

EMEP Assessment Report – Switzerland (Part 2: Particles)

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Abstract

Gravimetric measurements of TSP from Swiss sites are available since the early eighties. PM10 measurements started in 1997. Over all, a decreasing trend of PM10 (resp. TSP) concentrations is observed. Collocated parallel measurements of PM2.5 and PM10 were conducted at 7 sites in Switzerland since January 1998. Parallel PM2.5 and PM10 measurements were conducted since 1998. For sites within the Swiss plateau the differences of the PM2.5 concentrations are surprisingly low. 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 occur in winter. Exceptions are Chaumont (1140-m a.s.l.) and Jungfrauoch (3580-m a.s.l.), which are often positioned above the inversion layer (especially during wintertime), resulting in low concentrations. Despite of different exposition characteristics, the correlation of the daily values of PM2.5 (and PM10) between the sites of the Swiss plateau is high, indicating a dominant influence of regional meteorology over local events and sources.

1. Measurement programme, methods and quality assurance

Measurements of total suspended particulate matter (TSP) have been performed within the scope of the Swiss Federal Monitoring Network (NABEL) since the early eighties. In 1997 measurements of PM10, which is considered to represent the thoracic fraction of the ambient particles (ISO, 1995), have been included into the measurement programme. Due to the increasing interest for the alveolar fraction the measurement programme of the network has been extended to include PM2.5 measurements into the measurement programme at 7 sites already in 1998. Table 1 lists the sites where the PM measurements have been performed and gives information about the type of pollution exposition at each site. The map in figure 1 shows the geographical position of the investigated sites within Switzerland. A detailed description of the sites has been published in (EMPA, 2000).

All particle samplings were conducted with High-Volume-Samplers Digital 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) in its version for TSP measurements (sampling flow 40 m³/h). The sampling inlets for PM10 are operated at a flow of 30 m³/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 for the time being 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 on the Digital instrument with the WINS impactor, which is the 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 $\pm 10\%$ (95% confidence interval for single daily values) in the concentration range 10 – 30 $\mu\text{g}/\text{m}^3$. The detection limit was determined from the standard deviation of field blanks to be 1 $\mu\text{g}/\text{m}^3$. Because the only difference between the applied

method for PM₁₀, TSP and PM_{2.5} is the design of the sampling heads, the same measurement uncertainty can be assumed also for the TSP and PM_{2.5} measurements.

Table 1: Characterisation of the investigated sites (in parenthesis, the abbreviations of the station names, which are used in the figures)

Site	Characterisation of the site
Dübendorf (DUE)	Suburban, approx. 150 m distance to busy road (measurements only in 1998).
Basel (BAS)	Suburban, quiet situation in a park-like surrounding.
Bern (BER)	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)	Rural, elevated situation at 1140 m a.s.l.
Lugano (LUG)	Urban, situated in a park with trees, south of the Alps.
Payerne (PAY)	Rural, 490 m a.s.l. (Typical altitude of the Swiss basin).
Rigi (RIG)	Rural, elevated situation at 1030 m a.s.l.
Tänikon (TAE)	Rural, 540 m a.s.l. (Typical altitude of the Swiss basin).
Zürich (ZUE)	Urban background, courtyard in the city centre.
Jungfrauoch (JUN)	High alpine, 3580 m a.s.l.

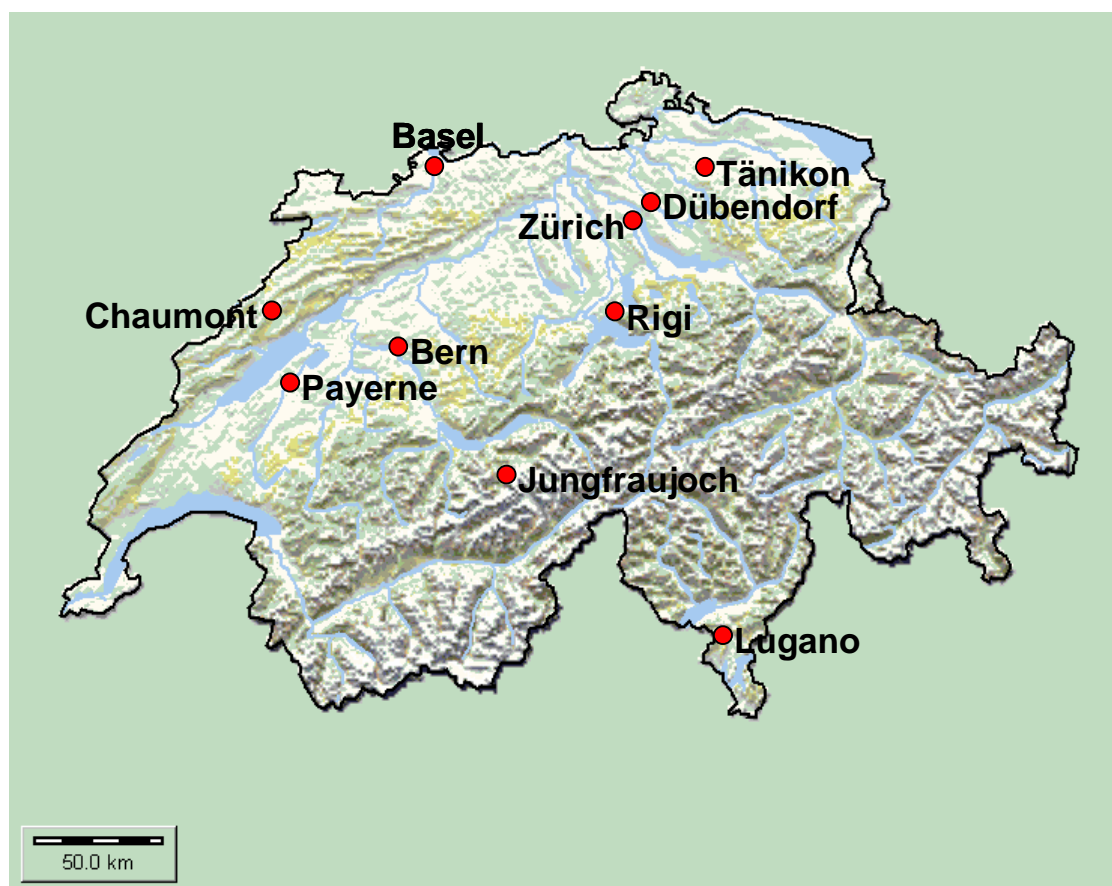


Fig. 1. Geographical position of the investigated sites of the Swiss National Monitoring Network (NABEL)

2. Results

2.1 Long term trend of TSP and PM10 concentrations at the Swiss EMEP sites

Table xy shows the yearly means of the TSP and PM10 measurements, which were performed at the Swiss EMEP sites. Until 1996 all sites had a TSP sampling head. From 1997 all sampling heads were changed to PM10 with the exception of Jungfrauoch, where for technical reasons the no changes were made. At several sites parallel TSP and PM10 sampling was conducted in 1997 and 1998 in order to get information on the relation of TSP and PM10. For rural sites ,which are not exposed to strong local sources of coarse particles, a very stable ratio PM10/TSP of 0.88 (± 0.02) could be observed for annual means (Gehrig and Hofer, 2000). This allowed an estimation of PM10 concentrations based on TSP for data before 1997. These estimated PM10 concentrations are given in italics in table 2. Figure 2 illustrates the decreasing trend of PM10 at all Swiss EMEP sites.

Table 2: Annual means of TSP and PM10 in $\mu\text{g}/\text{m}^3$ for the Swiss EMEP sites. Estimated PM10 concentrations (based on TSP measurements) are given in italics.

	JUN	CHA	PAY	RIG	TAE	CHA	PAY	RIG	TAE
Year	TSP	TSP	TSP	TSP	TSP	PM10	PM10	PM10	PM10
1981			35		35		<i>31</i>		<i>31</i>
1982	5.7		37		38		<i>32</i>		<i>34</i>
1983	8.5		40		40		<i>35</i>		<i>36</i>
1984	5.9		39		39		<i>34</i>		<i>35</i>
1985	5.1		44		43		<i>39</i>		<i>37</i>
1986	4.0		44		43		<i>39</i>		<i>38</i>
1987	6.7		39		33		<i>35</i>		<i>29</i>
1988	4.9		32		30		<i>29</i>		<i>27</i>
1989	4.5		37		36		<i>33</i>		<i>31</i>
1990	4.3		34		31		<i>30</i>		<i>28</i>
1991	4.0	22	34		35	<i>20</i>	<i>30</i>		<i>31</i>
1992	3.3	18	31	22	31	<i>15</i>	<i>27</i>	<i>19</i>	<i>27</i>
1993	5.5	17	30	16	28	<i>15</i>	<i>26</i>	<i>14</i>	<i>25</i>
1994		16	27	16	24	<i>14</i>	<i>24</i>	<i>14</i>	<i>22</i>
1995	8.4	16	27	16	26	<i>14</i>	<i>24</i>	<i>14</i>	<i>23</i>
1996	5.4	18	31	19	31	<i>16</i>	<i>27</i>	<i>17</i>	<i>27</i>
1997	3.5	16	29			14	26	14	27
1998	4.0		27			11	23	12	20
1999	3.8					12	21	12	19
2000	3.4					10	20	11	18
2001	3.2					11	19	12	18
2002	3.4					12	21	13	20

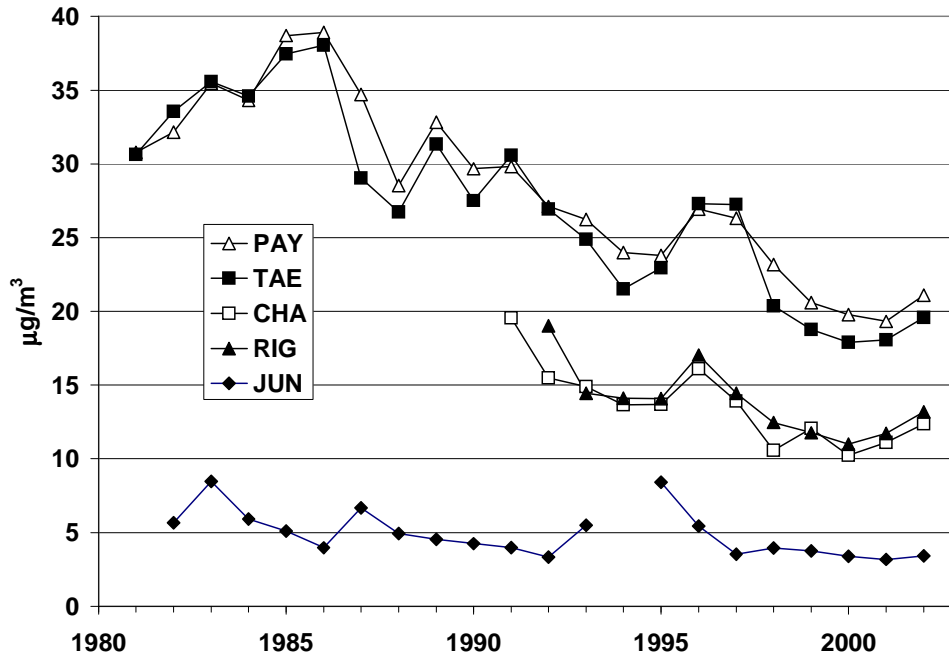


Fig. 2: Long-term PM10 trends at the Swiss EMEP sites

2.2 Mass concentration of PM10 and PM2.5 from 1998 – 2001

Table 3 gives an overview of the annual mean concentrations of PM10 and PM2.5. Table 4 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 ($7.9 \mu\text{g}/\text{m}^3$) were observed at the elevated site Chaumont (situated on an altitude of 1140 m a.s.l.), the highest ($24.4 \mu\text{g}/\text{m}^3$) at Lugano, situated south of the Alps. Apart from these two sites with their special situation the observed range of PM2.5 concentrations was considerably smaller (between $15.1 \mu\text{g}/\text{m}^3$ at Payerne and $20.8 \mu\text{g}/\text{m}^3$ 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 4). The only exception is the kerbside site of Bern, which is strongly influenced by coarse particles from traffic induced abrasion and resuspension processes ($\text{PM}_{2.5}/\text{PM}_{10}=0.59$).

Table 5 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 ($r^2 = 0.85$) $r^2 \geq 0.94$ are observed. 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 3

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

	PM10 ($\mu\text{g}/\text{m}^3$)					PM2.5 ($\mu\text{g}/\text{m}^3$)				
	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

Table 4

Mean PM2.5/PM10 ratios of daily values and standard deviations (STD) of the daily PM2.5/PM10 ratios

	1998	1999	2000	2001	1998-2001	STD
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

Table 5

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

2.3 Seasonal variations of PM10 and PM2.5 concentrations

It can be seen from figures 3 and 4 that for all sites, with the exception of the elevated sites of Chaumont and Jungfrauoch, 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 and Jungfrauoch show the lowest values in winter. This also illustrates the dominating influence of the meteorology. The sites are especially during wintertime often situated above the inversion layer, thus protected from the emissions of the lowlands of the Swiss basin. From April to September the variations at Chaumont and Jungfrauoch follow that of the other sites, though on a lower concentration level. Figure 5 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.

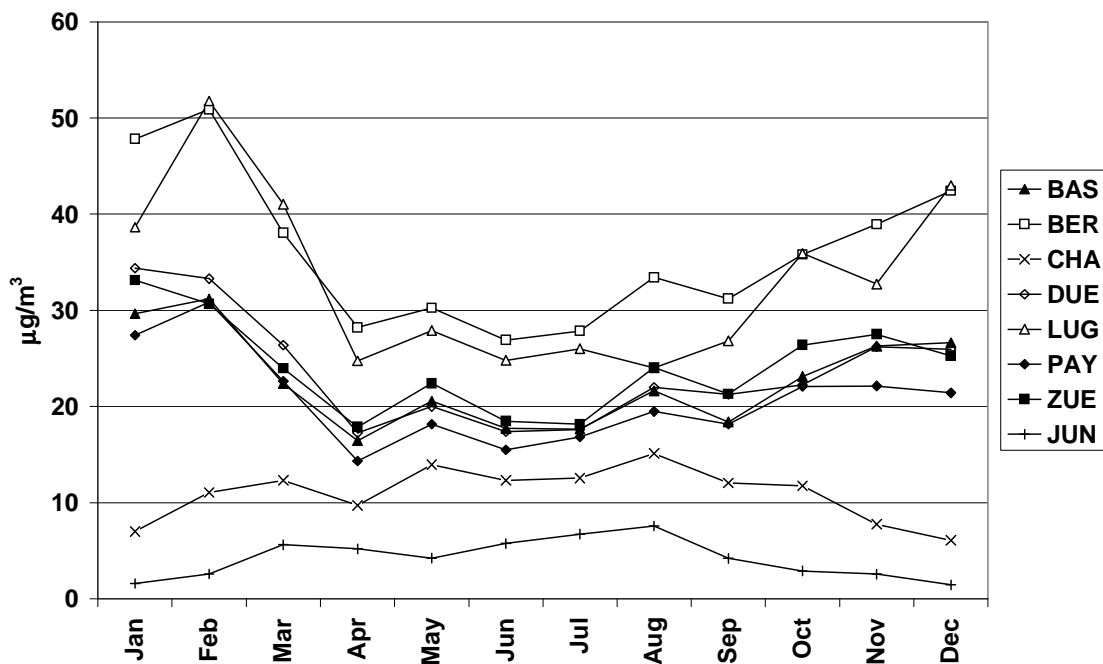


Fig. 3. Seasonal variations of PM10 monthly means 1998-2001 (JUN: TSP monthly means 1990-2002)

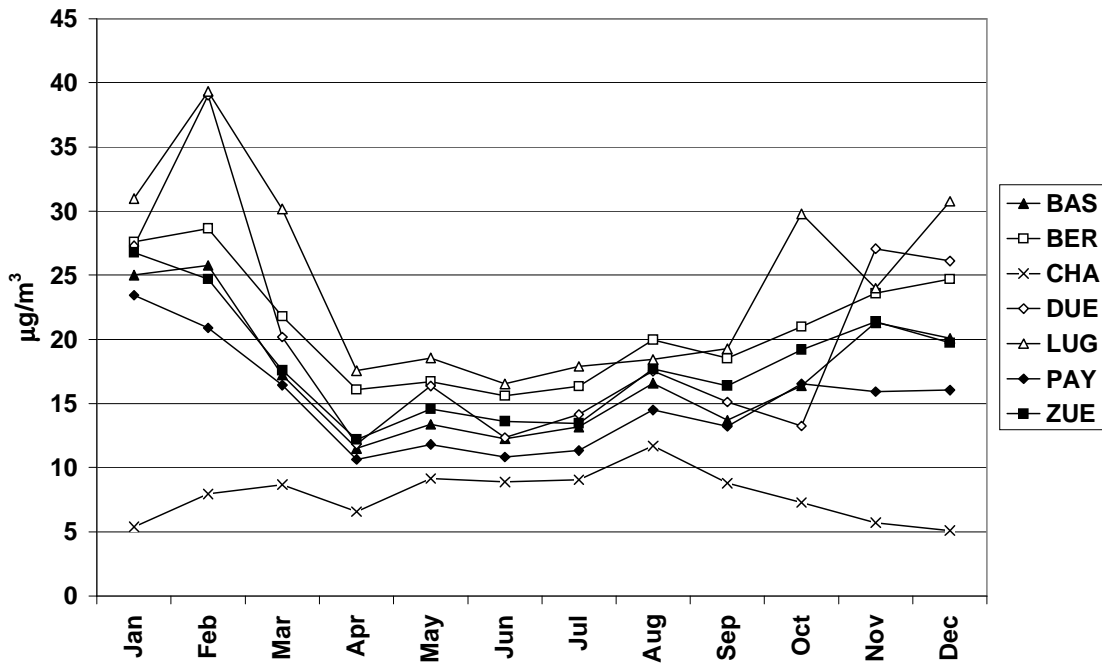


Fig. 4. Seasonal variations of PM2.5 monthly means 1998-2001

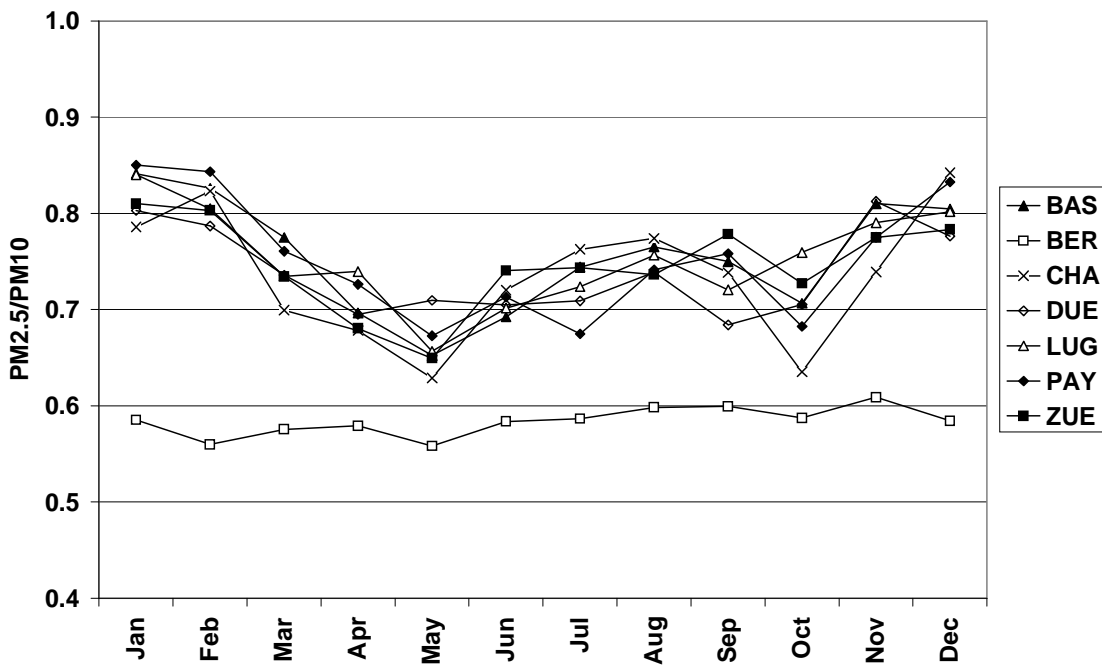


Fig. 5. Seasonal variations of the PM2.5/PM10 ratios (of the monthly means 1998-2001)

2.4 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. Table 6 gives 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 1998-2001. 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, Lugano, which is separated from the Swiss basin by the Alps, exhibit considerably lower or virtually no correlation.

Figure 6 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.

Table 6

Correlation (r^2) of the daily values of PM2.5 (and PM10 after slash) between the sites (all measurements 1998-2001; Dübendorf 1998 only, Lugano and Payerne 1999-2001 only).

	Dübendorf	Basel	Bern	Chaumont	Lugano	Payerne
Dübendorf	1.00					
Basel	0.85/0.82	1.00				
Bern	0.78/0.66	0.69/0.62	1.00			
Chaumont	0.11/0.13	0.22/0.22	0.17/0.12	1.00		
Lugano		0.12/0.15	0.16/0.19	0.04/0.05	1.00	
Payerne		0.72/0.75	0.71/0.68	0.30/0.27	0.19/0.23	1.00
Zürich	0.95/0.91	0.80/0.77	0.73/0.60	0.21/0.22	0.12/0.15	0.68/0.71

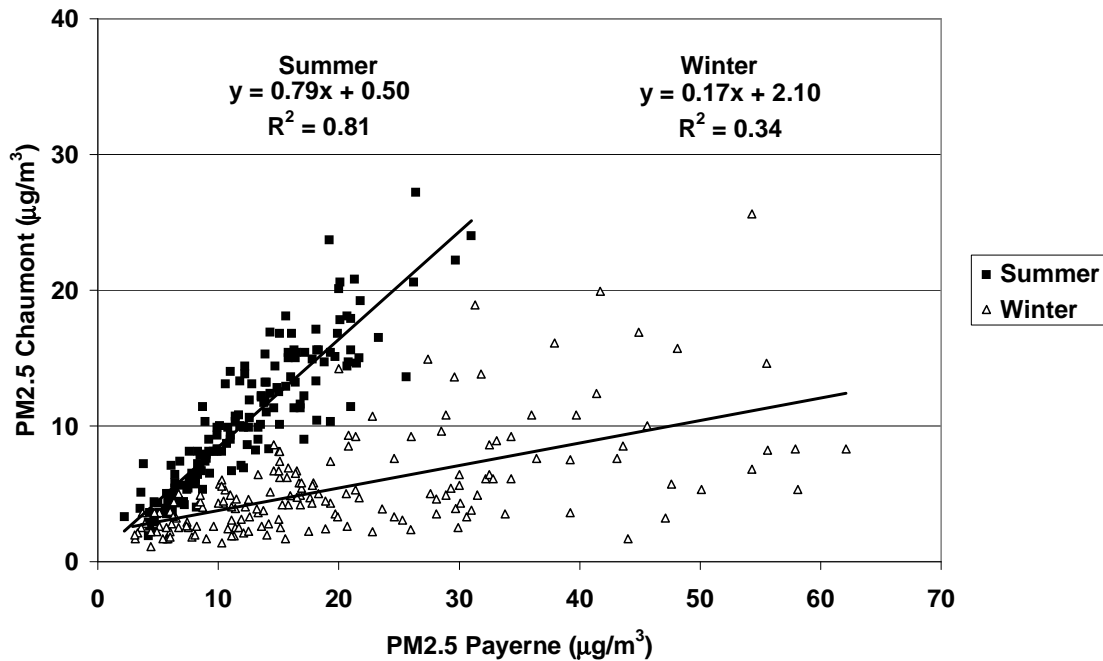


Fig. 6. Scatterplot and linear regression of the daily values of PM_{2.5} at Chaumont and Payerne during summer (June to August) and during winter (December to February)

3. Conclusions

It has been shown from the collocated measurements that there is a strong connection between PM₁₀ and PM_{2.5} 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 PM_{2.5} 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. PM_{2.5} 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 PM_{2.5} 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 PM_{2.5} 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.

4. References

- CEN, 1998. Air Quality - Determination of the PM₁₀ fraction of suspended particulate matter - Reference method and field test procedure to demonstrate reference equivalence of measurement methods, EN 12341.
- EMPA, 2000. Technischer Bericht zum Nationalen Beobachtungsnetz für Luftfremdstoffe (NABEL), pp. 164, Dubendorf, Switzerland.

- Gehrig, R., and Hofer, P., 2000. Parallel measurements of PM₁₀ and total suspended particles (TSP) - Estimation of PM₁₀-characteristics from TSP data, *Gefahrstoffe Reinhaltung Der Luft* 60(10), 389-394.
- Grell, G.A., Emeis, S., Stockwell, W.R., Schoenemeyer, T., Forkel, R., Michalakes, J., Knoche, R., and Seidl, W., 2000. Application of a multiscale, coupled MM5/chemistry model to the complex terrain of the VOTALP valley campaign, *Atmospheric Environment* 34(9), 1435-1453.
- Hoek, G., Meliefste, K., Cyrus, J., Lewne, M., Bellander, T., Brauer, M., Fischer, P., Gehring, U., Heinrich, J., van Vliet, P., and Brunekreef, B., 2002. Spatial variability of fine particle concentrations in three European areas, *Atmospheric Environment* 36(25), 4077-4088.
- ISO, 1995. Air quality - Particle size fraction definitions for health related sampling, ISO 7708.
- Johnston, C.J., Finkelstein, J.N., Mercer, P., Corson, N., Gelein, R., and Oberdorster, G., 2000. Pulmonary effects induced by ultrafine PTFE particles, *Toxicology and Applied Pharmacology* 168(3), 208-215.
- Oberdorster, G., 2001. Pulmonary effects of inhaled ultrafine particles, *International Archives of Occupational and Environmental Health* 74(1), 1-8.
- Prevot, A.S.H., Staehelin, J., Kok, G.L., Schillawski, R.D., Neininger, B., Staffelbach, T., Neftel, A., Wernli, H., and Dommen, J., 1997. The Milan photooxidant plume, *Journal of Geophysical Research-Atmospheres* 102(D19), 23375-23388.
- UMEG, 1999. Prüfung des DIGITEL Staubsammlers DHA80 mit PM₁₀-Einlass nach prEN 12341, pp. 87, UMEG Karlsruhe.
- VDI, 1996. Messen von Partikeln - Messen der Massenkonzentration (Immission) - Filterverfahren - Filterwechsler Digital DHA-80, VDI 2463, Blatt 11.