EMEP Assessment Report – Switzerland

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Part I: Gases and Precipitation

Abstract

In Switzerland, ambient air concentrations of sulphur dioxide and nitrogen dioxide have decreased during the last two decades. Sulphate in particulate matter and the wet sulphate deposition have also decreased. The wet depositions of nitrate and ammonium have not changed since 1985, although their concentrations in precipitation have decreased. The concentrations of lead and cadmium have decreased in the last decade. The annual mean of ozone has increased since 1990, whereas the AOT40-values for forests and for crops remained unchanged.

1. Monitoring stations and measurement programme

In Switzerland, five stations of the National Air Pollution Monitoring Network (NABEL) are instrumented as EMEP stations: Chaumont, Jungfraujoch, Payerne, Rigi-Seebodenalp and Tänikon (Fig.1 and Table 1). At all stations SO_2, NO_2, O_3, particulate sulphate and heavy metals in particulate matter are measured. Precipitation analysis is done at all stations with the exception of the station at Jungfraujoch.

Table 1: Characterisation of the Swiss EMEP sites.

Site	*Characterisation of the site*
Chaumont	Rural, elevated situation at 1140 m a.s.l.
Payerne	Rural, 490 m a.s.l. (Typical altitude of the Swiss Plateau).
Rigi-Seebodenalp	Rural, elevated situation at 1030 m a.s.l.
Tänikon	Rural, 540 m a.s.l. (Typical altitude of the Swiss Plateau).
Jungfraujoch	High alpine, 3580 m a.s.l.

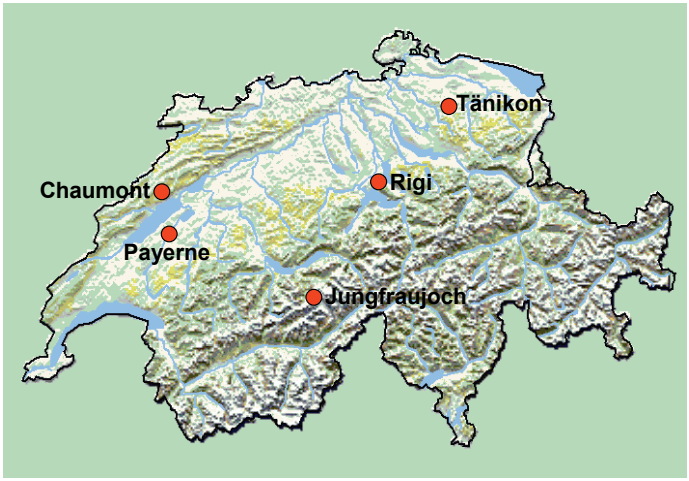


Fig. 1. Geographical position of the investigated sites of the Swiss National Air Pollution Monitoring Network (NABEL).

2. Emissions

Figure 2 shows the emissions to air of sulphur oxides, nitrogen oxides and non-methane volatile organic compounds in Switzerland. Emissions reductions were successful for all three substances. The strongest reduction was achieved for SO_x-emissions (Table 2) with a stronger reduction in the neighbouring countries (Austria, Germany, France and Italy) than in Switzerland, mainly after 1990. NO_x and NMVOC emission reductions were slightly stronger in Switzerland than in the neighbouring countries. Thus, local and regional emissions of important ozone precursors were reduced in the last two decades.

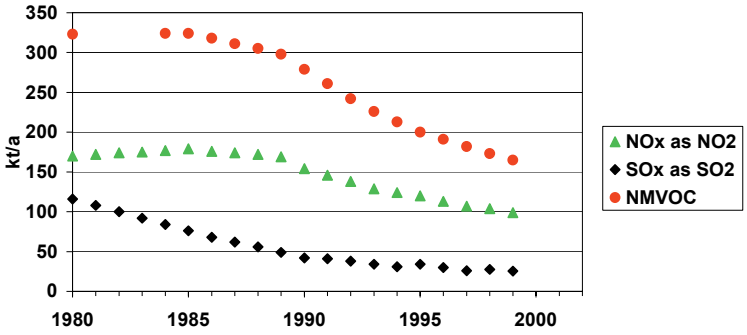


Figure 2.
Emissions of SO_x, NO_x and NMVOC in Switzerland in kt per year.

Table 2: Trends of emissions in Switzerland and in the neighbouring countries (Austria, Germany, France and Italy).

	SO_x		NO_x		NMVOC	
Trend	Switzerland	Neighbours	Switzerland	Neighbours	Switzerland	Neighbours
1980-1999	-78%	-86%	-51%	-31%	-62%	
1990-1999	-40%	-67%	-35%	-29%	-40%	-36%

3. Sulphur

3.1 Sulphur dioxide

At all sites, sulphur dioxide concentrations have significantly decreased (Fig. 3). At Payerne and Jungfraujoch, the SO_2-S has decreased by about 90% since 1980. This is in good agreement with the emission reductions in Switzerland and in neighbouring countries.

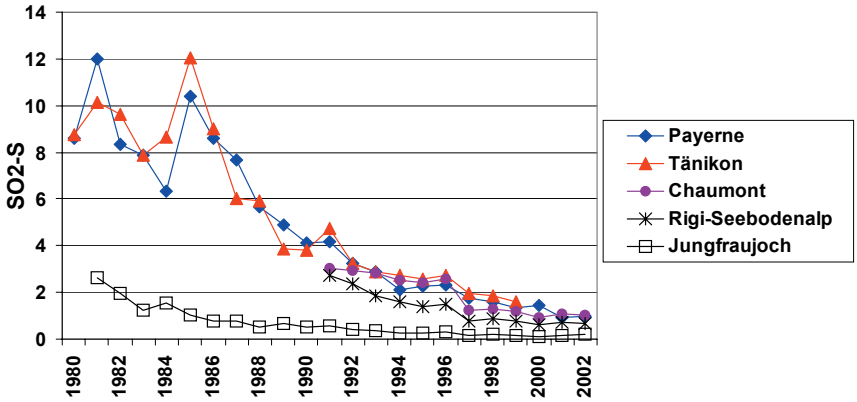


Figure 3. Annual means of SO_2-S at Swiss monitoring sites 1980-2002 (in µgS/m^3).

Using the daily sector values (based on backward trajectories), as provided by the EMEP MSC-W, the origin of high sulphur dioxide pollution observed at Jungfraujoch can be determined. In most years, the origin of the highest values lies in the northeast. On average over all years the SO_2 values from northeast direction are 80% higher than the values from the other sectors. The annual mean values of the northeast sector have decreased by 95% since 1985, whereas the mean values of the other sectors have decreased by about 80%.

3.2 Particulate sulphate

At Payerne, particulate sulphur decreased by 75%, at Jungfraujoch by 80% since 1981 (Fig. 4). The shorter time series of the other stations confirm this trend. Most of this reduction took place after 1990. The SO_x- emissions of the neighbouring countries (Fig. 2b) show the same behaviour, whereas the Swiss SO_x-emissions (Fig.2) were mostly reduced prior to 1990.

The daily sectors of air mass origin do not clearly indicate a main source sector for high particulate sulphate concentrations at Jungfraujoch, although the northeast sector has 45% higher mean values than the other sectors. Also the high values at Jungfraujoch in 1991 cannot be attributed to a specific source region, as all sectors show high values in that year. These high concentrations of sulphate may be caused by the eruption of Mount Pinatubo.

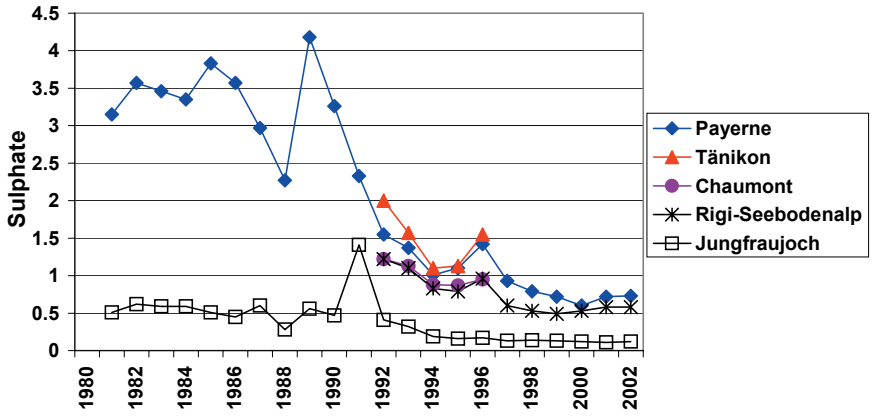


Figure 4. Annual means of particulate sulphate 1981-2002 (in µgS/m^3).

3.3 Wet deposition of sulphate

At Payerne, the concentration of sulphate in precipitation (Fig. 5a) has decreased by more than 60% since 1985 and also the wet S-deposition (Fig. 5b) has decreased by 50%. At the station Rigi the wet S-deposition has increased parallel to precipitation during the last six years.

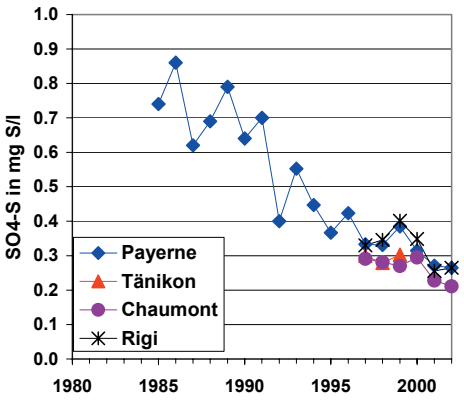


Figure 5a.
Annual mean concentrations of sulphate in precipitation 1985-2002 (in mgS/l).

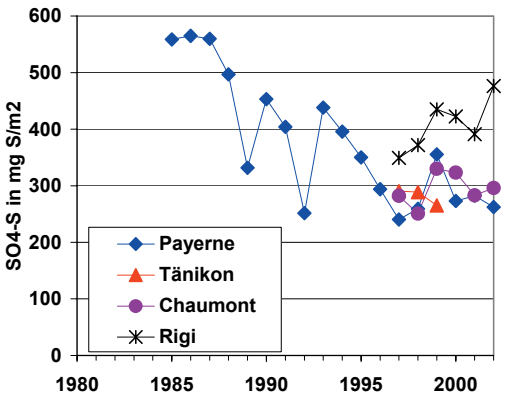


Figure 5b.
Annual wet depositions of sulphate in Switzerland 1985-2002 (in mgS/m^2).

4. Nitrogen

4.1 Nitrogen dioxide

At lower altitude sites (Payerne and Tänikon), the NO_2-nitrogen has decreased by 35% since 1985 (Fig. 6). At altitudes around 1000 m a.s.l. no trend in the NO_2-concentrations is observed. This may reflect the stronger reduction of NO_x-emissions in Switzerland compared to the European reductions.

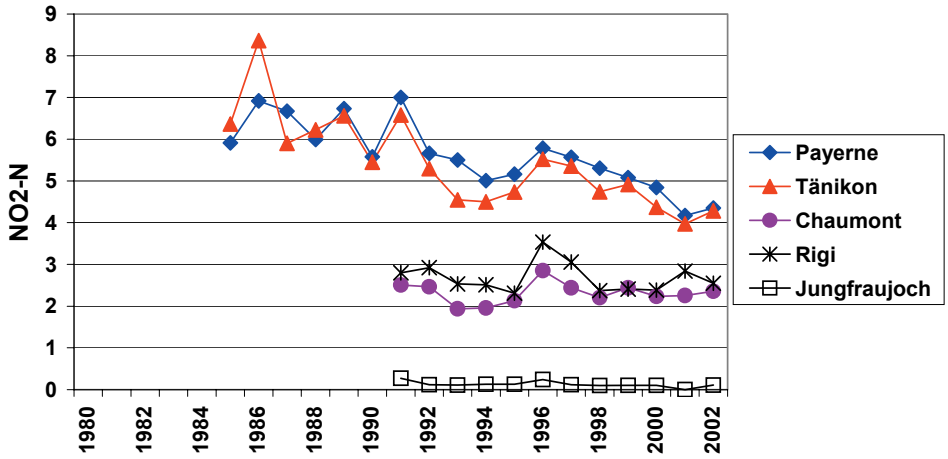


Figure 6. Annual means of NO_2-N at Swiss monitoring sites 1985-2002 (in µgN/m^3).

4.2 Wet deposition of nitrogen

The concentrations of nitrate and ammonia in precipitation have decreased since 1985 (Figs. 7a and 8a). However, the wet depositions of nitrate and ammonia have not changed (Figs. 7b and 8b) with exception of the station Rigi where precipitation and consequently the wet deposition have increased.

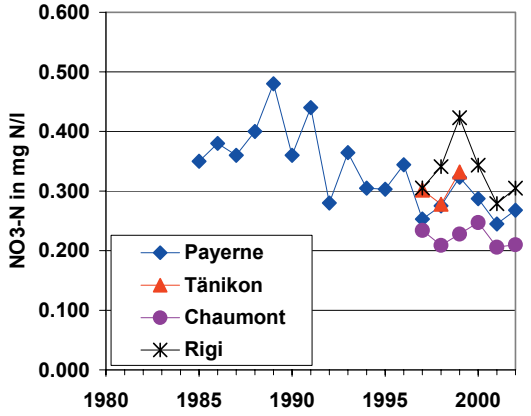


Figure 7a. Annual mean concentrations of nitrate in precipitation 1985-2002 (in mgN/l).

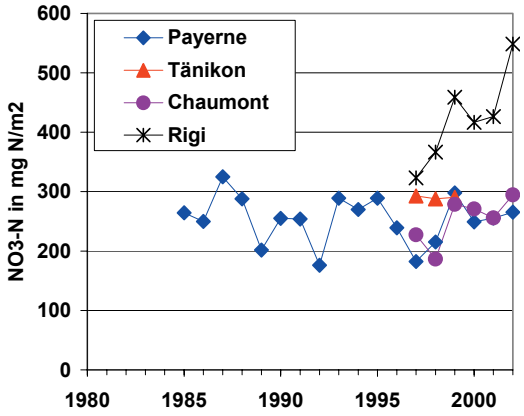


Figure 7b. Annual wet depositions of nitrate 1985-2002 (in mgN/m2).

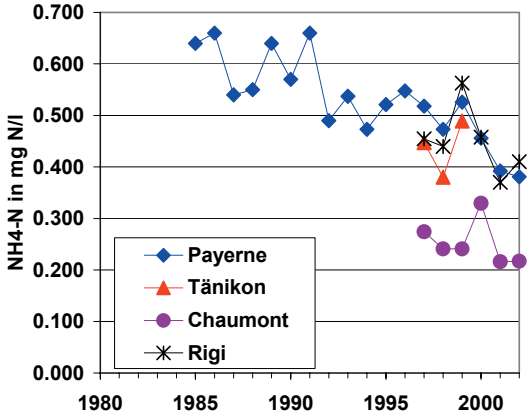


Figure 8a.
Annual mean concentrations of ammonium in precipitation 1985-2002 (in mgN/l).

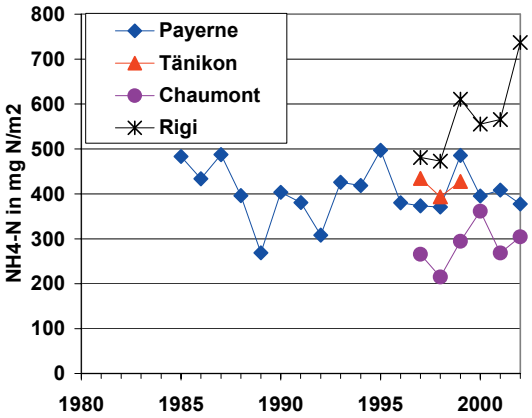


Figure 8b.
Annual wet depositions of ammonium 1985-2002 (in mgN/m^2).

5. Heavy metals

Both lead (Fig. 9a) and cadmium (Fig. 9b) in total suspended particles (TSP, measured before 1997) and PM10 (since 1997) have strongly decreased in the last 15 years. Lead concentrations in particulate matter went back by over 80% in the last 15 years. Cadmium concentrations went back by more than 60%.

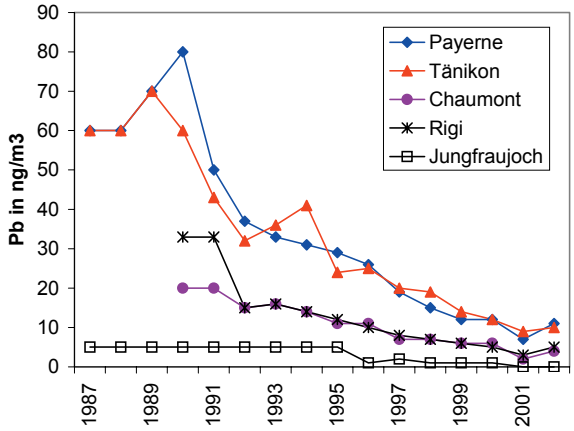
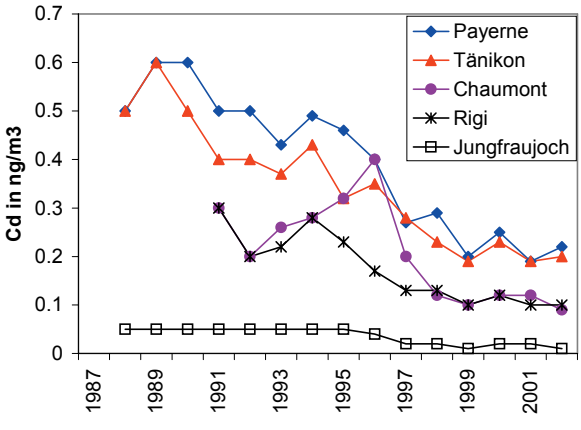


Figure 9a.
Annual mean concentrations of lead in TSP or PM10 at Swiss monitoring sites 1987-2002 (in ng/m^3).



Figur 9b.
Annual mean concentrations of cadmium TSP or PM10 at Swiss monitoring sites. 1987-2002 (in ng/m^3).

6. Ozone

Reliable ozone data are available since 1990. The annual mean of ozone concentration has increased at all sites (Fig. 10a). This increase may be a sign of a larger-scale increase of tropospheric ozone. The number of exceedances of the national air quality standard of 120 µg/m^3 for hourly means (Fig. 10b) has not changed in the last decade. The AOT40-value for forests (April-September, Fig. 11a) and for crops (May-July, Fig. 11b) have not significantly changed since 1990.

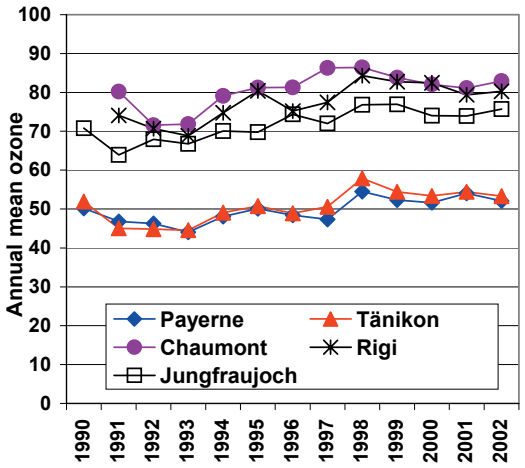


Figure 10a.
Annual mean concentrations of ozone at Swiss monitoring sites (in µg/m^3).

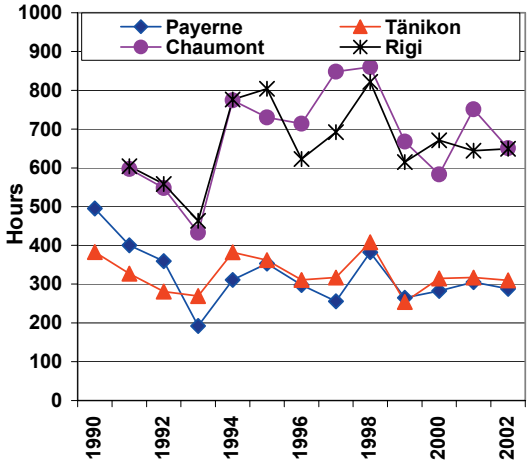


Figure 10b.
Annual number of hours with an ozone concentration > 120 µg/m^3.

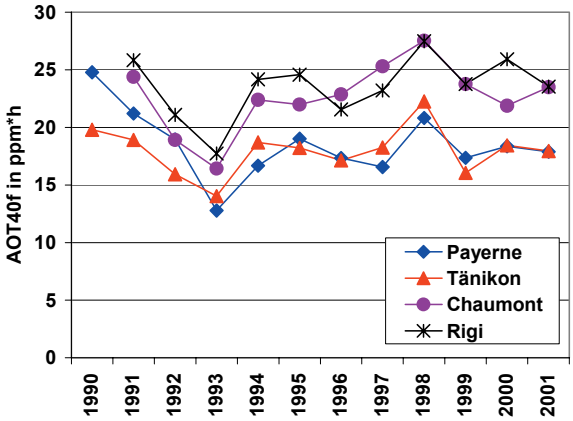


Figure 11.a
*AOT40 values for forests (April – September, daylight hours) in ppm*h.*

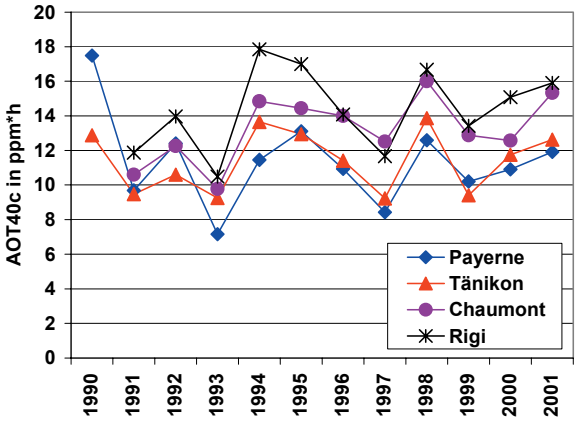


Figure 11.b
*AOT40 values for crops (May –- July, daylight hours) in ppm*h.*

Part II: Particles

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.

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 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) in its version for TSP measurements (sampling flow 40 m^3/h). The sampling inlets for PM10 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 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 Digitel 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 ± 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, TSP and PM2.5 is the design of the sampling heads, the same measurement uncertainty can be assumed also for the TSP and PM2.5 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.
Jungfraujoch (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 2 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 Jungfraujoch, 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 concetrations based on TSP for data before 1997. These estimated PM10 concentrations are given in italics in table 2. Figure 2 illustrates the dereasing trend of PM10 at all Swiss EMEP sites.

Table 2: Annual means of TSP and PM10 in μg/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		*31*		*31*
1982	5.7		37		38		*32*		*34*
1983	8.5		40		40		*35*		*36*
1984	5.9		39		39		*34*		*35*
1985	5.1		44		43		*39*		*37*
1986	4.0		44		43		*39*		*38*
1987	6.7		39		33		*35*		*29*
1988	4.9		32		30		*29*		*27*
1989	4.5		37		36		*33*		*31*
1990	4.3		34		31		*30*		*28*
1991	4.0	22	34		35	*20*	*30*		*31*
1992	3.3	18	31	22	31	*15*	*27*	*19*	*27*
1993	5.5	17	30	16	28	*15*	*26*	*14*	*25*
1994		16	27	16	24	*14*	*24*	*14*	*22*
1995	8.4	16	27	16	26	*14*	*24*	*14*	*23*
1996	5.4	18	31	19	31	*16*	*27*	*17*	*27*
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

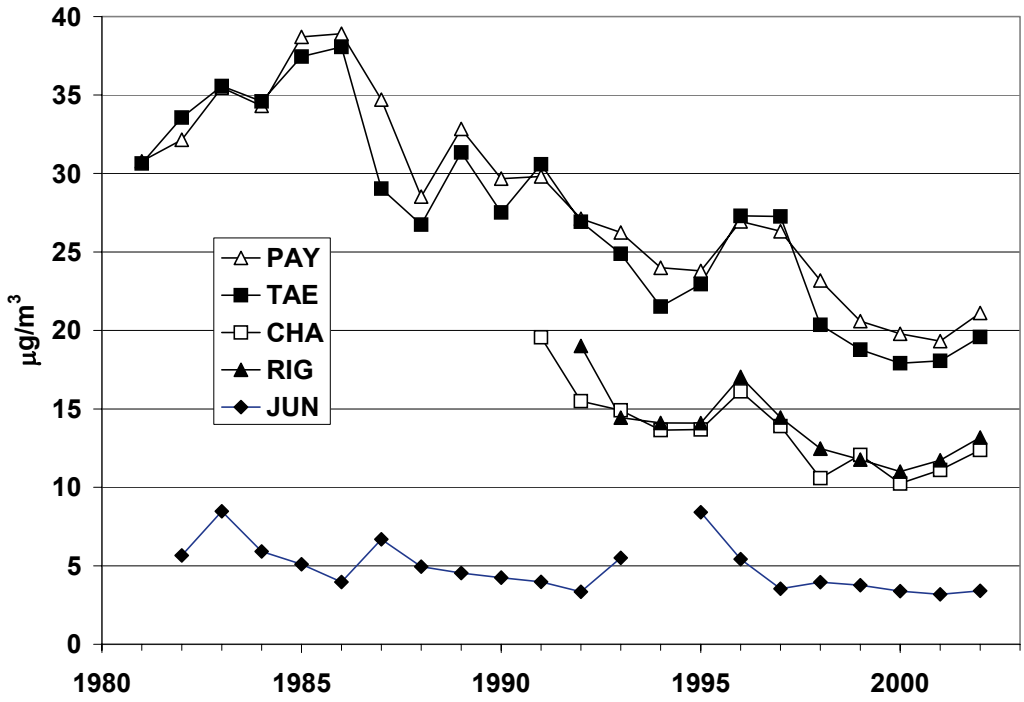


Fig. 2: Long-term PM10 trends at the Swiss EMEP sites

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 and the average ratios PM2.5/PM10. 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 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 4). 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).

A high correlation between the daily values of PM10 and PM2.5 was observed at all sites. With the exceptions of Bern ($r^2 = 0.86$) and Chaumont ($r^2 = 0.85$) r^2 was always ≥ 0.94. 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.
PM10 as well as PM2.5 exhibit characteristic seasonal trends with elevated concentrations during the cold season at all sites, with the exception of the elevated sites of Chaumont and Jungfraujoch. 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 Jungfraujoch 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 Jungfraujoch follow that of the other sites, though on a lower concentration level.
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. 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, typically in the range of r^2 0.7-0.9. 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.
A more elaborated discussion of the seasonal trends and the spatial variations can be found in (Gehrig and Buchmann, 2003).

Table 3
Long term mean values (1998-2001) of PM10 and PM2.5 concentrations and of daily PM2.5/PM10 ratios

	PM10 ($\mu g/m^3$)	PM2.5 ($\mu g/m^3$)	PM2.5/PM10
Dübendorf	22.9	19.9	0.74
Basel	22.5	17.2	0.75
Bern	35.9	20.8	0.59
Chaumont	11.0	7.9	0.75
Lugano	33.0	24.4	0.74
Payerne	20.7	15.1	0.75
Zürich	24.0	18.1	0.75

3. 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.

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, Switzerand.

Gehrig, R., and Buchmann, B., 2003. Characterising seasonal variations and spatial distribution of ambient PM10 and PM2.5 concentrations based on long-term Swiss monitoring data, Atmospheric Environment 37(19), 2571-2580.

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., Cyrys, 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 Massenkonzen-tration (Immission) -
Filterverfahren - Filterwechsler Digitel DHA-80, VDI 2463, Blatt 11.