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ATMOSPHERIC ENVIRONMENT

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Characterising seasonal variations and spatial distribution of ambient PM10 and PM2.5 concentrations based on long-term Swiss monitoring data

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Received 9 December 2002; received in revised form 11 March 2003; accepted 17 March 2003

Abstract

Collocated parallel measurements of PM2.5 and PM10 were conducted at 7 sites in Switzerland since January 1998, constituting now one of the longest comparative data sets for PM2.5 and PM10 in Europe. The range of the long-term mean concentrations of PM2.5 was between 7.9 μg/m^3 at Chaumont and 24.4 μg/m^3 in Lugano. For the sites within the Swiss plateau this range narrows from 15.1 μg/m^3 at the rural site of Payerne to 20.8 μg/m^3 at the directly traffic exposed site of Bern. The long-term averages of the PM2.5/PM10 ratios of the daily values vary only from 0.75 to 0.76, with the exception of the traffic exposed site of Bern (0.59). The correlation between the daily values of PM2.5 and PM10 at all sites is generally high. For PM10, as well as for PM2.5 the highest concentrations are normally observed during wintertime. An exception is Chaumont (1140-m a.s.l.), which is often positioned above the inversion layer during wintertime and, therefore, has the lowest concentration during wintertime. A minimum of the PM2.5/PM10 ratio is often found during spring, probably due to the influence of relatively coarse biogenic particles. Though the sites have quite different exposition characteristics, the correlation of the daily values of PM2.5 and PM10 between the different sites of the Swiss plateau is very high, indicating a dominant influence of regional meteorology over local events and sources. The findings imply that from the point of view of an efficient use of financial and personal resources, the number of collocated PM2.5 measurements at PM10 sites in a monitoring network can be kept quite limited. The saved resources could rather be used to investigate other particle related parameters providing substantial new information (e.g. on particle sources, formation and effects) like PM1, particle number concentrations, morphology or chemical composition.

Keywords: Fine particles; Parallel measurement; Collocated measurement; PM monitoring; PM spatial distribution

1. Introduction

Epidemiological studies in Switzerland as well as in other countries have shown a significant impact of fine particles below 10 μm (PM10) on human health (AckermannLiebrich et al., 1997; BraunFahrlander et al., 1997; BUWAL, 1996; Dockery and Pope, 1994; Kunzli et al., 2000; Pope et al., 2002). Therefore, measurements of PM10, which is considered to represent the thoracic fraction of the ambient particles (ISO, 1995), have been performed within the Swiss National Monitoring Network (NABEL) already since 1997. However, PM10 is by far not a specific indicator for anthropogenic fine dust as it contains also considerable amounts of wind-blown natural mineral particles. This holds even true for the finer PM2.5 fraction, which is now widely measured in Europe and US and is considered to represent the

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1352-2310/03/$ - see front matter © 2003 Published by Elsevier Science Ltd.
doi:10.1016/S1352-2310(03)00221-8

alveolar fraction of the ambient particles (ISO, 1995). Due to the increasing interest for the finer particles (PM2.5) the measurement programme of the network has been extended to include PM2.5 measurements into the measurement programme at 7 sites already in 1998. The Directive 1999/30/EC (EU-Commission, 1999) asks member states of the European Union to perform and report PM2.5 measurements in addition to PM10. However, extended data sets of parallel PM2.5 and PM10 measurements are still lacking for Central Europe. Even worldwide long-term comparative data for PM10 and PM2.5 are hardly available. Reported measurements either focus on short events (Claiborn et al., 2000), are limited to urban areas (Li and Lin, 2002; Marcazzan et al., 2001), or were measured under pollution conditions, which are not representative for Europe (Qian et al., 2001). Therefore, the Swiss data set forms a unique data basis for investigating the temporal and spatial behaviour of PM2.5 compared to the well-known PM10. It includes meanwhile 4 years of parallel PM2.5 and PM10 data at various sites representing important exposition types of the population.

2. Measurement programme, methods and quality assurance

Table 1 lists the 7 sites where parallel PM2.5 and PM10 measurements have been performed and give information about the type of pollution exposition at each site. The map in Fig. 1 shows the geographical position of the sites within Switzerland. A detailed description of the sites has been published in (EMPA, 2000).

All particle samplings were conducted with high-volume-samplers Digitel DA 80. The set-up of the instrument, which is of widespread use in Europe, has been described in detail in a VDI guideline (VDI, 1996). The sampling inlets are operated at a flow of 30 m^3/h and meet the requirements of EN 12341 (CEN, 1998) for reference equivalency, as has been shown in an extended field study (UMEG, 1999). For PM2.5 measurements there is still no defined reference method for Europe. Ongoing, still unpublished field measurements performed within the scope of the European standardisation for PM2.5 show a satisfactory consistence of the PM2.5 sampling inlet used in this study on the Digitel instrument with the WINS impactor, which is used as reference sampler in the US for PM2.5 sampling and also with the German low-volume-sampler (Kleinfiltergerät). Glass fibre filters of the type Ederol 227/1/60 were used for particle collection.

The measurement uncertainty for the PM10 measurements has been quantified from collocated parallel measurements. It is ±10% (95% confidence interval for single daily values) in the concentration range 10–30 µg/m^3. The detection limit was determined from the standard deviation of field blanks to be 1 µg/m^3. Because the only difference between the applied method for PM10 and PM2.5 is the diameter of the nozzles in the sampling heads, the same measurement uncertainty can be assumed for the PM2.5 measurements.

3. Results

3.1. Mass concentration of PM10 and PM2.5 from 1998 to 2001

Table 2 gives an overview of the annual mean concentrations of PM10 and PM2.5. Table 3 shows the average ratios PM2.5/PM10 and the standard deviations of the daily ratios. The completeness of PM10 and PM2.5 data series was on the average 96%, ranging from 87% to 100% for specific data series.

The lowest PM2.5 concentrations (7.9 µg/m^3) were observed at the elevated site Chaumont (situated on an altitude of 1140 m a.s.l.), the highest (24.4 µg/m^3) at Lugano, situated south of the Alps. Apart from these two sites with their special situation the observed range

Table 1
Characterisation of the sites with PM2.5 measurements (in parenthesis, the abbreviations of the station names, which are used in the figures)

Site	Start of PM2.5 meas.	Characterisation of the site
Dübendorf (DUE)	1998	Suburban, approx. 150 m distance to busy road (measurements only in 1998).
Basel (BAS)	1998	Suburban, quiet situation in a park-like surrounding.
Bern (BER)	1998	Urban, directly at the kerbside of a very busy transit road (approx. 60,000 vehicles/day), 4 m distance from the next lane, high buildings on both sides of the road.
Chaumont (CHA)	1998	Rural, elevated situation at 1140 m a.s.l.
Lugano (LUG)	1999	Urban, situated in a park with trees, south of the Alps.
Payerne (PAY)	1999	Rural, 490 m a.s.l. (Typical altitude of the Swiss basin).
Zürich (ZUE)	1998	Urban background, courtyard in the city centre.

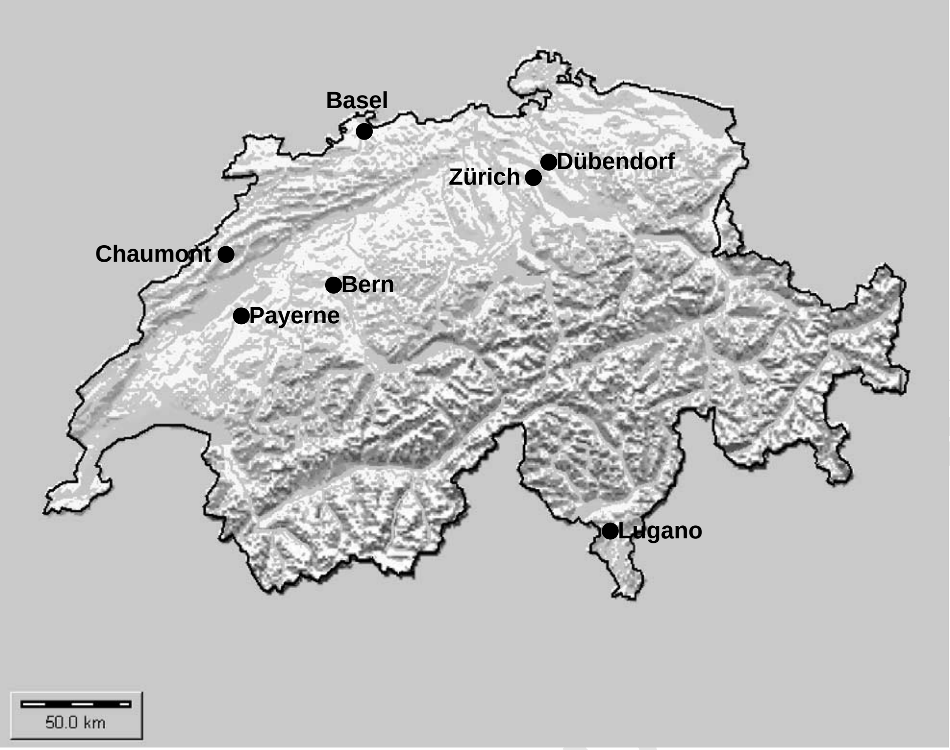

Fig. 1. Geographical position of the 7 investigated sites of the Swiss National Monitoring Network (NABEL).

Table 2
Annual mean values and long-term mean values of PM10 and PM2.5 concentrations

	PM10 (μg/m^3)					PM2.5 (μg/m^3)				
	1998	1999	2000	2001	1998–2001	1998	1999	2000	2001	1998–2001
Dübendorf	26.7	23.6	20.8	20.6	22.9	19.9				19.9
Basel	24.1	23.1	20.5	22.2	22.5	17.8	17.8	15.8	17.3	17.2
Bern	40.3	37.9	33.0	32.5	35.9	23.3	20.3	19.0	20.7	20.8
Chaumont	10.6	12.1	10.2	11.0	11.0	7.7	8.7	7.3	8.1	7.9
Lugano	35.7	30.9	33.8	31.8	33.0		24.3	24.9	24.0	24.4
Payerne	23.2	20.6	19.8	19.3	20.7		15.9	14.7	14.8	15.1
Zürich	24.3	25.3	23.2	23.4	24.0	18.9	18.7	16.9	17.8	18.1

of PM2.5 concentrations was considerably smaller (between 15.1 μg/m^3 at Payerne and 20.8 μg/m^3 at Bern). These are surprisingly low differences for these quite differently exposed sites.

Even at the extremely traffic-exposed site of Bern lower concentrations of PM2.5 were observed than at the site of Lugano, which, though urban, is not directly exposed to traffic emissions. It seems that the Swiss territory south of the Alps generally show a higher concentration level of PM2.5 than the northern parts. The most probable reason for this is the vicinity of the heavily polluted Milan area with its high emissions of primary aerosols as well as gaseous precursors for secondary fine aerosols. Regrettably no PM2.5 data are available for rural sites of the southern part of Switzerland to confirm this statement, but a strong influence of the Milan plume on the air quality of southern Switzerland has already been shown (Grell et al., 2000; Prevot et al., 1997).

The means of the daily PM2.5/PM10 ratios are rather constant at the different sites and vary only between 0.75 and 0.76 in the long-term average (Table 3). The only exception is the kerbside site of Bern, which is strongly influenced by coarse particles from traffic-induced abrasion and resuspension processes (PM2.5/PM10 = 0.59).

Fig. 2 shows the frequency distribution of the daily PM2.5/PM10 ratios. All sites, with the exception of Bern have the maximum between 0.75 and 0.80. At Bern, however, the already mentioned influence of traffic-induced coarse dust is obvious and causes a shift of the maximum to 0.65. The standard deviations of the daily ratios (Table 3) are quite low and vary only from 0.08 to 0.13, except for Chaumont (0.22). This higher standard deviation can at least partly be explained with the higher relative measurement uncertainties due to the low absolute concentrations.

Table 4 shows, that there is a high correlation between PM10 and PM2.5 at all sites. With the exceptions of Bern ($r^2 = 0.86$) and Chaumont ($r^2 = 0.85$) $r^2 \geqslant 0.94$ are observed. For the sites Basel and Bern the connection between PM2.5 and PM10 is visualised in Fig. 3. As shown in an earlier paper for a similar comparison of TSP and PM10, the correlation is good enough and the mean PM2.5/PM10 ratios from year to year are constant enough to allow quite reasonable estimates of long-term (e.g. yearly) PM2.5 concentrations and number of days per year exceeding some threshold values from PM10 data at sites, which are not heavily influenced by local sources of coarse dust (Gehrig and Hofer, 2000). The estimation of the PM2.5 concentration from PM10 for a specific day, however, is subject to larger uncertainties. Using the average ratio PM2.5/PM10 (together with the standard deviation of the daily PM2.5/PM10 ratios, see above) this uncertainty (on the 95% confidence level) is in the range of 16–26% except Chaumont (44%).

Table 3
Mean PM2.5/PM10 ratios of daily values and standard deviations (S.D.) of the daily PM2.5/PM10 ratios

	1998	1999	2000	2001	1998–2001	S.D.
Dübendorf	0.74				0.74	0.08
Basel	0.72	0.77	0.75	0.78	0.75	0.13
Bern	0.58	0.55	0.59	0.65	0.59	0.09
Chaumont	0.74	0.79	0.74	0.73	0.75	0.22
Lugano		0.77	0.72	0.73	0.74	0.11
Payerne		0.78	0.72	0.76	0.75	0.12
Zürich	0.76	0.75	0.73	0.76	0.75	0.11

The lower correlation at Bern reveals that the traffic-induced coarse particles from abrasion and resuspension contained in PM10 follow different temporal emission patterns than PM2.5, which is dominated by exhaust pipe emissions. This is plausible because mechanically produced particles, and in particular resuspension, depend not only on the vehicle frequency but also on the condition of the carriageway (e.g. clean/dirty, wet/dry). At the site of Chaumont the lower correlation can be explained with the generally lower concentrations and the correspondingly higher relative measurement uncertainties.

Table 4
Correlation (r^2) between PM2.5 and PM10 daily values (1998–2001)

Site	Correlation coefficient (r^2)				
	All data	Dec–Feb	Mar–May	Jun–Aug	Sep–Nov
Dübendorf	0.98	0.99	0.91	0.91	0.98
Basel	0.95	0.97	0.90	0.91	0.95
Bern	0.86	0.83	0.84	0.77	0.88
Chaumont	0.85	0.83	0.85	0.89	0.82
Lugano	0.96	0.97	0.94	0.95	0.95
Payerne	0.94	0.97	0.94	0.94	0.93
Zürich	0.95	0.97	0.92	0.92	0.93

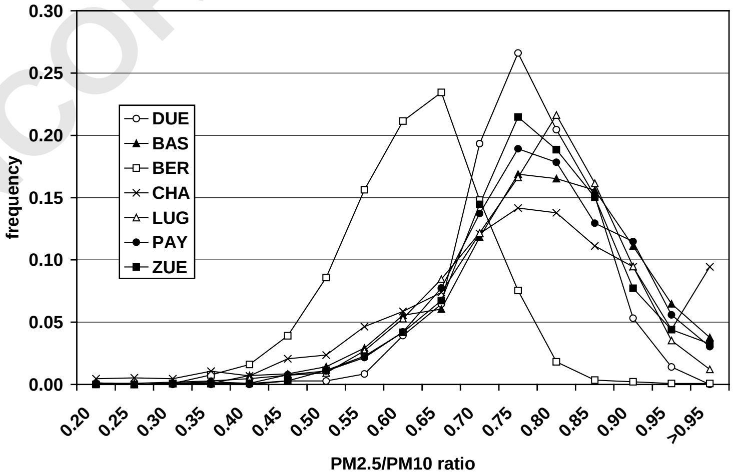

Fig. 2. Frequency distributions of the PM2.5/PM10 ratios of the daily values (all measurements 1998–2001).

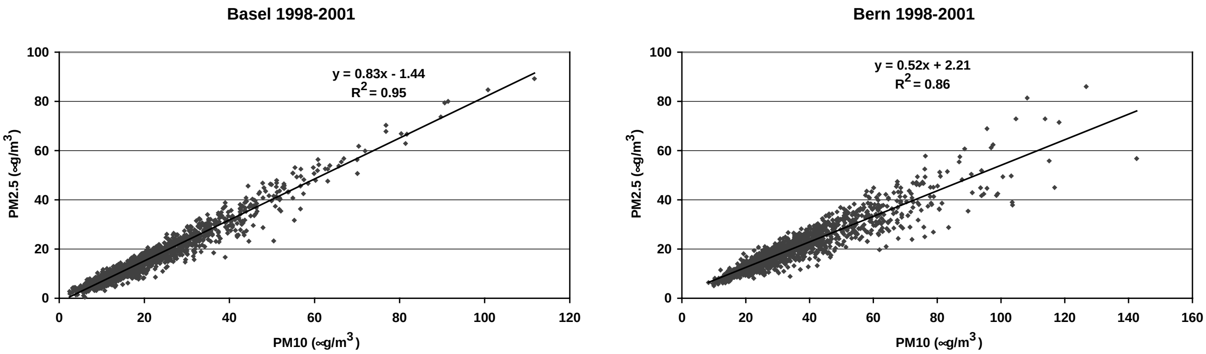

Fig. 3. Scatterplots of PM2.5 versus PM10 at Basel and Bern (daily values 1998–2001).

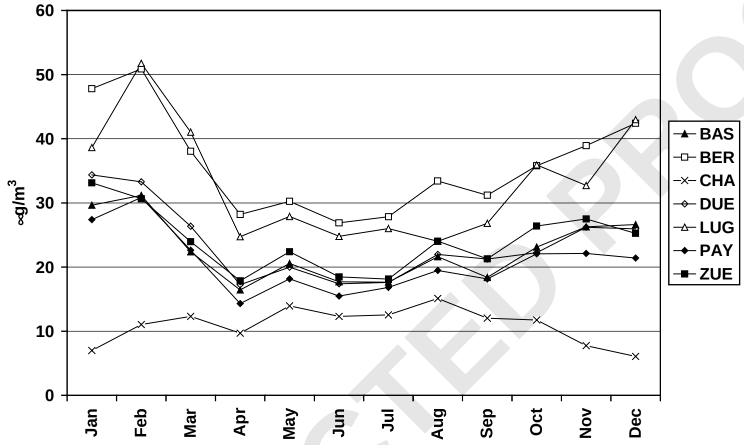

Fig. 4. Seasonal variations of PM10 monthly means 1998–2001.

3.2. *Seasonal variations of PM10 and PM2.5 concentrations*

It can be seen from Figs. 4 and 5 that for all sites, with the exception of the elevated site of Chaumont, a characteristic seasonal variation can be observed for PM10 and PM2.5 with elevated concentrations during the cold season. The reasons for this are not primarily caused by seasonal fluctuations of the emissions, but rather by meteorological effects. This is already well known from similar variations of other parameters like sulphur dioxide or nitrogen oxide (frequent inversions during winter and good vertical mixing during summer). In contrast, Chaumont shows the lowest values in winter. This also shows the dominating influence of the meteorology. The site is situated on an altitude of 1140 m a.s.l. and, therefore, during wintertime in the majority of cases above the inversion layer, thus protected from the emissions of the lowlands of the Swiss basin. From April to September the variations at Chaumont follow that of the other sites, though on a lower concentration level, due to the better vertical mixing of the lower atmosphere during the warmer season. This effect is further discussed in Section 3.3. Fig. 6 shows that the PM2.5/PM10 ratios are not constant over the year. In general lower values were observed during spring and partly also during summer, indicating presumably the occurrence of coarse biogenic dust (e.g. pollen). At Bern, this seasonal variation of the PM2.5/PM10 ratios cannot be observed. Obviously, if present at all, it is masked by the massive influence of locally produced exhaust and road dust.

3.3. *Spatial distribution of PM10 and PM2.5 in Switzerland*

Interesting information about the spatial distribution of the PM10 and PM2.5 concentrations over Switzerland can be obtained when analysing the correlation of the daily values between the different sites. Tables 5 and 6 give an overview of the correlation coefficients (r^2) of the daily means for PM10 and PM2.5 between all sites over the whole measurement period from 1998 to 2001.

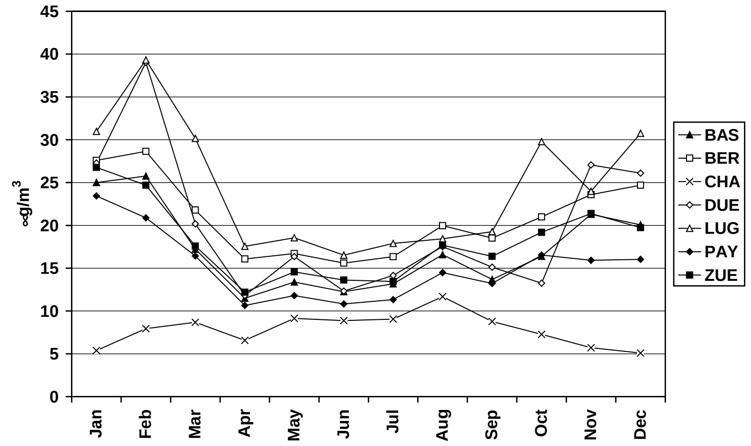

Fig. 5. Seasonal variations of PM2.5 monthly means 1998–2001.

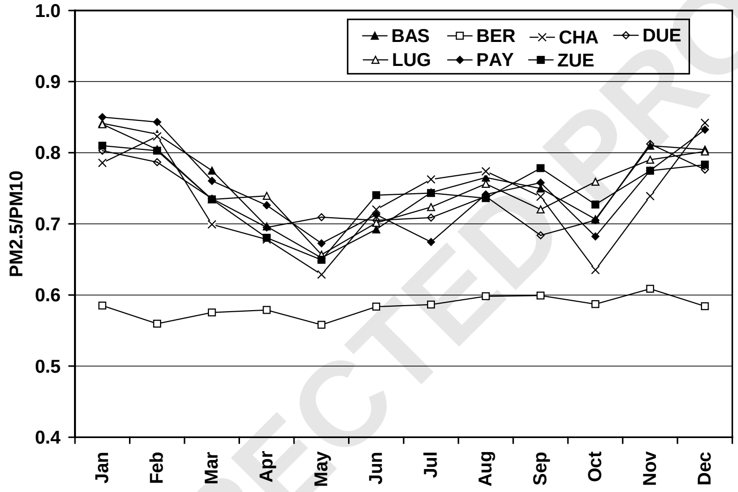

Fig. 6. Seasonal variations of the PM2.5/PM10 ratios (of the monthly means 1998–2001).

Table 5
Correlation (r^2) of the daily values of PM2.5 between the sites (all measurements 1998–2001; Dübendorf 1998 only, Lugano and Payerne 1999–2001 only)

PM2.5	Dübendorf	Basel	Bern	Chaumont	Lugano	Payerne
Dübendorf	1.00					
Basel	0.85 (0.75/0.78)	1.00				
Bern	0.78 (0.77/0.60)	0.69 (0.68/0.56)	1.00			
Chaumont	0.11 (0.19/0.59)	0.22 (0.27/0.69)	0.17 (0.16/0.61)	1.00		
Lugano		0.12 (0.02/0.24)	0.16 (0.01/0.33)	0.04 (0.02/0.24)	1.00	
Payerne		0.72 (0.68/0.70)	0.71 (0.65/0.71)	0.30 (0.36/0.85)	0.19 (0.06/0.34)	1.00
Zürich	0.95 (0.95/0.90)	0.80 (0.76/0.80)	0.73 (0.73/0.69)	0.21 (0.18/0.64)	0.12 (0.02/0.31)	0.68 (0.63/0.70)

In parenthesis the correlations (r^2) for the winter months from December to February and for the summer months June–August are given separately in the format (winter/summer).

Table 6
Correlation (r^2) of the daily values of PM10 between the sites (all measurements 1998–2001)

PM10	Dübendorf	Basel	Bern	Chaumont	Lugano	Payerne
Dübendorf	1.00					
Basel	0.82 (0.78/0.77)	1.00				
Bern	0.66 (0.67/0.41)	0.62 (0.60/0.42)	1.00			
Chaumont	0.13 (0.17/0.61)	0.22 (0.22/0.71)	0.12 (0.13/0.50)	1.00		
Lugano	0.17 (0.06/0.16)	0.15 (0.05/0.16)	0.19 (0.05/0.18)	0.05 (0.04/0.20)	1.00	
Payerne	0.75 (0.67/0.63)	0.75 (0.70/0.67)	0.68 (0.68/0.57)	0.27 (0.29/0.82)	0.23 (0.12/0.27)	1.00
Zürich	0.91 (0.94/0.86)	0.77 (0.73/0.79)	0.60 (0.59/0.51)	0.22 (0.17/0.70)	0.15 (0.05/0.21)	0.71 (0.61/0.69)

In parenthesis the correlations (r^2) for the winter months from December to February and for the summer months June–August are given separately in the format (winter/summer).

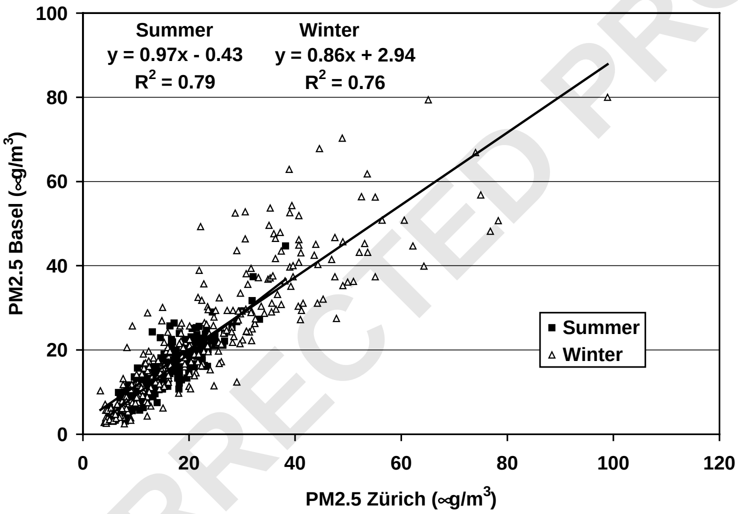

Fig. 7. Scatterplot and linear regression of the daily values of PM2.5 at Zürich and Basel during summer (June to August) and during winter (December–February).

There are only small differences between the behaviour of PM10 and PM2.5, with slightly higher correlation for PM2.5. This was to be expected as the finer fraction has a longer lifetime in the atmosphere and hence tend to a more homogenous distribution.

The correlation coefficients between the sites Dübendorf, Basel, Bern, Payerne and Zürich, which are all situated in the lowlands of the Swiss basin, are surprisingly high. This indicates that meteorological conditions and emissions from sources, which are effective over all the area (e.g. traffic), rather than specific local sources and events dominate the relative variations of the concentrations of fine dust. As expected, Chaumont, which is often above the inversion layer and Lugano, which is separated from the Swiss basin by the Alps, exhibit considerably lower or virtually no correlation.

For some sites an interesting dependence of the correlation coefficients from the season can be observed. Tables 5 and 6 give the correlation coefficients for winter (December–February) and summer (June–August) separately. Especially during summer, when good vertical mixing of the lower atmosphere is prevailing, the correlations are often higher. This is elaborated in more detail by means of three examples.

Fig. 7 shows the correlation for PM2.5 between the sites Basel and Zürich. Though the distance between these two sites is 72 km and they are separated by the

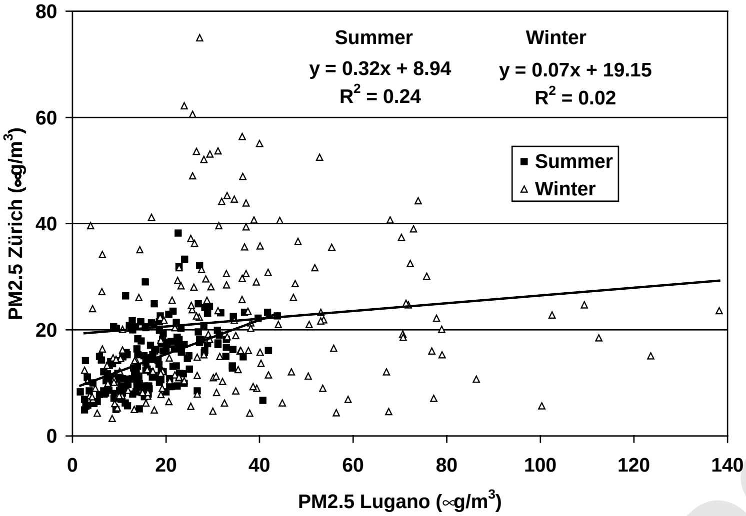

Fig. 8. Scatterplot and linear regression of the daily values of PM2.5 at Lugano and Zürich during summer (June–August) and during winter (December–February).

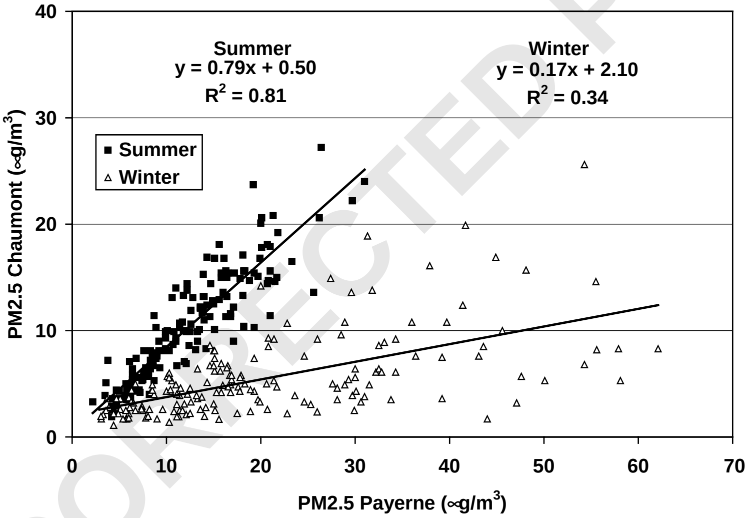

Fig. 9. Scatterplot and linear regression of the daily values of PM2.5 at Chaumont and Payerne during summer (June–August) and during winter (December–February).

600–800 m high Jura Mountains high correlation can be observed in particular in summer, but also during wintertime. Considering the very similar results for the sites Dübendorf, Bern and Payerne (Tables 5 and 6) the high interconnection of the relative variations of PM10 and PM2.5 concentrations can be generalised for the whole lowlands north of the Alps (Swiss basin). For PM2.5 even the absolute concentrations at these sites are very similar (Fig. 5).

Fig. 8 compares Zürich, north of the Alps with Lugano, south of the Alps. As expected no correlation can be observed during wintertime. During summertime the correlation is somewhat higher but still very weak. This shows clearly, that the high mountains of the Alps form an efficient obstacle for the distribution and homogenisation of fine particles.

Fig. 9 shows a comparison of the two sites Payerne and Chaumont for PM2.5. The sites are located quite close together (distance 24 km) but on different altitudes. Chaumont is situated 650 m higher than Payerne. A high correlation can be observed during summertime, when the vertical mixing of the lower atmosphere is generally good and the absolute concentration level of the mountain site is only about 20% lower than at Payerne, which is situated within the Swiss basin in a rural environment. However, during wintertime, when the meteorology is characterised by frequent inversion, the observed PM2.5 levels are largely decoupled. The correlation is very low and the absolute concentration level at the mountain site Chaumont reaches only about 20% of Payerne.

4. Conclusions

It has been shown from the collocated measurements that there is a strong connection between PM10 and PM2.5 concentrations, with the exception of sites, which are influenced by nearby strong and variable local sources (kerbsides, construction sites, strongly dust emitting industries). Furthermore, in absence of dominating local sources PM2.5 concentrations tend to be quite evenly distributed over surprisingly large areas unless these are not separated by topographic obstacles like high mountains, which induce different meteorological regimes. PM2.5 concentration levels in typical situations can reasonably be estimated from a limited number of measurement sites. Therefore, from the point of view of an efficient use of financial and personal resources, the number of additional collocated PM2.5 measurements at PM10 sites can be kept quite limited. The saved resources could then be used to investigate other interesting particle related parameters, which, in contrast to PM2.5 measurements, provide substantial new information (e.g. on particle sources and ageing) like PM1, particle number concentrations, morphology or chemical composition. Such additional monitoring work will become increasingly important, as recently published papers give serious indications about adverse health effects of nanoparticles (Hoek et al., 2002; Johnston et al., 2000; Oberdorster, 2001). However, due to their negligible mass these nanoparticles are virtually not reflected by gravimetric PM measurements.

Acknowledgements

We gratefully acknowledge the financial support of the Swiss Federal Office for the Environment, Forests and Landscape. We thank the NABEL team for their dedicated and reliable work on the sites as well as in the analytical laboratory.

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