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RELATIONSHIPS BETWEEN PM_{10} MASS CONCENTRATIONS AND AEROSOL OPTICAL PARAMETERS OVER MAGURELE, ROMANIA

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Abstract. This study is focused on relationship between ground-level Particulate Matter (PM_{10}) and columnar AOD (aerosol optical depth) and AE (Angstrom exponent) at Magurele, Romania. A two years data set (2014-2015) of measurements was used. The annual means of PM_{10} were about 25 µg m^{-3} (2014) and about 28 µg m^{-3} (2015) with highest level attained in cold season (about 29 µg m^{-3}). Seasonal variations of PM_{10} mass concentrations and AOD show PM_{10} has a seasonal pattern with a main maximum in winter (DJF), while AOD reached maximum values in summer (JJA). We obtained single peak frequency distributions for both AOD and AE, with mode values around 0.15 (for AOD) and 1.6 (for AE). Fine mode aerosols dominate the PM_{10} mass concentrations during the entire period. Best correlation coefficients between PM_{10} and AOD and between PM_{10} and AE were found during warm season and for the days when the columnar optical parameters were obtained from measurements with observation time for more than 10 hours a day.

Key words: aerosol optical depth, angstrom exponent, PM_{10}, air pollution.

1. INTRODUCTION

The aerosol or particulate matter (PM) is a very important component of atmosphere that plays a crucial role in regional and global climate change, contributes to changes of air quality and visibility and may determine serious health problems (Pope and Dockery, 2006; IPCC, 2013). Because the particulate matter covers a wide range of sizes and chemical compositions and has a large spatial and temporal variability, estimation of its effects is a challenge task for the scientific community. Aerosol effects are more pronounced and complex as smaller fractions PM_{10}, $PM_{2.5}$ or PM_1 (particles with a diameter less of 10, 2.5 or 1 µm) of PM are involved (e.g.Wilson and Suh, 1997; Seinfeld and Pandis, 2006). Therefore, the investigations of aerosols and their properties in various regions worldwide, including *in situ* and remote measurements, are necessary for understanding their effects. Ground-based measurements of aerosol optical properties are performed at global level by several known networks: AERONET (Aerosol Robotic Network), PHOTONS (Photometrie pour le Traitement Operationnel de Normalisation Satellitaire), SKYNET (Skyradiometer Network), AEROCAN (Canadian Sunphotometer Network, sub-network of AERONET) etc. (Holben et al., 1998; Bokoye et al., 2001; Uchiyama et al., 2005; Goloub et al., 2008). Recently, increasing studies have focused on differences in aerosol columnar properties, as well as on their relationship with ground measured particulate matter (e.g. PM_{10}), cloud condensation nuclei (CCN) and aerosol chemical composition (Engel-Cox et al., 2006; Cheng et al., 2008; Garland et al., 2008; Andreae, 2009; Esteve et al., 2012). The primary aerosol parameter provided by remote sensing is the aerosol optical depth (AOD), describing the extinction at a given wavelength of the electromagnetic radiation in an atmospheric column, attributed to aerosols present in column (Cachorro et al., 2008; Kokhanovsky et al., 2009; Toledano et al., 2012). Angstrom exponent (AE) derived from AOD wavelength dependence (Eck et al., 1999) helps us to distinguish between aerosol types. AE will show the smallest values for the largest sizes of particles. For aerosols of diameter less than 0.1 µm, the Angstrom exponent is greater than 1.8 and the fine mode aerosol dominates the atmosphere. The particles in accumulation mode (diameters between 0.1 and 1 µm) have AE values between 0.7 and 1.8. When particles with diameters greater than 1 µm (e.g., dust) prevail in the atmosphere, AE has values smaller than 0.7 (Kaufman, 1995). The Angstrom exponent AE can be expressed as:

$$\mathrm{AE}_{440-870} = -\frac{\Delta \ln(\mathrm{AOD})}{\Delta \ln \lambda} \tag{1}$$

where AOD is the aerosol optical depth for the paired wavelength (λ) of 440 and 870 nm. However, the link between measured columnar properties and the surface concentrations of PM_{10} (or $PM_{2.5}$) is not a straightforward problem (among interfering factors: variability of size distributions, atmospheric humidity, variations of planetary boundary layer, precipitations, aerosol lifetime). Ground-based measurements of PM_{10} are also subject to different systematic errors depending on the measurement technique. Therefore, despite their limited spatial representativeness, empirical regression models and relationships are usually established and used (e.g. Koelemeijer et al, 2006; Rohen et al., 2011; Estelles et al., 2012; Bilal et al., 2017) in the main effort of scientists to improve the understanding on PM, which is necessary for monitoring compliance.

Our study is focused on the analysis of the correlations between coincident measurements of optical columnar parameters (AOD and AE) and ground-level PM_{10} aiming to establish a relationship between them, relationship that could be further used to estimate PM_{10}. We selected Magurele (latitude 44.38N, longitude

26.03E, elevation 65.0m) in order to perform our analysis, as the small city is located at 10 km SW from Bucharest, the largest and most developed city in Romania. Previous studies on temporal and spatial variations of air pollutants, including PM in Bucharest greater area (Iorga et al., 2015; Stefan et al., 2015), showed that Magurele city suffers from the air pollution in Bucharest, as the topographic characteristics of the region and meteorological factors favor this situation (Figure 1). Some optical properties of aerosol in Bucharest area have been evaluated in other few papers (Radu et al., 2008; Mihai and Stefan, 2011; Filip and Stefan, 2011; Gothard et al., 2014; Toanca et al., 2017; Cazacu et al., 2017). The single previous analysis (Filip and Stefan, 2011) focused on PM_{10}-AOD relationship we found was made on few short periods in 2007 and 2009, and we shall discuss our results by comparison to. To our knowledge, no study aimed to analyze the coincident measurements of optical aerosol columnar properties for such long time and ground mass concentrations of aerosol. Present study extends knowledge about pollution at local scale, estimated using data from AERONET that may thus help to improve or replace the ground-level PM_{10} measurements when the monitoring station encounters technical difficulties.

The details of measurements performed at Magurele are presented in Section 2, together with data processing methods, and the results of the analysis are discussed in Section 3.

2. DATA AND METHODS

PM_{10} mass concentrations are monitored in Magurele on a daily basis at the suburban background station of the Air Quality Automatic Monitoring Network of Bucharest, maintained by the National Environmental Protection Agency (EPA-B). A parallel data set of daily PM_{10} mass concentrations (between 1st June and 31st December 2014), was manually collected using a low-volume sampler (sampling rate 2.3 $m^3 h^{-1}$) on the roof of Faculty of Physics (nearby of the sampling locations of EPA-B and of sun photometer, within 200 m distance). This data set was used for comparison with the derived PM_{10}, calculated as explained at the end of the section. The ground-level PM_{10} mass concentrations were obtained by gravimetric method following the standards SR EN 12341:2002 (ASRO, 2002) and SR EN 14907:2006 (ASRO, 2006).

The columnar optical parameters of aerosol were obtained using a CIMEL Automatic Sun Tracking Photometer CE 318, installed at National R&D Institute for Optoelectronics (INOE 2000), Department of Atmospheric Observatory (RADO), which is part of AERONET. We used the third version of data, acquired every 15-minutes from AERONET level 1.5 (cloud-screened) with retrieval error less than 5%. From the 302 daily columnar optical observations available from 1st January 2014 to 31st December 2015, we selected 243 days that matched simultaneous ground-level PM_{10} measurements. There were with 22% more recorded values in 2014 than in 2015 because for April, May and June 2015 data were not available (Figure 2).

Fig. 1 – Map (created using Google Earth) showing location of Magurele in the vicinity of Bucharest and the position of the sun photometer belonging to AERONET and that of the Air Quality Monitoring Station (located within 200 m distance).

To establish the relationship between AOD, AE and PM_{10} mass concentration, correlation analysis, expressed by Pearson coefficients, statistically significant at 95% confidence interval, was performed for all pairs of observations.

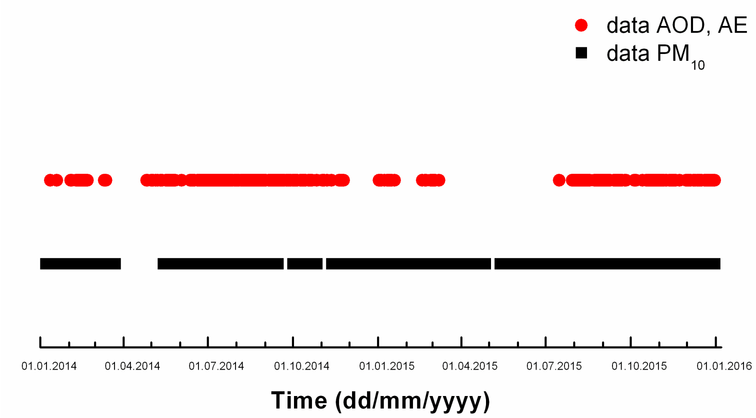

Fig. 2 – Temporal data coverage with collocated recorded values for aerosol optical parameters from from sun photometer and ground-level PM_{10} mass concentration during 2014-2015.

Correlation analysis has been also done for two separate time-periods: cold season (from 16th October to 15th April in both years) and warm season (from 16th April to 15th October in the both years), because these periods corresponds (within few days) to the onset/offset of the domestic heating period in the area. Besides the seasonal correlation, we examined the correlations PM_{10}-AOD and PM_{10}-AE for a daily observation time of sun photometer greater than 10 hours. This means that we selected the coincident days with ground-level PM_{10} measurements and with AOD and AE recorded data more than 10-hours per day. We prove in next section that this criterion of selection helps to grow the correlation between daily averages of aerosol optical parameters with the 24-hour values of PM_{10} mass concentration.

The linear regressions between daily PM_{10} as dependent variable and AOD at 340, 500 and 1020 nm and AE as independent variables were examined, as well. The resulted coefficients were used to derive PM_{10} mass concentrations, which were then compared with the independent PM_{10} data set, obtained by the parallel measurement campaign in 2014.

3. RESULTS AND DISCUSSIONS

3.1. VARIATIONS OF PM_{10} MASS CONCENTRATIONS AND OPTICAL PARAMETERS

The temporal variations of both PM_{10} mass concentrations and optical parameters AOD and AE and the relationships between them were analyzed using daily data from AERONET and EPA-B and Table 1 shows their annual and seasonal averages.

Table 1

Annual and seasonal averages of PM_{10} mass concentrations, AOD (500 nm) and AE (440-870 nm)

	Annual average (2014)	Annual average (2015)	Warm season average	Cold season average
PM_{10} (µg m^{-3})	24.48±14.02	27.73±16.40	23.37±8.93	28.84±19.56
AOD	0.20±0.12	0.23±0.12	0.24±0.11	0.19±0.10
AE	1.51±0.34	1.49±0.28	1.56±0.17	1.44±0.33

We note that PM_{10} level was higher in 2015 than in 2014, with relatively the same presence of fine-mode particles, but finer particles appear during warm season. The seasonal means show lower value of PM_{10} mass concentration and higher value of AOD in warm season than in cold season. Our results are similar to those in previous studies for Magurele (e.g. Gothard et al., 2014; Iorga et al., 2015; Stefan et al., 2015). Table 1 shows that annual means of PM_{10} mass concentrations are below the EU-imposed annual limit value of 40 µg m^{-3} for both years (EC, 2008), although PM_{10} mass concentration daily variations (Figure 3) indicate few pollution episodes in February 2014 and November 2015. Figure 3 also shows daily AOD variations between 0.05 and 0.90, AE ranges from about 0.5 to 2.4, and greater values for fine mode (FM) than coarse mode (CM) most of the time with few exceptions in the first part of year 2014 when some values of CM exceeded FM. Correspondingly, AE values were small and AOD high. These associate to local pollution episodes, when higher pollutant levels due to local anthropogenic activities in presence of a high-pressure system over the area, were detected. (Sandu et al., 2008; Iorga et al, 2015).

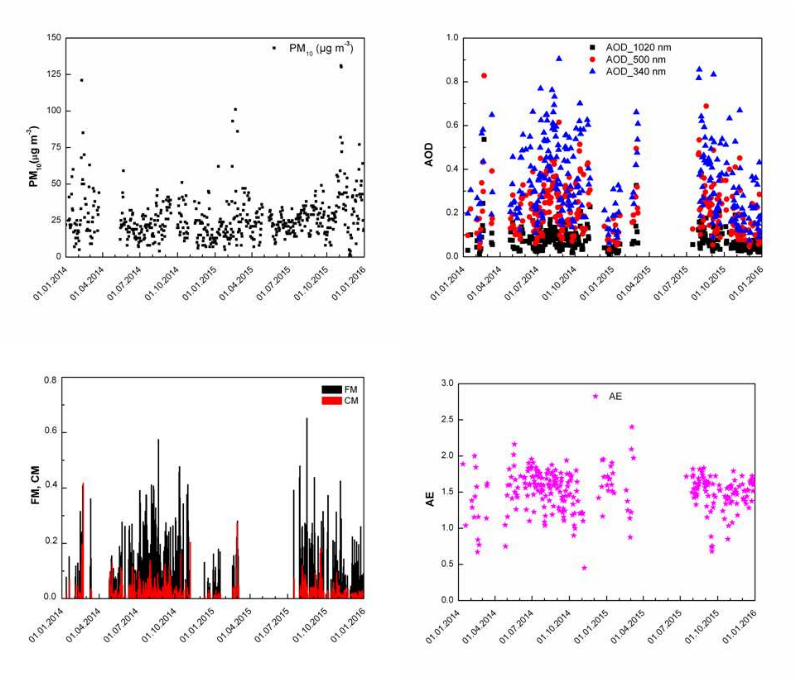

Fig. 3 – Daily variations of ground-level PM_{10} mass concentration (up-left), AOD at 340, 500, 1020 nm (up-right), fine and coarse fraction (down-left) and of AE_440-870 nm (down-right).

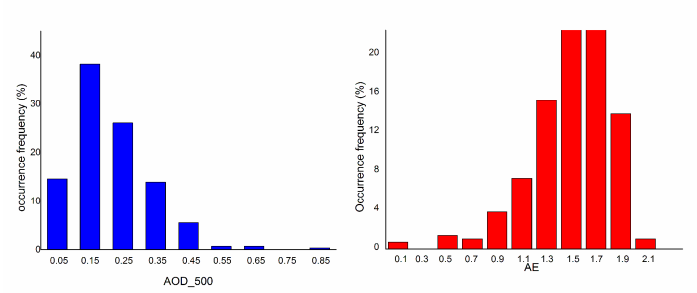

Fig. 4 – Frequency distributions of AOD (500 nm) and AE (440-870 nm) in the period 2014-2015.

Frequency distributions of AOD (500 nm) and AE in Figure 4 for all daily data reveals single peak distributions for both AOD and AE, with modal values of around 0.15 and 1.6, respectively. The cumulative frequency of AOD in the range of 0.15 - 0.40 is about 70% and that of AE in the range of 1.0 – 2.1 is about 75%, This result is concordant to the results of Iorga et al. (2015), which reported a large contribution (70–80%) of fine particles to the particulate mass in Bucharest Greater Area over a five-years (2005-2010) study.

AOD (500 nm) monthly values (Figure 5) shows a similar variation during the year, with greatest values in spring/summer and minimum in December and January and a clear and persistent presence of fine mode aerosols during the entire studied period. The seasonal AOD averages are 0.14 in winter, 0.20 in spring, 0.28 in summer and 0.21 in autumn. This result is similar with the monthly variation of AOD averaged over Europe found by (Koelemeijer, 2006). The explanation might consist in contribution to loading of the atmosphere with aerosols from other sources than local in spring/summer.

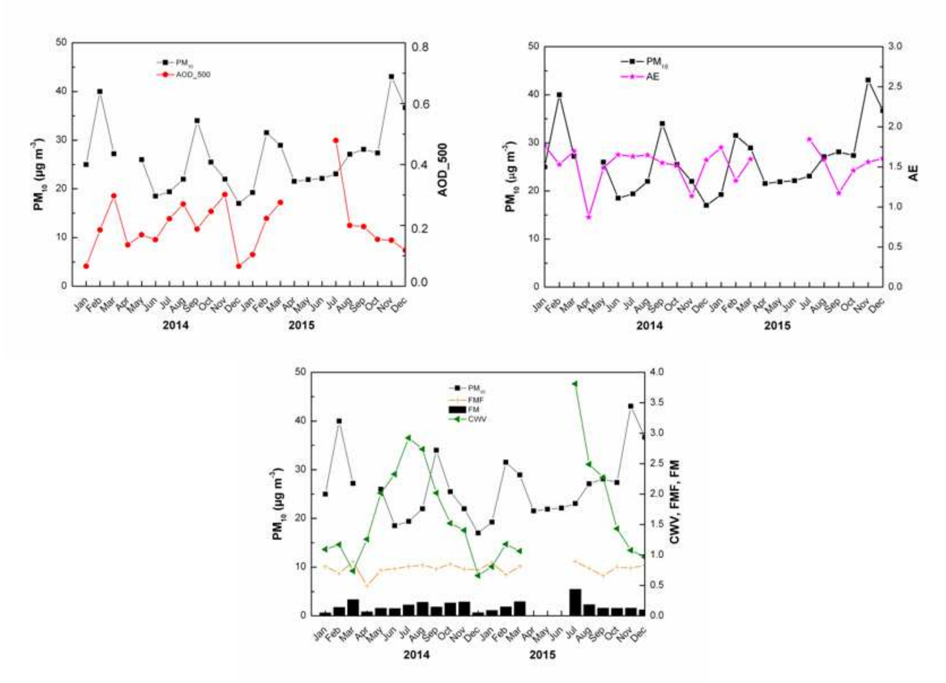

Fig. 5 – Monthly variations of ground-level PM_{10} mass concentration, AOD (500 nm), AE_440-870 nm, AOD fine mode (FM) and fine mode fraction (FMF) and content of water vapor (CWV).

The air mass back-trajectories and atmospheric circulation over Bucharest-Magurele area support it (Nicolae et al., 2008; Gothard et al., 2014; Sandu et al., 2008). Moreover, Iorga et al. (2015) showed that emissions from the relatively uniform sources in inner Bucharest spread out over the larger area, leading to 29% from pollution episodes per year to be of regional nature.

Some seasonality could be seen in monthly variations of fine mode AOD (FM) and the associated fine mode fraction (FMF) but this finding should be seen with caution because monthly averages in April 2014, November 2014 and July 2015 have a low confidence level as they were obtained from a reduced number of measurements. As FMF exceed 0.6, the dominance of fine aerosol appears once again obvious during the entire period, following the model proposed by Lee et al. (2010). Content of water vapor (CWV) attains its highest values in summer and lowest in winter, following the climate characteristics (Sandu et al., 2008) in the area (cold and dry weather in winter, hot and humid in summer). AE monthly means (between 1.0 and 2.0) show low seasonality but fit within the ground-level values (between 1.4 and 2.3) obtained by Mihai and Stefan (2011) using a nephelometer at 450 nm and 700 nm in a study covering 2008 and 2009.

Overall, the atmosphere at Magurele is dominated by small particles (diameter less 1 μm) showing low seasonality along the year, and with AOD variations determined mostly by regional influences.

3.2. CORRELATION BETWEEN PM_{10} MASS CONCENTRATIONS AND OPTICAL PARAMETERS

A synthesis of correlations between daily PM_{10} mass concentration and aerosol optical depth AOD (340 nm, 500 nm, 1020 nm), and Angstrom exponent is presented in Figure 6.

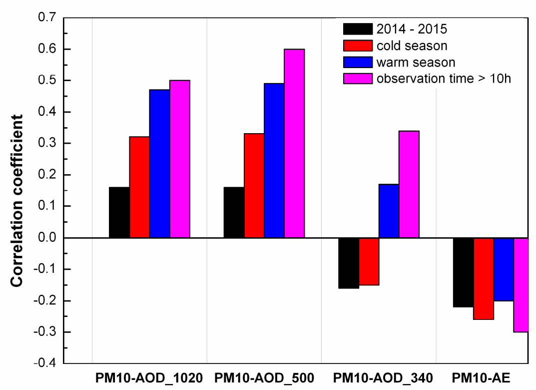

Fig. 6 – Pearson's correlation coefficient for PM_{10}-AOD (340, 500, 1020 nm), and PM_{10}-AE during different periods: 2014-2015; cold season (16th October-15th April); warm season (16th April-15th October); selected measurements with observation time more than 10 hours.

We identified a weak to moderate positive correlation that could be due to variability of the ground-level PM_{10} but also due to different intrinsic measurement techniques (24-h measurements for PM_{10}, while daily sun-photometer data are based on nearly instantaneous observations, with values recorded every 15-min, from morning to evening hours with short observation time for AOD). Greater correlation coefficients were obtained for PM_{10} – AOD (or AE, respectively) relationship with daily AOD and AE calculated from measurements with observation time more than 10 hours a day. In this case, the daily averages of AOD and AE are more confident in connection with the cumulative mass concentrations of PM_{10}. Another explanation could be related to the size distributions of particles varying from medium-to-fine size. These aerosol particles have a large influence on AOD but contribute much less to mass of PM_{10} in comparison to larger particles. The correlation of AOD (340 nm) and PM_{10} is positive only for the warm season and AOD recorded values more than 10-h daily. A possible explanation could be related on the interaction of UV radiation with fine particles of $PM_{10.}$ Angstrom exponents are negatively correlated with PM_{10} mass concentrations with small correlation coefficients in all the seasons.

Figure 7 shows the scatter-plots and relationships between PM_{10} and AOD (340, 500 and 1020 nm), and between PM_{10} and AE (440-870 nm) from measurements with observation time for more than 10 hours a day.

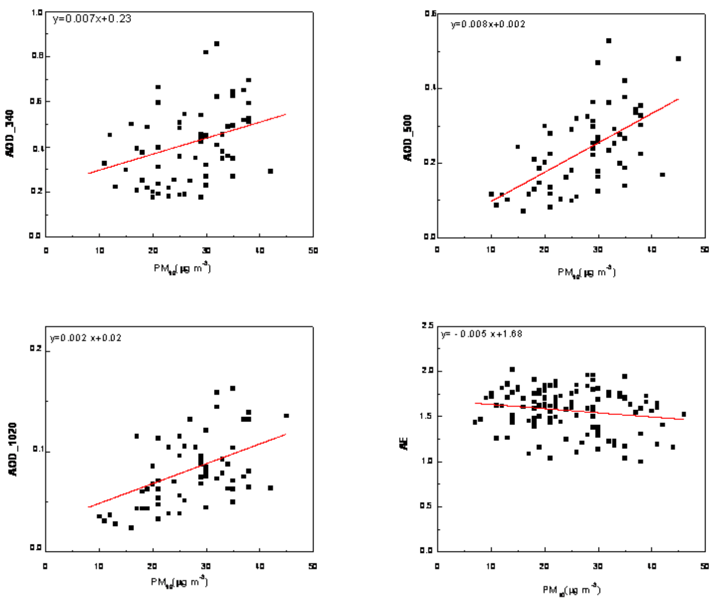

Fig. 7 – Correlations between PM_{10} and AOD (340, 500, 1020 nm) wavelength, and between PM_{10} and AE for measurements with observation time more than 10 hours during entire period 2014-2015.

The PM_{10} – AOD relationship appear to exhibit similar trends for daily values (Figure 7) in all spectral ranges. The linear regression functions of the daily AOD (y) with the ground-level PM_{10} concentration (x) were: y = (0.007 ± 0.002) x + (0.230± 0.071) for AOD at 340 nm; y = (0.008 ± 0.001) x + (0.002± 0.003), for AOD at 500 nm; and y = (0.0020±0.0046) x + (0.0210±0.0120) for AOD at 1020 nm. The linear regression function corresponding to the PM_{10} – AE relationship is: y = -(0.005 ± 0.002) x + (1.680± 0.061). The corresponding correlation coefficients are: 0.34; 0.60; 0.35 and -0.30, respectively. The previous study of Filip and Stefan (2011) revealed higher correlations (around 0.69 in 2007; 0.74 in 2008) for PM_{10}-AOD (500 nm) relationship could be obtained if all data, ground-level and columnar, are normalized to their annual means and the measurements are performed in a stable atmosphere. We found a good agreement (PM_{10} absolute error less than 10-15 μg m^{-3}) between our and their results despite the fact that our relationships were obtained covering all atmosphere regimes; therefore have the potential to cover a wider range of situations. We do not expect a perfect agreement between columnar optical parameters and ground-level PM_{10} mass concentrations even under ideal conditions, as other factors can contribute to scatter of data, as for example, variability in vertical distributions of PM_{10} mass concentrations, variations of height of boundary layer, meteorological parameters. Therefore, the above relationships can be used with cautions to derive daily averaged PM_{10} concentrations from the observed AOD and AE. Figure 8 shows a comparison between PM_{10} mass concentrations, calculated using the linear regression coefficients obtained from the correlation PM_{10} - AOD at 500 nm, and the independent, ground-level PM_{10} mass concentrations data set obtained in our parallel measurement campaign in 2014.

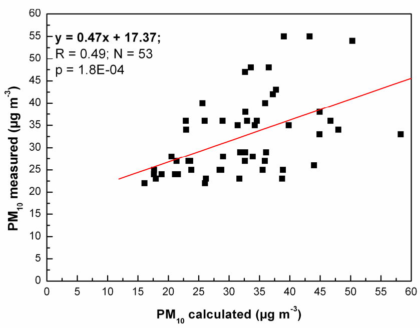

Fig. 8 – Ground-level measured versus calculated PM_{10} mass concentrations for measurements with observation time more than 10 hours during entire period 2014-2015.

4. CONCLUDING REMARKS

The analysis of optical aerosol parameters of the columnar aerosol and measured surface mass concentrations in an area influenced by regional pollution showed the difficulty to conclude when the columnar optical parameters characterize with accuracy the ground-level PM_{10} measurements. Some local characteristics of atmosphere loading with aerosol were nevertheless, emphasized: The seasonal averages of AOD at 500 nm present the greatest value in summer (0.28), while the smallest is in winter (0.14). Almost 70% of AOD (500 nm) values situate between 0.15-0.4. The daily values of AE varied from 0.1 to 2.1 with frequency distribution mode of 1.6. However, our results indicate that AOD measurements using sun photometers from ground-based AERONET network can be a useful tool to improve the monitoring of PM mass concentrations (correlation coefficient obtained equals 0.49). Further studies on correlations AOD-PM_{10} will include scaling of the AOD with the meteorological parameters, as meteorology day-to-day variations influence both ground-level and columnar aerosol properties.

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