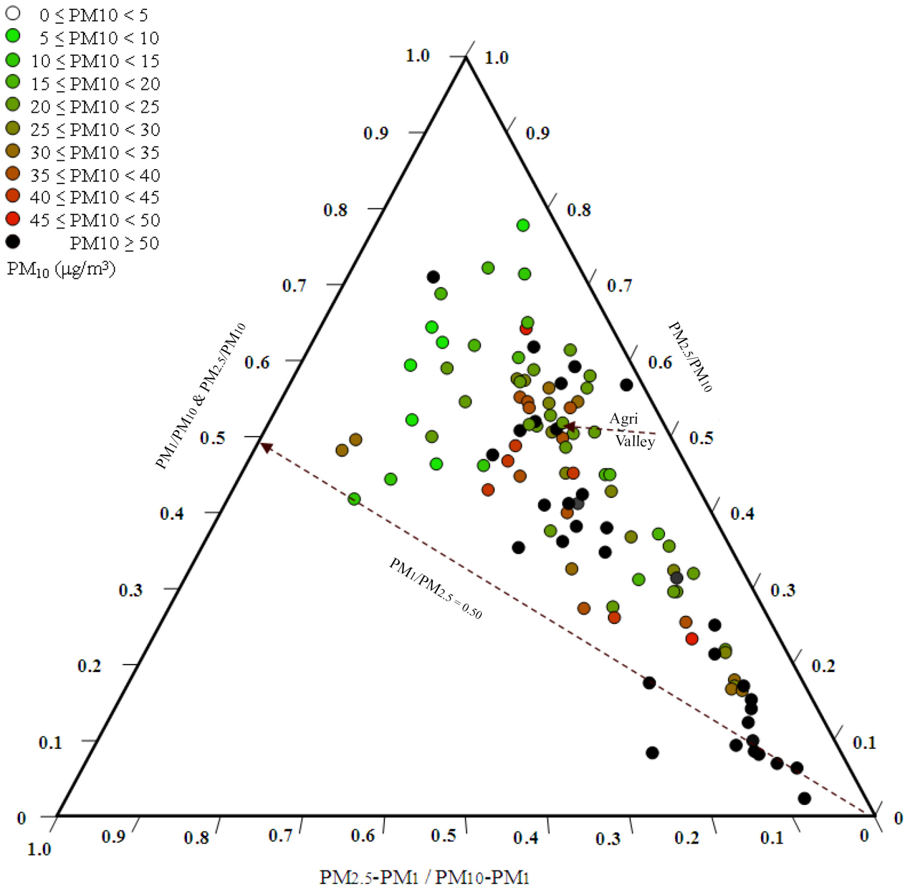

Figure 3. The triangular diagram of PM concentration ratios further categorized according to the PM_{10} concentrations. The line in the center of the triangular diagram represents a $PM_1/PM_{2.5}$ concentration ratio of 0.5.

very much comparable to those recorded at a typically urban site. All these results may be explained by considering the peculiarity of the area under study, and they are consistent with the emissions features of rural areas, where anthropogenic activities typical of small urban settlements and industrial plants processing oil/gas can be found.

4 Conclusions

A novel approach based on the use of a modified version of Sneed and Folk's triangular diagram was proposed and used to compare and graphically represent simultaneous measurements of PM_{10}, $PM_{2.5}$ and PM_1 concentrations recorded in the Agri Valley with similar measurements reported in the literature. With this aim, PM_1/PM_{10} and $PM_{2.5}/PM_{10}$ concentration ratios were used. Focusing on the $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios reported for the Agri Valley, they are plotted in an 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. Moreover, the Agri Valley $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} concentration ratios are both different from those reported for rural and semirural sites, and quite different from those referring to arid sites or sites affected by dust storms. Using PM_{10} concentration data, the results are that, among the identified urban environments, the values of PM_1, $PM_{2.5}$ and PM_{10} concentrations measured in the Agri Valley are comparable with those recorded in Vienna.

Therefore, even though the Agri Valley is an area mainly characterized by rural features, the presence of anthropogenic activities such as the oil/gas pre-treatment plant makes this area comparable to an urban site both in terms of PM concentration ratios and PM levels.

In conclusion, this work shows that the suggested approach allows a simple and clear identification of sites with comparable atmospheric PM concentrations.

As future work, the proposed approach could be used to evaluate both how the PM concentration ratios can depend on the seasons of sampling and to assess the 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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