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Fast Track Article

Influence of ABL stability on the diurnal cycle of PM10 concentration: illustration of the potential of the new Veneto network of MW-radiometers and SODAR

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Abstract

Stable boundary layer conditions are long known for causing increasing concentration of pollutant. The region of the Po Valley has a particularly unfortunate topographical conformation which favours low wind and strong inversion situations, a meteorological characteristic that leads to so-called episodic conditions, especially during the cold season. ARPAV-CMT has recently installed a network of boundary layer profilers consisting of four passive microwave radiometers and four SODAR, funded in the framework of the DOCUP 2000–2006 project (DOCumento Unico di Programmazione) and of the Project 'Air pollution in the city of Padua'. This network is briefly presented and an illustration is given as to its potential to analyse the PM10 (particulate matter with diameter lower than 10 ?m) concentration evolution. An entire year of data, as well as for a high-concentration episode is analyzed. In particular, emphasis is given to the documentation of the diurnal cycle of PM10 in relation to the atmospheric stability. It was found that PM10 is strongly modulated by synoptic-scale forcing and exhibits a strong diurnal cycle, especially in synoptically undisturbed conditions, when PM10 can exhibit large variations in just a few hours. A conceptualization is proposed which accounts for the daily emission cycle for the interpretation of the diurnal cycle of PM10 concentrations in anticyclonic conditions. It pinpoints the fundamentally different accumulation and removal mechanisms of the particulate matter from the atmosphere in stable and unstable conditions, highlighting the importance of the atmospheric stability at the time of the evening emission peak during the cold season.

Zusammenfassung

Es ist bekannt, dass stabile Grenzschichtbedingungen zu erhöhten Schadstoffbelastungen führen können. Die Poebene hat eine besonders unglückliche topographische Beschaffenheit, die Schwachwindlagen mit starken Temperaturinversionen begünstigt; meteorologische Eigenschaften, die besonders in der kalten Jahreszeit zu sogenannten episodischen Bedingungen führen. ARPAV-CMT hat kürzlich ein Netzwerk von Grenzschichtprofilern installiert, das aus vier passiven Mikrowellenradiometern und aus vier SODAR-Geräten besteht. Die Finanzierung kam dabei vor allem aus dem DOCUP Projekt 2000–2006 (DOCumento Unico di Programmazione). Dieses Netzwerk wird vorgestellt und beispielhaft angewendet, um die PM10-Entwicklung für ein ganzes Jahr und für eine Episode mit hohen PM10-Konzentrationen zu analysieren. Insbesondere wird dabei die Beschreibung des PM10-Tagesgangs in Beziehung zu den Eigenschaften der atmosphärischen Stabilität gesetzt. Es wurde herausgefunden, dass PM10 stark von synoptisch-skaligem Antrieb moduliert wird und einen ausgeprägten Tagesgang besitzt, vor allem in synoptisch ungestörten Verhältnissen, wenn PM10 große Variationen in wenigen Stunden aufweisen kann. Ein Schema für die Interpretation des PM10-Tagesgangs während stabiler Hochdrucklagen wird vorgestellt, das den Tagesgang der Emissionen berücksichtigt. Es hält die fundamental verschiedenen PM10-Abbauprozesse in stabilen und instabilen Verhältnissen fest und unterstreicht die Wichtigkeit der atmosphärischen Stabilität zur Zeit des abendlichen Emissionsmaximums in der kalten Jahreszeit.

1 Introduction

Compliance with the Council Directive 1999/30/EC for concentrations of PM10 is an increasing challenge for many European cities. As a matter of fact, the dispersion of pollutants is strongly subject to meteorological action so that a region's burden not to exceed PM10 concentration limits depends on its climatology. The region of the Po Valley has a particularly unfortunate topographical conformation which favours low wind and strong inversion situations, a meteorological characteristic that leads to so-called episodic conditions, especially during the cold season (e.g. KUKKONEN et al., 2005).

In order to respond to these requirements, the Meteorological Centre of Teolo (CMT) of the regional agency of environmental protection of the region Veneto (ARPAV), has recently installed a boundary layer profiler network (Fig. 1), which measures wind and temperature via remote sensing techniques and consists of one HATPRO microwave radiometer, three MTP5-HE microwave temperature profilers, and four PCS-2000 SODAR. The network is the first of its kind in Italy and its

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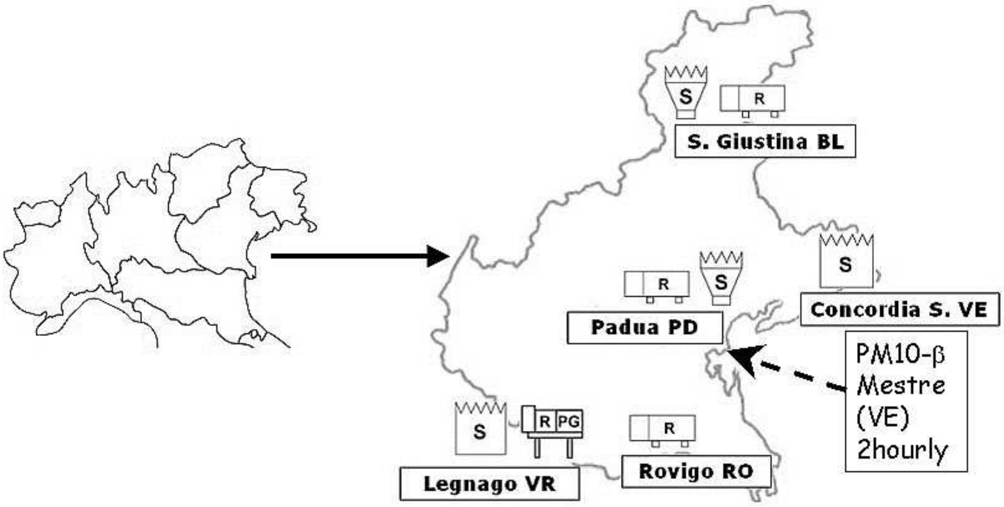

Figure 1: Northern Italy (left panel) and location of the ABL network (right panel). The acronyms stand for S = PCS2000 SODAR, R = MTP5-HE Radiometer, RPG = HATPRO RPG Radiometer). Legnago is located in the flat countryside in the south of the Province of Verona (VR); Rovigo is in the city centre in the Province of Rovigo (RO), Concordia Sagittaria is in the country side in the north of the Province of Venezia (VE) and S. Giustina is in the main valley in the Province of Belluno; Padua is in the city center in the Province of Padua. The 2-hly PM10 measurement site (dashed arrow) is located in the city center of Mestre, in the Province of Venice.

principal applications are in the field of environmental emergency management as well as regional air-quality short time forecasting.

Inversions are tied to very stable conditions and inhibit vertical mixing. They are, therefore, long known for causing increasing concentration of pollutant (e.g. MILIONIS and DAVIES, 1994 and references therein). The presence of a temperature inversion or stable stratification at ground level causes the response of PM10 concentrations to the morning traffic in a much more pronounced way (JANHÄLL et al., 2006), diurnal behaviour of PM10 and PM2.5 are influenced by temperature inversions on episodic basis.

Wind is another very important factor for the dispersion of pollutants by generating mechanical turbulence. A simple surface wind measurement or profile allows a qualitative estimate of turbulent activity, while assessment of turbulent fluxes, which govern the evolution of turbulence (PIRINGER et al., 2007), would permit a more quantitative estimation. The general capability of ground-based remote-sensing techniques for the direct estimation of turbulent fluxes with SODAR is reported in Engelbart et al. (2007, this volume). However, the demands for the data quality and signal processing are very high, so that an operational application is still far away.

In this paper an illustration of the potential of the Veneto ABL profiler network is presented as data from the Padua MTP5-HE radiometer and PCS-2000 SODAR are used to analyze the PM10 concentration evolution. This is done for an entire year, as well as for a high-concentration episode (19–26 December 2005). In particular, emphasis is given to the documentation of the diurnal cycle of PM10 in relation to the presence and characteristics of thermal stability.

2 Data set

SODAR (S in Fig. 1) is an established technology for determining the vertical wind profile with high temporal resolution. The instruments installed in Veneto are two PCS2000-24 (Padua and S. Giustina) and two PCS2000-64 (Legnago and Concordia S.), manufactured by Metek. The nominal measurement accuracy ranges from 0.1 to 0.3 m/s for horizontal wind speed and from 1° to 5° for horizontal wind direction. The wind profile measured by the Padua SODAR has a time resolution of 15 minutes and a vertical resolution of 20 m with data availability in the first year of deployment of 75 % for the first level (40 m), decreasing rapidly with height with to only 24 % at 200 m.

The radiometer MTP5-HE (R in Fig.1), manufactured by Attex and distributed by Kipp & Zonen, measures the temperature up to 1000m with a vertical resolution of 50 m and a time resolution of 5 min. The HATPRO (RPG in Fig. 1), manufactured by Radiometer Physics GmbH measures temperature and humidity up to 10000 m with a vertical resolution of 200 m, integrated water vapour, and liquid water content. An additional vertical scan is present, so that also temperature profile with a resolution of about 50–75 m is available every 20 minutes up to 2000 m.

The availability of MTP5-HE radiometer data in the first year of deployment for Padua is very high reaching 97 %. Nominal accuracy is 0.3–0.4 K for adiabatic and 0.8–1.2 K for inversion conditions, while a good agreement within 1 K with the closest soundings in homogeneous atmospheric conditions (FERRARIO et al., 2006 and references therein). This result is in line with the generally good accuracy of the radiometer measurements of temperature profiles reported in the literature.

In the centre of Mestre (broken arrow in Fig. 1), a city close to Venice and about 25 km northeast of the Padua, bi-hourly measures PM10 concentrations were available. This data are measured by a ENVIRONNEMENT MP101M, using the method of beta-ray attenuation; the daily averages are validated through comparison with a near by gravimetric instrument.

3 Results

In order to illustrate the usefulness of the CMT boundary layer profiler for air quality issues, the data of the Padua MTP5-HE radiometer and the SODAR are analyzed in relation to PM10 concentrations observed at the Mestre site in two-hourly intervals (Fig. 1). Given the meteorological conditions and the climatology of the Po Valley it is believed that the ABL measurements of Padua are representative for the Mestre site (there is no two-hourly PM10 measurement site in Padua). For this analysis, both the radiometer and the SODAR data are averaged on the corresponding two-hour interval preceding the validity time. For the characterization of the thermal structure of the boundary layer the potential temperature difference between 200 m and ground level ($\Delta\theta_{200}$) was used. Note that this is a good indicator for the diurnal cycle of the atmospheric boundary layer (ABL), especially in undisturbed synoptic conditions. The influence of transport and mechanical turbulence is estimated by the vector average of the wind at 100 m above ground (v_{100}). Other intuitively relevant variables like temperature inversion strength and depth were tested but did not yield better results (not shown); moreover such parameters are only defined when temperature inversions are present, whilst a continuous series of data was necessary for this analysis. The choice of v_{100} as measure for the wind was dictated by the scarce data availability above 100 m, making it difficult to evaluate a dynamical measure like strength of nocturnal boundary layer jet.

3.1 Statistical analysis

The time series of PM10 concentrations (not shown) clearly exhibit a diurnal cycle, a fact that is well reflected in the Fourier spectrum (not shown). Therefore the time series of all parameters considered are decomposed into a 24 h centred running average and a high-frequency part. The first gives every two hours a value that is the mean of the previous 10 hours, the present and the following 12 hours; the second is simply the difference between the full signal and the centred running daily average and should reflect the intra-diurnal variability (diurnal cycle of PM10 concentration and of the ABL).

Table 1: Correlation coefficients of PM10 vs potential temperature difference 200 m surface ($\Delta\theta_{200}$) and wind velocity at 100 m (v_{100}) for the two-hourly (original), 24 h average, and to the two-hourly high-frequency part of the time series, for the entire year and for the December episode (6 days).

R	**1 year**		**from h 14 of 20th to h 12 of 26th December 2005**	
PM10 vs	$\Delta\theta_{200}$	V_{100}	$\Delta\theta_{200}$	V_{100}
2 h original	0.37	–0.29	0.48	–0.18
24 h average	0.52	–0.42	0.84	n. s.
2 h high frequency	0.38	0.00	0.49	–0.24

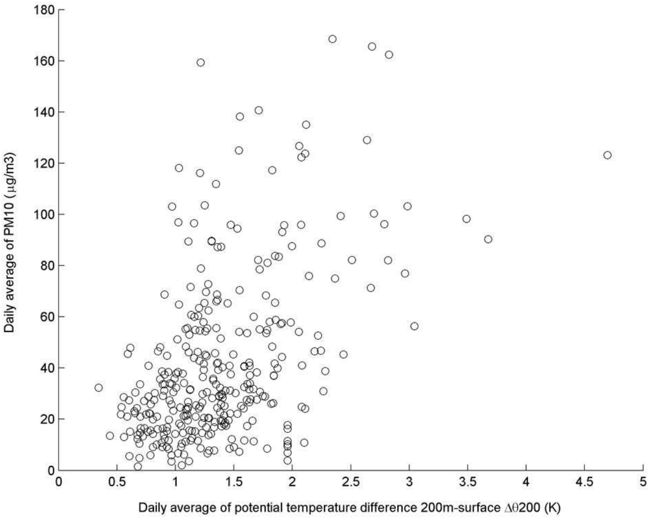

Figure 2: Scatter plot of 1 year data of PM10 vs $\Delta\theta_{200}$ (daily average).

Statistical results are reported in Table 1, where the correlation coefficients are reported for the original bi-hourly signal (first row), for the daily average centred al 00 LT (second row, 364 data in one year) and the bi-hourly filtered signal (third row). All data reported have significance higher than 95 % regarding the t-test for correlation.

The scatter plot of the daily averaged PM10 and $\Delta\theta_{200}$ (Fig. 2) shows that the PM10 concentration tends to be higher with more stratified ABL conditions. The first 2 columns of Table 1 report the correlation values between the PM10 concentration and $\Delta\theta_{200}$ and v_{100} for the entire year. For 200 the value 0.37 increases to 0.52 for daily averages (Fig. 2), indicating that the variability due to synoptic disturbances is an important modulator of PM10 concentration, coherent with values found by SCHÄFER et al. (2007) for the mixing height in Hanover. The high frequency part of the series features a lower

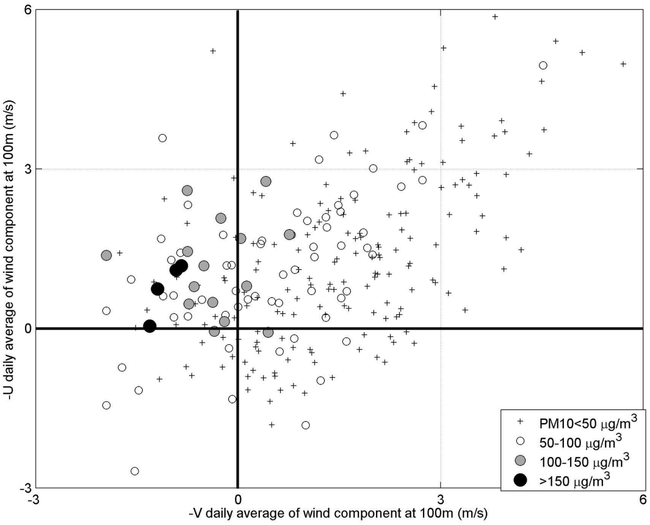

Figure 3: Daily wind distribution at 100 m for various PM10 concentration ranges.

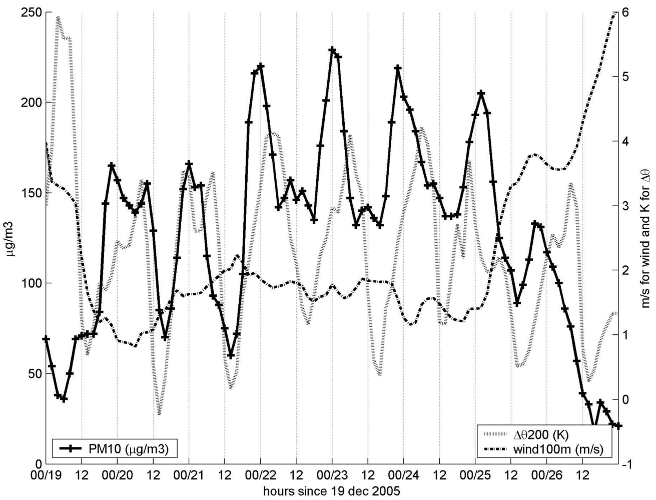

Figure 4: Pollution episode of 19–26 December 2005, with 2-hly data PM10 (solid line in μ/m^3, left axis), and $\Delta\theta_{200}$ (dotted line in K) and v$_{100}$ (dash-dotted line in m/s, both right axis).

correlation value of 0.38 as, over the period of the entire year, the synoptic variability is frequently disturbing the diurnal ABL cycle.

The dependence of the PM10 on wind is well known as advection of clean air and turbulent vertical mixing both reduce pollutant concentrations. Correlations in Table 1 are, therefore, negative, but not very pronounced with values of –0.29 for the full time series. Figure 3 shows that the high-concentration days are all characterized by relatively low winds of less than about 3 m/s, and that the wind direction for such days is predominantly from the north-west and north. Lower concentrations frequently are tied to stronger winds coming from north-east to south-east, denoting advection of clean air from the sea. The 24 h running average shows indeed a better correlation of –0.42, which is coherent with what was found by SANSONE et al. (2006) for wind measured at 10m.The high-frequency part is completely uncorrelated (0.00), probably because especially in winter a clear diurnal wind signal, e.g. from sea breeze, is absent.

3.2 Description of the diurnal cycle of PM10 during an episode

A high-concentration episode, the strongest of the analyzed data set, occurred in Mestre in the Christmas week 2005 when a stable high pressure system persisted for several days. Figure 4 shows the PM10 concentration evolution for this period with values below 50 μ/m^3 at December 19 that increased over the next days to 150 μ/m3 around noon, and peaks over 200 μ/m^3 during the night. Eventually, on December 26 the synoptic conditions changed and brought the episode to an end. One very obvious feature of this evolution is the pronounced diurnal cycle, both of the PM10 concentration and the ABL stability.

In fact, a correlation of the pollutants with 200 for the most stable part of the period from midday 20 to midday December 26 (Table1, third and fourth columns) yields values of 0.84 for the daily averages. As can be seen in Fig. 5a this high correlation is due to the accumulation of PM10 when the ABL became more stable from day to day, and the dilution of PM10 with the synoptic disturbance which, in addition to the wind, destabilized the ABL (indeed, the anti-correlation with wind is also high as –0.74, but not over at the 95 % of significance for the t-test). For the high-frequency part, on the other hand, the correlation value for this period is 0.49, i.e. higher than for the entire year but still modest. Figure 5b shows that the main contribution to the correlation between the high-frequency time series of PM10 concentration and ($\Delta\theta_{200}$ is due to the accumulation of PM10 when the ABL becomes more stable in the late afternoon, suggesting a direct cause-effect. Closer inspection, however, reveals that the PM10 concentration is not in phase with 200 for the entire day, as actually the PM10 concentration is increasing after the traffic peak (about 20 LT) but then decreasing during the night when the boundary layer is still further stabilizing. The strong increase during the first part of the night is probably related to the condensation of secondary particles (NH_4NO and organic matter) as suggested in PUTAUD et al. (2004) for other cities in the Po-Valley (Milan and Bologna). This hypothesis has been recently confirmed for the near-by city of Padua (TARGA, 2007). The following decrease is probably related mostly to normal processes of lateral dispersion, being this station a hot spot for pollution. The combination of these correlated and anti-correlated periods explains the modest coefficients found (Table 1). The wind during the event tends to be north-westerly. This is probably not identifying the source of pollution but is just the typical wind regime of

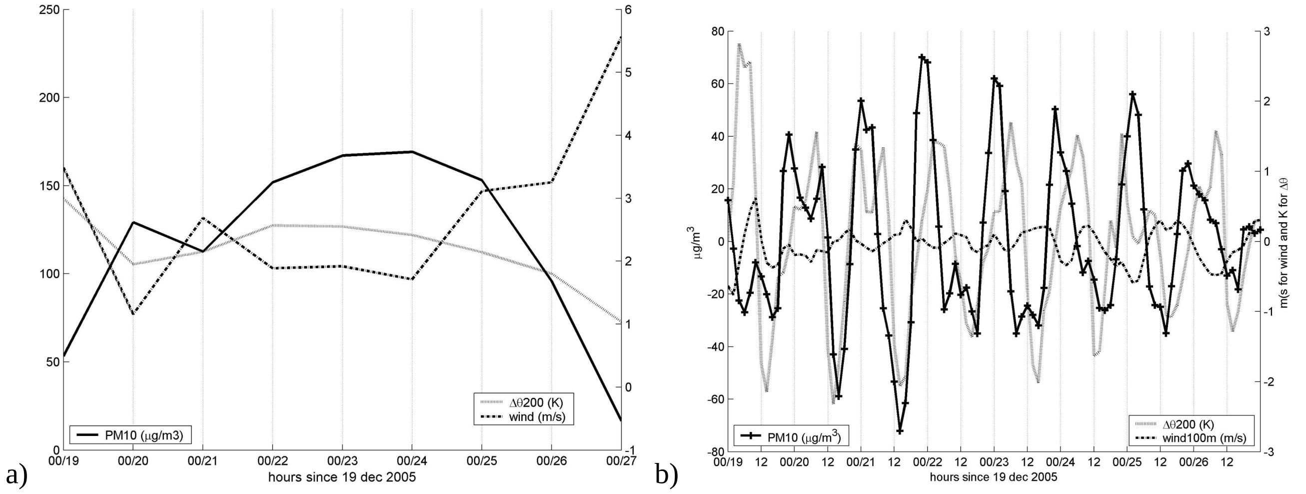

Figure 5: a) as Fig. 4 but for the 24h centered averages. b) as Fig. 4 but for the 2-hly high frequency part.

a very stable condition in a city close to the coast (in the SE direction), given that the sea surface temperature was about 10°C in those days, while there was a minimum of –1°C and a maximum of 8°C in the city.

For the considered episode, assuming a constant emission cycle with two peaks in the morning and evening rush hours, the time evolution of the PM10 concentrations (Fig. 4) can be conceptualized in the following way:

1. The peak concentration occurs at around midnight and subsequently decreases during the night when emissions are significantly reduced. This reduction takes place while the boundary layer stability is still increasing, which yields an anticorrelated behaviour.

2. The morning emission peak occurs in stable conditions so that the PM10 concentration increases (JANHÄLL et al., 2006), but is counteracted by the destabilization of the boundary layer during the morning hours. Consequently, the resulting concentration signature, which does not always show as a distinct maximum, is much lower compared to the peak of the night and occurs before noon.

3. The minimum concentration values are reached mid afternoon when the emissions are lower and stability is at its minimum.

4. The main and sharp build-up of the PM10 concentration values comes about when the evening peak emissions due to traffic and other sources are coincident with the stabilization of the boundary layer and therefore vertical mixing is reduced. Especially at low temperatures these precursors are likely to cause the formation of secondary PM10 that condensate particles from the gaseous phase (namely NH_4NO_3 and organic matter (PUTAUD et al., 2004).

In the episode under consideration the build-up in the evening hours is much more significant (100–150 μ/m^3 in 4–6 hours) than the day-to-day variations. Hereby, the peak concentrations appear to depend on the stability, consistent with the high correlation of the daily running averaged time series for the episode (0.84). Note that in this episode the day-to-day variations of PM10 does not follow a monotonous increase but, rather, exhibits a step in which the peak value of about 160 μ/m^3 sharply rises to 220 μ/m^3 in the evening of 21 December, when the boundary layer stability settles on a slightly higher level. These conditions persist for the following two days with similar peak and daily averaged concentration values. The nocturnal reduction of PM10, on the other hand, is more regular over the entire period reducing the concentrations by about 80 μ/m^3.

The analysis of this episode within the description above, suggests that the peak concentration values, given a constant emission cycle, depend on the stability of the boundary layer at the time of the evening emission peak. The more stable the conditions close to the ground the higher pollutant concentration which can be sustained by the emissions and consequent formation of secondary PM10, particularly near the sources. The night-time reduction testifies to the fact that such high levels of concentration are not sustained in the absence of sources. In this light, a fundamental difference between the cold and the warm season is that the evening emissions peak occurs in well mixed conditions in summer, i.e. before sunset, while in winter the pollutants are emitted into the stabilizing boundary layer. By the same token, any traffic limitation measures should be more effective during the stable conditions, starting from the late afternoon hours, than during daytime.

4 Summary and discussion

In this study a recently installed network of boundary layer profilers was presented, along with an attempt to understand the PM10 concentration evolution in terms of boundary layer evolution, assuming for PM10 a constant emission diurnal cycle. The main results of the activity that was performed mostly within the DOCUP project are:

- Eight profilers (four METEK-SODARs, three MTP5-HE radiometers, and one HATPRO radiometer) were successfully installed in the Italian region Veneto, and have been working reliably for almost two years now.
- PM10 concentrations are strongly modulated by synoptic-scale forcing, like frontal passages, precipitation and strong winds, so that the correlation coefficient between $\Delta\theta_{200}$ and PM10 is unexpectedly low over the entire year.
- PM10 exhibits a strong diurnal cycle, especially in synoptically undisturbed and low wind conditions, featuring concentration variations much stronger than day-to-day variations of daily averages (100–150 μ/m^3 in 4–6 hours in the evenings of 21–24 December, see Fig. 4).
- A schematic description of the diurnal PM10 cycle is proposed for anticyclonic conditions, which account for the complex interaction between stabilization of the atmosphere and emissions peak, leading to the formation of secondary PM10. Hereby, the late afternoon onset of the stabilization of the ABL and its overall stability appear to play a significant role in determining the peak and daily average PM10 concentration in synoptically undisturbed conditions.

In summary, boundary layer profilers, like radiometers and SODAR, help characterizing the state of the ABL. One striking result of this analysis is the unexpectedly low correlation between wind and static stability and PM10 concentration (Table 1, first row). As indicated above, synoptic variability is a strong modulator of PM10 in the atmosphere, as it can act to exchange or strongly modify the air mass under consideration, e.g. by precipitation (SANSONE et al., 2006). Another reason why the correlation of the daily running average data (Table 1, second row) is low over the entire year is the fact that the static stability and wind can act in distinct ways that lead to strong increase or decrease of PM10 concentration. For example, there can be strong static stability with low winds (as in the case study reported) favouring the accumulation of PM10 and a good correlation between $\Delta\theta_{200}$ and PM10. On the other hand, there can be an increase in stability with a strengthening of the winds which then act to reduce PM10 yielding an anti-correlated behaviour between $\Delta\theta_{200}$ and PM10 (e.g. for the period 15–19 February 2006 a correlation coefficient of 0.9 was found).

The case study approach based on which the conclusions are drawn is certainly a limitation, as is the qualitative consideration of role of weak winds for local transport and sources and sinks of PM10. Hence, future work will comprise stratifying the data set according to meteorological situations in which the main factors that influence PM10 concentration are similar, and a more thorough treatment of the important processes governing this pollutant. Also, in light of the recent progress reported in ENGELBART et al. (2007, this issue), the feasibility of direct determination of turbulent fluxes with the METEK-SODAR will be explored.

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