

THE ROLE OF REGIONAL ATMOSPHERIC STABILITY IN HIGH-PM₁₀ CONCENTRATION EPISODES IN MIERCUREA CIUC (HARGHITA)

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Abstract: The conditions of accumulation of the particulate matter with diameter $<10\mu\text{m}$ - PM₁₀ in 2 episodes of thermal inversion from the Ciuc Depression were studied, with focus on the intradiurnal variation. The data taken from the regional monitoring station of the air quality from the Jigodin-Ciuc have been used. In accordance to the observations made in the Venice-Mestre area, when the intradiurnal variation of PM₁₀ values is greater than the daily average, static stability has an important role in the accumulation of PM₁₀, without having a linear behaviour. A relative increase of the static stability in Brunt Väisälä terms is enough to produce increases over the daily limit values ($50\mu\text{g}/\text{m}^3$) of the PM₁₀. The diurnal behaviour is highlighted by a maximum, respectively a minimum value; the maximum value occurs especially in the late afternoon and in the early evening, and the minimum values occur especially in the early morning. If we report the maximum and minimum values to the diurnal cycle of the atmospheric pressure variation, we can notice that these values fit to the time limit periods between them. If we strictly refer to the distribution of the minimum and maximum values, as it's well-known, taking into account the values of the static stability, we can notice that the increased values of the static stability induce maximum values of PM₁₀, while its low values determine an increase of the frequency of minimum diurnal values of PM₁₀.

Keywords: PM₁₀, Static stability, Brunt-Väisälä frequency, potential temperature, high concentration episodes, aerosol chemistry and geochemistry

1. INTRODUCTION

Particulate matter (PM) is one of the most important atmospheric pollutants in the European countries (Pateraki, et al., 2012), and present the most frequently exceeded of normative. A particular case represents the stable boundary layer conditions and is long known for causing increasing concentration of pollutant concentrations (Peringotti, et al., 2007). This value very frequently exceeds of the thresholds of PM₁₀ concentrations set by the EU air quality Directive 2008/50/EC (Jimmink, et al., 2010) has set target values and limit values for PM₁₀ concentrations in ambient air: the annual average should not exceed $40\mu\text{g}/\text{m}^3$ and the daily averages $50\mu\text{g}/\text{m}^3$ on more than 35 days per year.

The same behaviour was also observed in the urban site of Venice-Mestre in such stable boundary conditions, when the daily average reaching 100-150 $\mu\text{g}/\text{m}^2$ in just 4-6 hour (Peringotti, et al., 2007).

Despite of the increasing number of recent studies around the world (e.g. Kukkonen, et al., 2005; Viana, et al., 2003; Pateraki, et al., 2008; Vecchi, et al., 2007; Perrino, et al., 2008; Rodríguez, et al., 2008; Stefan, et al., 2010; Tai, et al., 2010; Dayan, et al., 2011; Pearce, et al., 2011), the understanding of the connection between temperature, relative humidity, wind and atmospheric circulation with particulate matter, is still not clear (Pateraki, et al., 2012).

As a result of the above we have found that similarly to the Venice-Mestre area in the Po plain, when the variation of the PM₁₀ concentration over the course of one day is often the same as, or higher than the daily average values (Rossa, et al., 2007), static stability plays a role in the accumulation of PM₁₀, but this does not appear to be a linear connection (Barnpadimos, et al., 2011), neither with the absolute values, nor with the increase of the PM₁₀ concentration.

The aim of this paper is to study the daily cycle of the PM₁₀ accumulation mentioned above considering the causing criterions of the temperature inversion conditions, first of all the stable stratification of the surface air layer with negative or zero gradients (Apăvăloaie, et al., 1988) and the different surface temperature.

In this study we focus on the relation between temperature inversion (in static stability conditions) and PM₁₀ concentration evolution at the Miercurea Ciuc. In this situation the air close to the (ground) surface is colder and heavier than the one related to particles, as a consequence reduced vertical mixing occurs between the air layers. The thermal inversion is associated to one extreme stability, and appears after cold nights with clear skies when near the ground surface there was not time for warming up, while the upper layers are already warmed by solar radiation (Fritz, et al., 2008). The temperature inversion conditions may lead to extremely high PM concentrations, especially during the cold season (Kukkonen, et al., 2005). In these conditions we would like to test the mechanism proposed by Rossa et al., 2007 which presents a daily emission cycle for the interpretation of the diurnal cycle of PM₁₀ concentration in anticyclonic conditions.

2. MATERIAL AND METHODS

2.1. Study area

The thermal profile (sampling site) is situated in Jigodin suburban area of the Miercurea Ciuc town (latitude 46°21'0''N, longitude 25°48'0''E).

The geographical position of the Ciuc depression - situated mostly in the shelter of the orographic dam determined by the limitrophic mountain frame of the Eastern Carpathians - is similar to a depresional groove in which thermal inversions are remarkable (Bogdan, & Niculescu 2004). Formation of fog and thermal inversion is characteristic especially during winter (Kristó, 1994).

In the Ciuc Depression, the average temperature is just with 0.2 grades smaller than on the limitrophic mountains with the height of 1800 m latitude, but with 1-2 grades smaller than on the slopes with 1200-1400 m, which stays warmer (Bogdan, & Niculescu, 2004).

Because of this fact the inversion can be very intense, and it may occupy the whole topoclimatic space, having a width of 500-600 m. On the other hand in case of polar air invasion determined by Eastern European anticyclone this may expand up to 1200 m (Bogdan & Niculescu, 2004). The main wind direction is westerly and north-westerly (Kristó,

1994).

2.2. Sampling

Atmospheric particles PM₁₀ were sampled with an Automatic analyzer LSPM₁₀ equipped with PM₁₀ and PM_{2.5} impactors, and Low volume gravimetric sampler for PM₁₀/PM_{2.5} - lead analysis (FOX Pump and Sentinel). PM₁₀ daily data were provided in continuous basis by the nearby station, which is included in the national monitoring network of air quality - RNMCA. The duration of the experimental campaign was 24 days (2 inversion periods), covering two periods with atmospheric stability, one from 2007-2008 and other from 2011.

The air temperature were sampled by a TS Thermometer sensor with measuring range between -30°C and +50°C installed at 2 m meter above the ground, and the pressure with an ORION (Mod BP-S) measuring sensor.

The meteorological data that we have measured at two points in Miercurea Ciuc (662m altitude) and in Jigodin Bath (717m altitude). As we already have presented in the introductory part of this paper, the PM₁₀ is one of the most widespread pollution especially in urban areas. In the Ciuc basin the most significant pollution is as well the PM₁₀ in spite of the fact that it is an agrarian region with wide forests. In our situation, in case of a stable stratification the flow of background layer cannot be determined owing to the fact that the consolidated uniformly stratified free flow waves extend to the convection field of the whole basin. Since the extension of the stable stratified flows is limited with a z upper limit which is the height of the ascending inversion, the stability of the flow can be described making use of frequency Brunt-Väisälä N^2 , which present at zero the most instable stratification (Britter, et al., 1981).

$$N^2 = -\frac{g}{\rho_0} \frac{d\rho}{dz} \quad (1)$$

N - Brunt-Väisälä frequency (Hz)

$d\rho$ - the difference in density measured at two measuring stations (kg/m^3)

dz - elevation difference between the two measuring stations (m)

g - gravitational acceleration (m/s^2)

$\rho_0 \equiv \rho(z_0)$ - the density measured at the z_0 level.

The revised parametric method for the density of moist air (Picard, et al., 2008) were determinate from measurements of air pressure, temperature, humidity and (and in our case because is required a high accuracy) carbon dioxide concentration using an equation (Giacomo, 1982., Davis, 1992)

recommended by the "Comité International des Poids et Mesures" (CIPM) derived by Giacomo and modified by Davis.

By means of the potential temperature, θ , as well as of the potential temperature gradient thereof, $\delta\theta/\delta z$, one can confirm the atmospheric stability. The sense of the potential temperature gradient determines the nature of the boundary layer stratification. If $\delta\theta/\delta z > 0$, the stratification is stable, if the gradient value is negative it is unstable, while in case of the existence of the condition $\delta\theta/\delta z = 0$, one can speak of a neutral stratification of the boundary layer (Hanna, et al., 1982).

$$\theta = T \left(\frac{P_0}{P} \right)^{\frac{R}{C_p}} \quad (2)$$

θ - potential temperature °C

T - current absolute temperature (K)

P - pressure at level "h"

P_0 - pressure at the reference surface

R - gas constant (J/kmol.K)

C_p - specific heat capacity at a constant pressure (J/kmol.K).

3. RESULTS AND DISCUSSIONS

High concentration episodes were analyzed in 2 inversion periods 12.21.2007 - 01.01.2008 and

01.25.2011 – 02.05.2011, when a stable high pressure system persisted for several days. In the figure 1 and 2, the PM_{10} concentration evolution for these periods with values below $50\mu g/m^3$ increase over the next days to $413.20\mu g/m^3$ in 26 December 2007 and 210.419 in 03th February 2011. In 01 January 2008 and 05 February 2011 the synoptic conditions changed and brought the episode to an end.

Identical to the situation described by Rossa et al., 2007, one very obvious feature of this evolution is the pronounced diurnal cycle, both of the PM_{10} concentration and the PBL (Planetary Boundary Layer) stability.

For example on the day of 23.12.2007 the PM_{10} concentration increased from 78.54 to $261.66 \mu g/m^2$ in just 9 hours; we found the same situation on the day of 26.01.2011 where the PM_{10} concentration increased from 65.015 to $181.069 \mu g/m^2$. The same behaviour was also observed in the urban site of Venice-Mestre in such stable boundary conditions, when the daily average has reached $100-150\mu g/m^2$ in just 4-6 hour (Peringotti, et al., 2007).

As a result of the above one finds that similarly to the Venice-Mestre area, when the variation of the PM_{10} concentration over the course of one day is often the same as, or higher than the daily average values (Rossa, et al., 2007), static

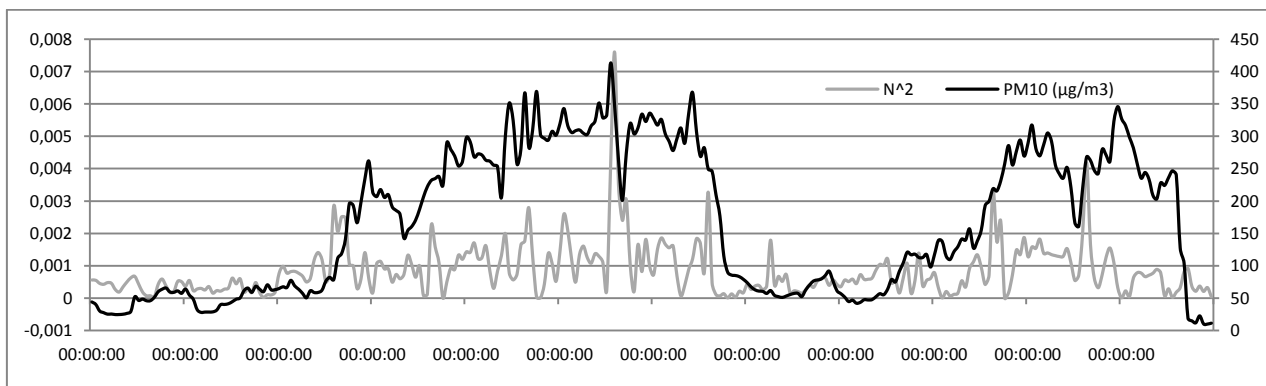


Figure 1. Hourly PM_{10} values (right axis) in relation to N^2 in terms of Brunt Väisälä (left axis) - for 12.21.2007 - 01.01.2008

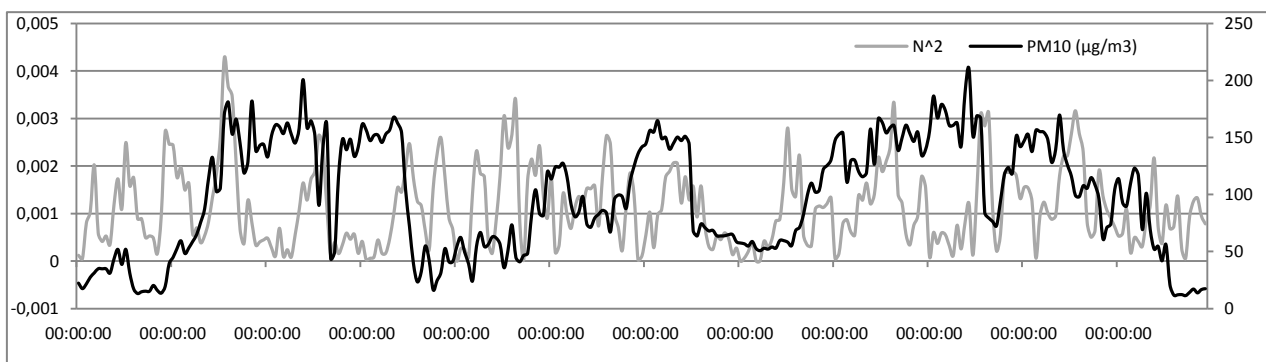


Figure 2. Hourly PM_{10} values (right axis) in relation to N^2 in terms of Brunt Väisälä (left axis) - for 01.25.2011 – 02.05.2011

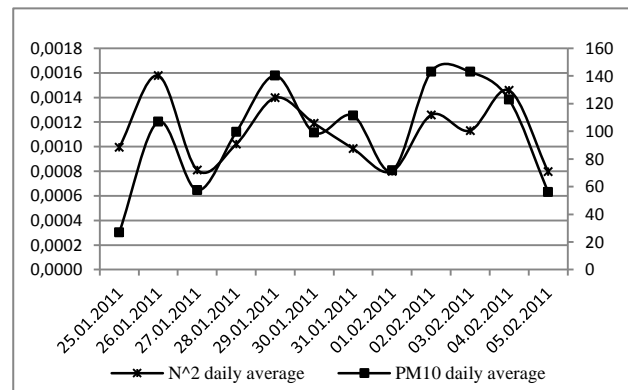
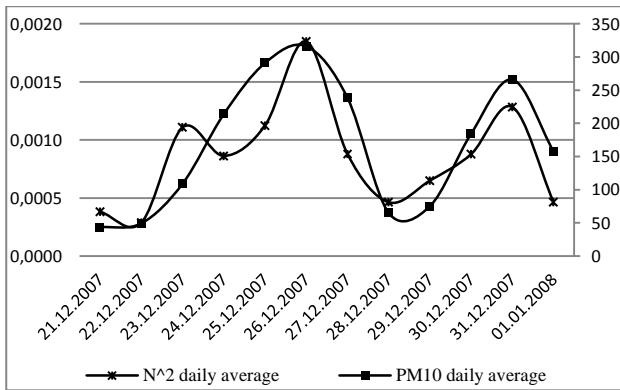


Figure 3. The left side - diurnal PM_{10} average concentration (right axis) in relation to diurnal average N^2 in terms of Brunt Väisälä (left axis) - for 12.21.2007 - 01.01.2008. The right side - diurnal PM_{10} average concentration (right axis) in relation to diurnal average N^2 in terms of Brunt Väisälä (left axis) - for 01.25.2011 - 02.05.2011

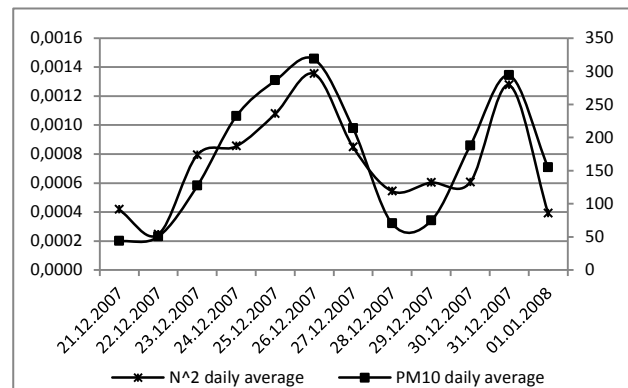
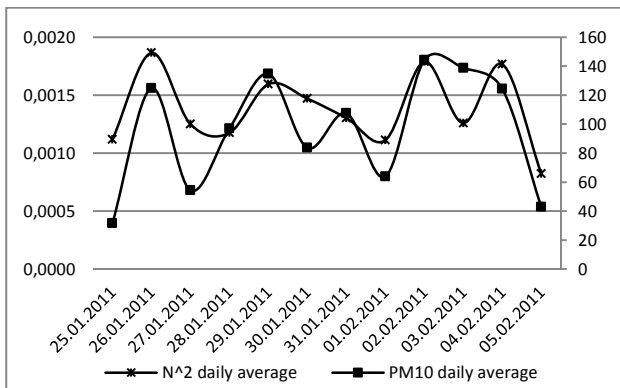


Figure 4. The left side - average of 12 hours (06-18 hours) PM_{10} concentration (right axis) in relation to the average of the 12 hours N^2 (left axis) - for 12.21.2007 - 01.01.2008. The right side - average of 12 hours (06-18 hours) PM_{10} concentration (right axis) in relation to the average of the 12 hours N^2 (left axis) 25.01.2011 - 02.05.2011

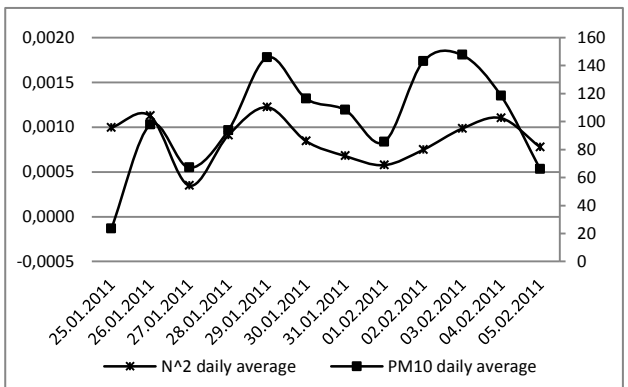
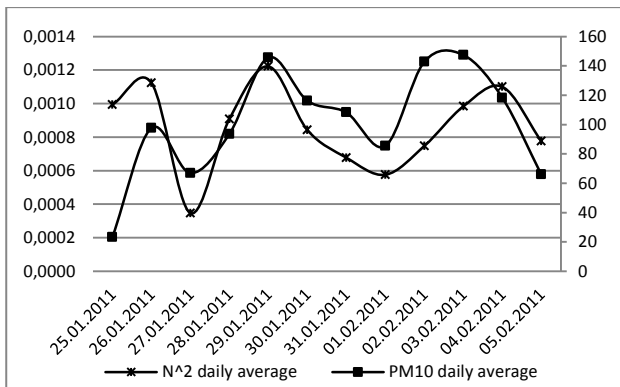


Figure 5. The left side - average of 12 hours (18-06 hours) PM_{10} concentration (right axis) in relation to the average of the 12 hours N^2 (left axis) - for 12.21.2007 - 01.01.2008. The right side - average of 12 hours (18-06 hours) PM_{10} concentration (right axis) in relation to the average of the 12 hours N^2 (left axis) 25.01.2011 - 02.05.2011

stability plays a role in the accumulation of PM_{10} , but this does not appear to be a linear connection, neither with the absolute values, nor with the increase of the PM_{10} concentration; a relatively moderate static stability, in terms of the Brunt-Väisälä frequency N^2 , is enough for the generation of a significant accumulation (Figs. 1 and 2).

All these observations were also tested with potential temperature. By analyzing figures 6 and 7, we will find that according to those established by Hanna, et al., (1982), for the increased values of the potential temperature there occurs a significant decrease of the values of PM_{10} concentration, under the conditions of decrease values of air stratification

stability. At the end of inversion period, when in the Depression there penetrates unstable cyclonic air masses, the PM_{10} values significantly decrease with the increase of potential temperature (Figs. 6 and 7).

The anticorrelation between the hourly value of the potential temperature and the concentration of PM_{10} is much more evident (Figs. 6 and 7) in comparison with the hourly values of N^2 . The analysis of daily average values by PM_{10} and potential temperature enhances much more this tendency (Figs. 8, 9 and 10).

The analysis of these values by Spearman's method (Hauke & Kossowski, 2011) from the studied periods supports the correlation mentioned above; the critical values for Spearman's rank correlation coefficient at a level of significance (α) 0.100, one tailed test at a number of 12 data pairs in each period is 0.406 (Gauthier, 2001).

In most of the cases the correlations between the average values of PM_{10} and N^2 and values of PM_{10} and θ are statistically significant. For the present case it is also interesting the sign of the correlation which supports the relationship between the static stability and potential temperature with the variation of PM_{10} (Tables 1 to 6). One may find weaker correlations related to the average daily values which are related to certain instability effects of air mass during different periods of the day.

The negative correlations of the average daily values between PM_{10} and θ caused by the destabilisation of air mass in different periods of the day in most cases in daytime (tables 3, 4 and 5).

By analyzing the static stability values in terms of Brunt - Väisälä and of the potential temperature θ , we were able to highlight, by means of two different methods, the conditions that lead to the creation of the thermal inversion phenomenon in the Depression of Ciuc, which implicitly causes the PM_{10} levels to exceed the daily average values of $50 \mu g/m^3$.

Taking into account the diurnal cycle of the atmospheric pressure (Dai, et al., 1999 In references list is Dai & Wang) one can notice a relative uniformity regarding the distribution of the

maximum PM_{10} values in the two inversion periods, with a minimum concentration value overnight (Figs. 11 and 12).

	PM_{10}	N^2	$\theta_{Jigodin} ^\circ C$
PM_{10}	1	0.895	-0.168
N^2	0.895	1	-0.350
$\theta_{Jigodin}$	-0.168	-0.350	1

Table 1. Spearman correlation coefficients of daily averages for the period 21.12.2007 - 01.01.2008

	PM_{10}	N^2	$\theta_{Jigodin} ^\circ C$
PM_{10}	1	0.783	-0.021
N^2	0.783	1	-0.364
$\theta_{Jigodin}$	-0.021	-0.364	1

Table 2. Spearman correlation coefficients for the mean values between 06-18 hours for the period 21.12.2007 - 01.01.2008

	PM_{10}	N^2	$\theta_{Jigodin} ^\circ C$
PM_{10}	1	0.902	-0.483
N^2	0.902	1	-0.503
$\theta_{Jigodin}$	-0.483	-0.503	1

Table 3. Spearman correlation coefficients for the mean values between 18-06 hours for the period 12.21.2007 - 01.01.2008

	PM_{10}	N^2	$\theta_{Jigodin} ^\circ C$
PM_{10}	1	0.577	-0.371
N^2	0.577	1	0.294
$\theta_{Jigodin}$	-0.371	0.294	1

Table 4. Spearman correlation coefficients of daily averages for the period 01.25.2011 – 02.05.2011

	PM_{10}	N^2	$\theta_{Jigodin} ^\circ C$
PM_{10}	1	0.650	-0.503
N^2	0.650	1	-0.650
$\theta_{Jigodin}$	-0.503	-0.650	1

Table 5. Spearman correlation coefficients for the mean values between 06-18 hours for the period 01.25.2011 – 02.05.2011

	PM_{10}	N^2	$\theta_{Jigodin} ^\circ C$
PM_{10}	1	0.769	-0.469
N^2	0.769	1	-0.531
$\theta_{Jigodin}$	-0.469	-0.531	1

Table 6. Spearman correlation coefficients for the mean values between 18-06 hours for the period 01.25.2011 – 02.05.2011

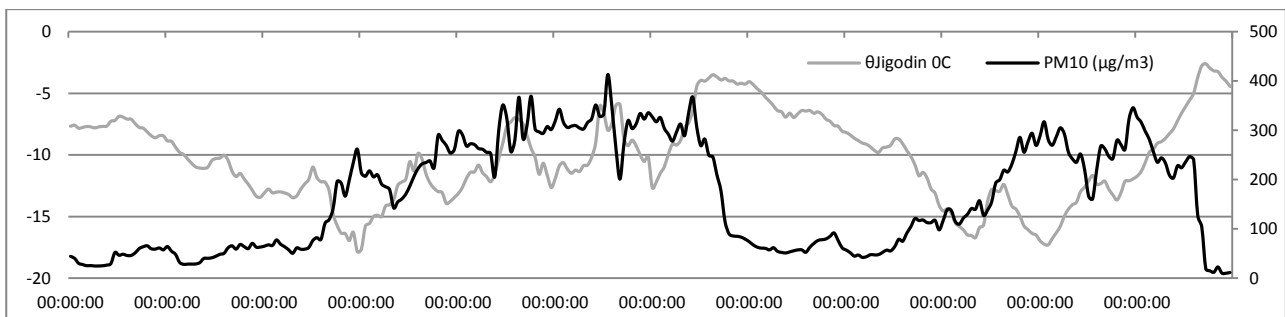


Figure 6. Hourly PM_{10} values (right axis) in relation to $\theta_{Jigodin} ^\circ C$ - potential temperature (left axis) - for the period 12.21.2007 - 01.01.2008

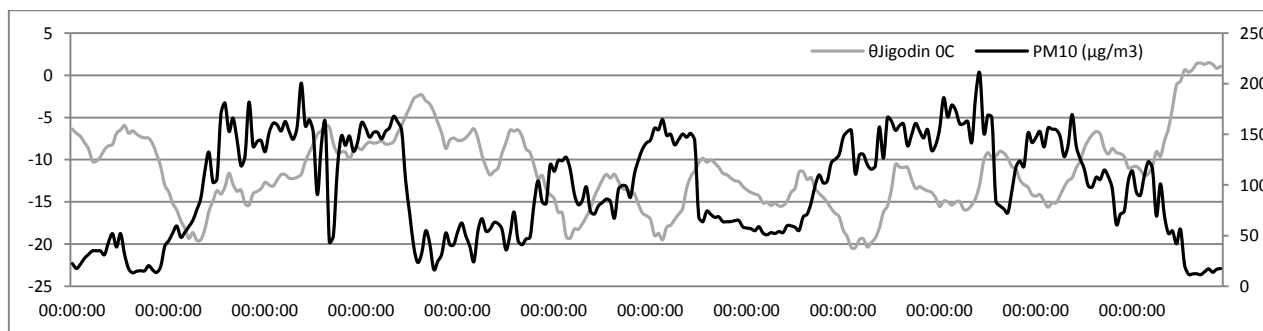


Figure 7. Hourly PM₁₀ values (right axis) in relation to $\theta_{Jigodin}^0C$ - potential temperature (left axis) - for 01.25.2011 – 02.05.2011

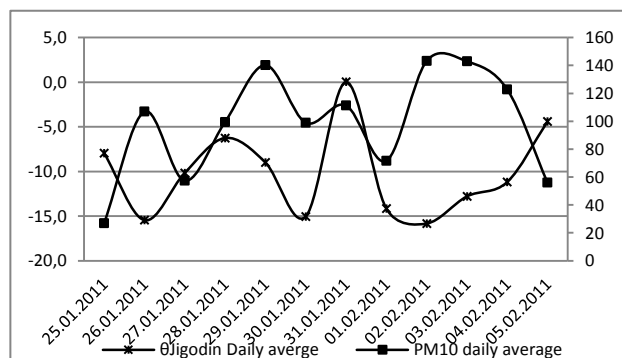
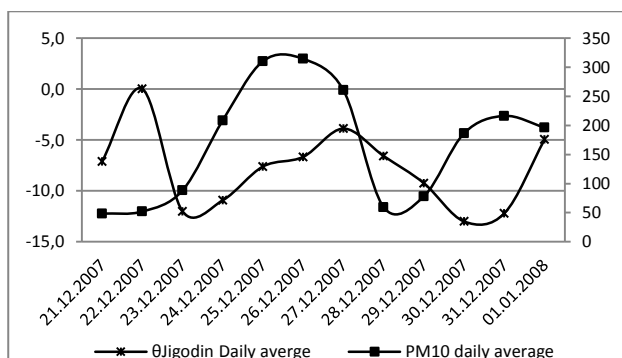


Figure 8. The left side - diurnal PM₁₀ average concentration (right axis) in relation to diurnal average of $\theta_{Jigodin}^0C$ (left axis) - for the period 21.12.2007 - 01.01.2008. The right side diurnal PM₁₀ average concentration (right axis) in relation to diurnal $\theta_{Jigodin}$ - potential temperature average (left axis) - for the period 01.25.2011 – 02.05.2011

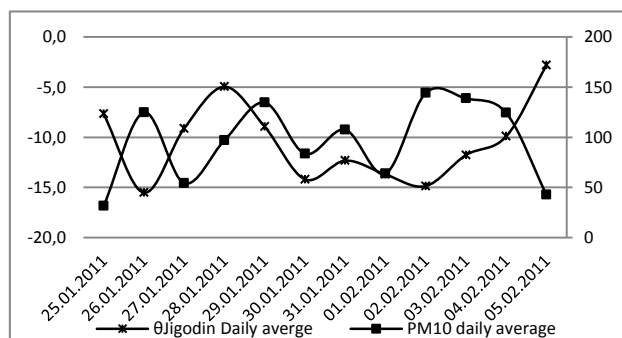
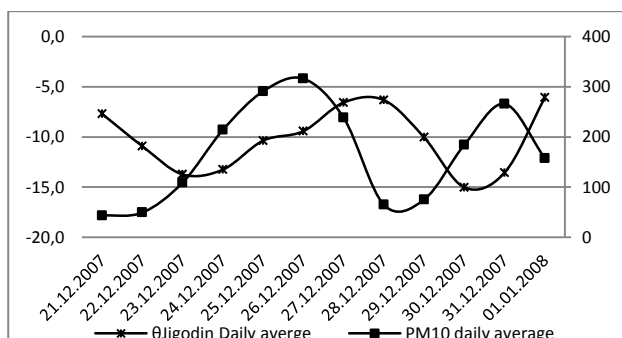


Figure 9. The left side - average of 12 hours (06-18 hours) PM₁₀ concentration (right axis) in relation to the average of the 12 hours $\theta_{Jigodin}$ (left axis) - for the period 21.12.2007 - 01.01.2008. The right side - average of 12 hours (06-18 hours) PM₁₀ concentration (right axis) in relation to the average of the 12 hours $\theta_{Jigodin}^0C$ (left axis) for the period 25.01.2011 - 02.05.2011

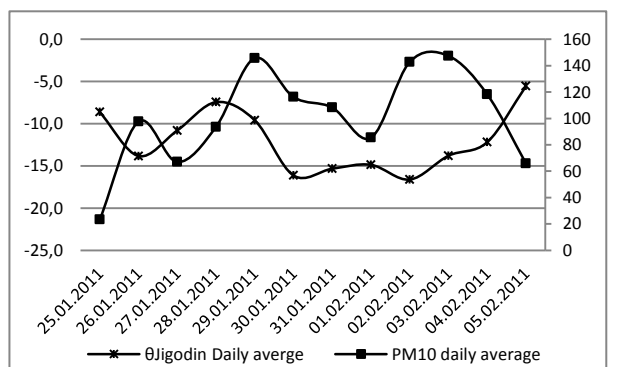
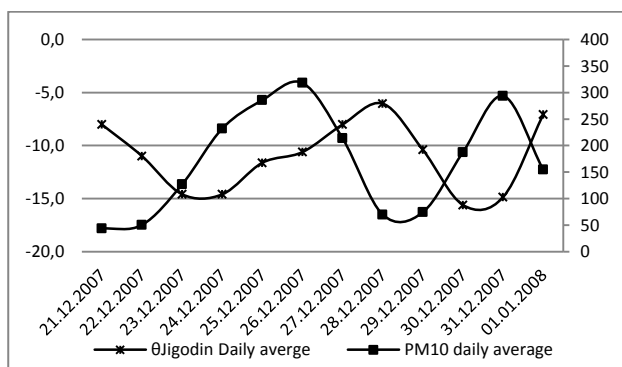


Figure 10. The left side - average of 12 hours (18-06 hours) PM₁₀ concentration (right axis) in relation to the average of the 12 hours $\theta_{Jigodin}$ (left axis) - for the period 21.12.2007 - 01.01.2008. The right side - average of 12 hours (18-06 hours) PM₁₀ concentration (right axis) in relation to the average of the 12 hours $\theta_{Jigodin}^0C$ (left axis) for the period 25.01.2011 - 02.05.2011

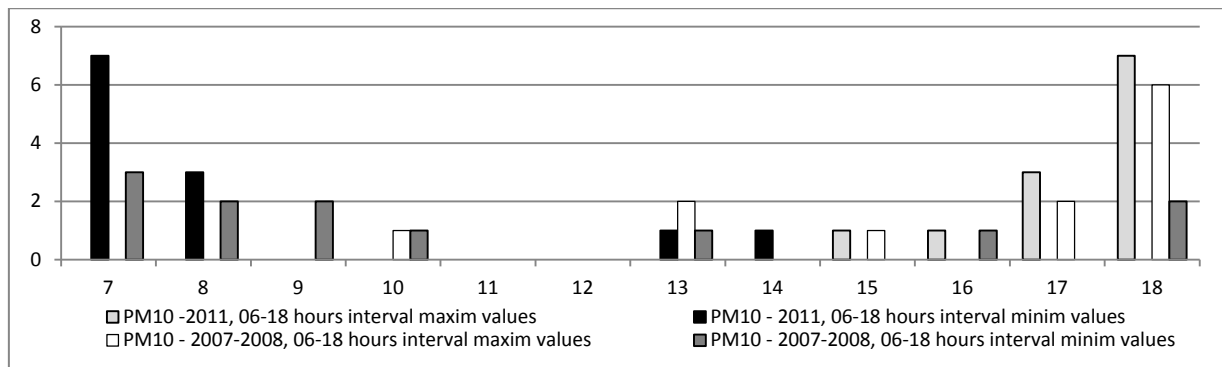


Figure 11 - Twelve hour's distribution (06-18) of minim and maxim values in the two studied high atmospheric stability episode (frequency is represented on the ordinate; in the abscissa are shown the hours of daytime)

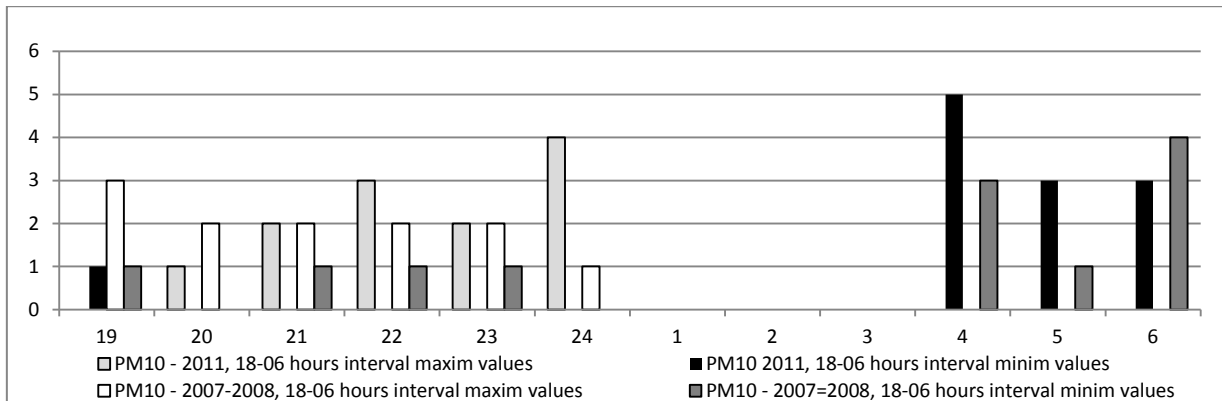


Figure 12 - Twelve hour's distribution (18-06) of minim and maxim values in two studied high atmospheric stability episode (frequency is represented on the ordinate; in the abscissa are shown the hours of night-time)

Meanwhile the minimum values, contrary to those observed in the Po valley, are distributed much less uniformly throughout the day (Rossa, et al., 2007); one peaks were observed, mostly during the morning hours (Figs. 11 and 12).

In the 2 examined inversion periods, the average level of the maximum and minimum variations (amplitude) of the PM_{10} in different periods of the day is of $269.52 \mu g/m^3$, which are, of course, dependent on the peak concentration and the static stability of the air; the daily peak concentration reaches a hourly value of over $400 \mu g/m^3$, which represents an excess that is more than eight-fold.

The peak concentration occurs in the late afternoon and in the evening hours and it subsequently decreases during the night (tables 11 and 12), when the emissions are also significantly reduced and the deposition and chemistry of this pollutant probably takes a downturn, as well.

This generic, non-linear, late-night decrease takes place while the stability of the boundary layer continues to increase (Rossa, et al., 2007).

From the analysis of the figures 1 and 2 respective figures 6 and 7 at the first sight is difficult to talk about correlation between PM_{10} concentration and N^2 (static stability).

This is due to the fact that the response of the PM_{10} concentration arrives after some time the stability establishes. The diurnal average values in the 2 studied periods, a positive correlation can be observed, which sets in with the increase of the stability and shows a trend of increase in the levels of PM_{10} (Fig. 3). The positive correlation is evident in case of 12 hour intervals of 06-18 hours (Fig. 4) and nights of 18-06 hours (Fig. 5). The positive correlations during night are more enhanced due to the stability of air mass (Vecchi, et al., 2007) +0.902 for the period 21.12.2007 – 01.01.2008 in comparison with the one during the day +0.783; when a light destabilisation is possible even in case of relatively strong anticyclonic period, because of the rise of temperature. The same situation was observed for the period of 01.25.2011 – 02.05.2011, when the correlation value during the 18-17 hourly period is higher +0.769 than in the daytime +0.650 (Tables 5 and 6). Daytime the PM_{10} concentration may be higher than during the night, which is caused by the particles resuspension, caused by the car traffic and domestic wood combustion (Grange, et al., 2013).

If normally the early morning emission peak occurs under stable conditions, so that the concentration of PM_{10} continues to increase (Janhäll, et al., 2006); in the Ciuc Depression this is

counterbalanced by a significant destabilization of the boundary layer in the morning hours (Peringotti, et al., 2007) which decrease the PM₁₀ concentration (Figs. 11 and 12).

The minimum concentration values often occur early in the morning, when the emissions are at a lower level and the stability is at its minimum (Wang, et al., 2009). The early morning low concentration period is also due to the low levels of domestic wood combustion (Grange, et al., 2013) and the significant decrease in traffic (Gehrig et al., 2004). The most powerful and sharpest PM₁₀ concentration values build up when the emissions due to evening traffic (Kakoli, & Gupta, 2006), including resuspension (Amato, et al., 2009) and other sources, coincide with the stabilization of the boundary layer and are not thinned during the vertical mixing process (Rossa, et al., 2007). All this occurs under a second progressive daily pressure cycle.

Overall, the peak concentrations of the minimum values develop under increasing air pressure, while the downturn occurs during the descending period of the air pressure, when the destabilization of the air is increasing. The exceeded concentrations can be observed outspokenly during inversions.

This mechanism does not account for aerosol chemistry and geochemistry, which plays an important role in the production of secondary aerosol PM (Miracolo, et al., 2011; Rossa, et al., 2007). Most PM₁₀ peaks, however, occur in moderately stable condition (Figs. 1 and 2), but there are several cases when one can observe minimum values in case of high values of static stability. This is in agreement with the conclusions of P07 who claim that the diurnal PM₁₀ cycle cannot be explained with atmospheric parameters alone but need to take into account the emission cycle. (Peringotti, et al., 2007).

4. CONCLUSION

The diurnal PM₁₀ concentration in 2 pollution episodes that occurred in stable conditions in the period of 12.12.2007 - 01.01 2008 and 25.01.2011 – 02.05.2011 was analyzed. In such regime we found that the intra diurnal PM₁₀ concentration variation can be much stronger than day to day variation of daily averages. For example in the day of 26.12.2007 the PM₁₀ concentration increases from 58.874 to 196.018µg/m³ in just 9 hour. A lot of features found by Peringotti, et al., (2007) and Rossa, et al., (2007) could be recognized in the studied inversions periods from Ciuc Depression.

Yet, until now, for the Ciuc Depression and

other depressions where it was noticed the phenomenon of thermal inversion, there were not made determinations of the conditions which determine this phenomenon, and which practically represent the basis of air pollution with particulate matter and other chemical compounds. Thus, in the whole county of Harghita, there was installed a single background station of monitoring the air quality, on the grounds that there are no major pollutants. In reality, as we have shown in the present work, there exists a real danger for the health of the population in these areas. The acknowledgement, respectively the extension of researches of the diurnal cycle of PM₁₀ is extremely necessary in order to decrease the exposure to this type of pollutant.

In both periods of inversion which have been studied in winter 2007, respectively 2011, static stability does play a role in the accumulation of PM₁₀, but there seem not to be a linear relation neither with the maximal values or with the increase of PM₁₀; a relatively moderate static stability seems to be sufficient to allow significant accumulation, while marked decrease is observed also in strongly stable conditions (Rossa, et al 2007);

The diurnal behaviour is highlighted by a maximum, respectively a minimum value; the maximum value occurs especially in the late afternoon or in the evening, and the minimum values occur especially in the early morning.

The amplitude of the minimum and maximum values of the PM₁₀ concentration frequently depasses 100µg/m³ and obviously depends on the peak concentrations;

From this perspective, it is obvious that the diurnal cycle of PM₁₀ is extremely complex and cannot be synthesized only by the study of its intradiurnal variation, even in static stability conditions. All these depend to a great extent on the emissions cycle, and on the aerosols chemistry and geochemistry.

Acknowledgments

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