Report on the Sigma-2 Pilot Project in the Canton of Aargau

Which air quality issues can be addressed by passive dust sampling using a Sigma-2 passive sampler with subsequent microscopic analysis?

Client

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1 Summary

In 2011, the Department of the Environment of the Canton of Aargau decided to conduct a one-year pilot project using the Sigma-2 passive sampler for coarse dust measurements. The project was carried out in cooperation with the German Meteorological Service (analysis) and the company Particle Vision GmbH (concepts and data evaluation).

Between week 41 (the week commencing 10 October) 2011 and week 40 (the week commencing 1 October) 2012, the Sigma-2 passive sampler was used to take particle samples at six sites in the Canton of Aargau. At the Sisseln and Baden sites, these measurements were conducted in parallel to the mass concentration measurements already being conducted with the TEOM FDMS.

The one-year measurement programme reveals that coarse dust pollution levels differ from site to site. Over the course of the year, peak pollution levels – some of them pronounced – occurred at individual sites. The fact that such peaks could be identified is evidence that the Sigma-2 passive sampler is well-suited to monitoring coarse dust levels, e.g. when complaints are made about dust levels or for the purposes of monitoring construction sites in densely populated areas.

The coarse mode fraction (PM10-2.5) allows the PM10 pollution level at any site to be modelled. In terms of its quality, this guideline PM10 measurement is equivalent to the NO_2 passive sampler measurements that have been in use for many years. Thus the Department of the Environment will in future have a cost-efficient method of conducting extended PM10 measurements at its disposal.

Unfortunately, the adhesive particle slide used by the German Meteorological Service is only suitable to a limited extent for scanning electron microscopy, with which a morpho-chemical characterization of the dust samples can be carried out if necessary. Adhesive carbon pads, which are routinely used in scanning electron microscopy, have proven to be one suitable method. These are inexpensive and, with a little extra effort, can be affixed to the adhesive slide used by the German Meteorological Service.

The pilot project was brought to a successful conclusion. All questions relating to methods were answered. As regards the complaint cases, indications of what the sources may be were identified.

2 Introduction

The Canton of Aargau operates a network of three measurement stations that perform continuous measurements. To assess PM10 immission levels, the mass concentration is determined. This parameter provides no information about the sources or chemical composition of the particles, however, which can pose a problem, especially when complaints have been made about dust pollution.

In 2011, Particle Vision approached the Canton of Aargau and proposed that the Sigma-2 be trialled for a more advanced particle characterization. Contact between Particle Vision (PV) and the German Meteorological Service (DWD) presented an opportunity to have the DWD handle the hardware and optical microscopy.

The screening part of this project was not of any particular interest to the DWD because such tasks are a routine part of the work it carries out when conducting spa town certification. What the DWD did find interesting, however, was the chance to analyse samples of special interest using Particle Vision's scanning electron microscopy (SEM-EDX). The opportunity to use the knowledge of the different organizations in one joint project led to the three partners planning a one-year measurement campaign which they carried out in 2011/2012.

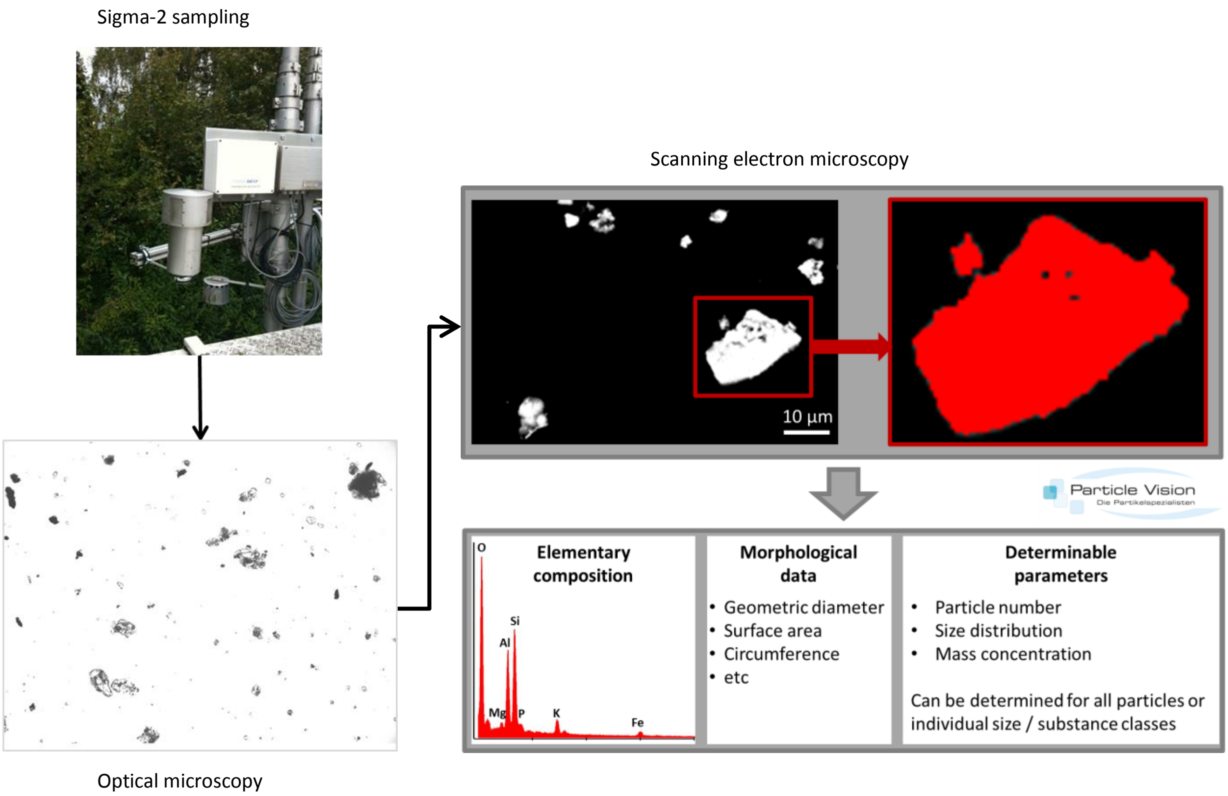


Figure 1: Sampling and analysis procedures

Using the Sigma-2 as the sampling system and then performing optical microscopy, it is possible for the first time to differentiate between different types of dust pollution even during routine measurements according to the following aspects:

- particle size distribution
- mass determination of the different size and particle classes
- rough chemical differentiation into an opaque (anthropogenic) and transparent particle class

For samples which appear to show anomalies, a morpho-chemical analysis can subsequently be performed by means of SEM-EDX. This allow the substances that are causing the anomalies to be identified, which in turn allows conclusions to be drawn about the causal sources.

3 Objectives

The project is intended to provide answers to the following questions:

- When complaints about dust are made, can the Sigma-2 passive sampler be used to compile basic data that can be used to identify the cause(s)?
- Can the Sigma-2 sampling system and subsequent optical microscopic particle analysis be used as a cost-efficient means of determining the annual average PM10 level?
- Can a morpho-chemical particle characterization be carried out on the Sigma-2 slides by means of scanning electron microscopy?

To address the questions listed above, the sites in Table 1 were selected.

Site	Region	Characteristics
Möhlin	Upper Rhine, the three sites are located on a 10 km west-east line	High wood firing density (small and large plants)
Wallbach		Site on the Rhine with high dustfall pollution levels in the past
Sisseln (TEOM)		Industrial site, widespread agricultural use, close to motorway
Bözen	Fricktal, site between the Upper Rhine and the Swiss Plateau	Village character, close to motorway
Baden (TEOM)	Swiss Plateau Urban and rural sites	Urban background, housing estate, zone 30
Hallwyl		Rural site

Table 1: Selection of sites and problem definition

Figure 2: Overview of sites

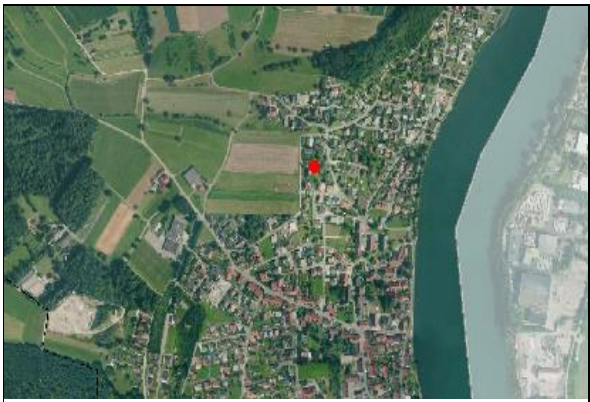
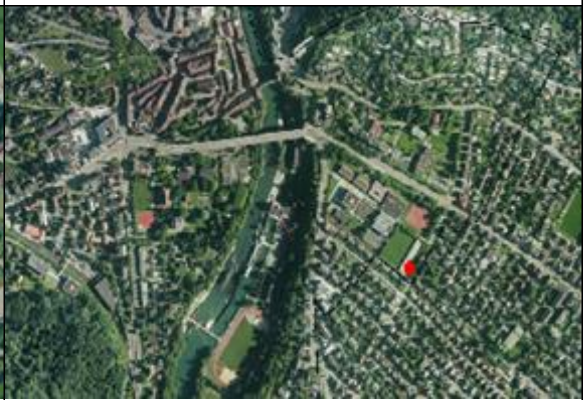
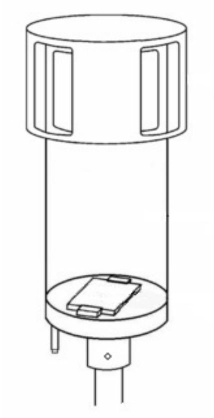
	
Wallbach	Bözen
	
Möhlin	Baden
	
Sisseln	Hallwyl

Table 2: Detailed view of measurement sites

4 Methods

4.1 Passive sampling

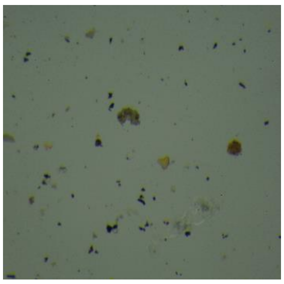
The coarse dust was sampled using the Sigma-2 passive sampler. The particles, which are transported by the natural air flow, enter the wind-sheltered interior of the passive sampler, where particles with a geometrical diameter of between 2.5 and 80 µm are deposited and settle on a transparent slide measuring 65 x 65 mm. The upper side of this slide is covered with roughly 50 µm of weather-resistant adhesive to ensure that the particles are not lost while being transported to the laboratory.



Sampling is carried out over the course of seven days. In isolated cases, e.g. when no personnel is available, other sampling periods have occurred, though these were recorded and taken into account in the respective calculations. The exposed adhesive slide is placed into interim storage in a dry and dark place at room temperature. Every two to three months, the collected slides are taken to the laboratory of the German Meteorological Service.

4.2 Optical microscopic particle analysis

At the laboratory of the German Meteorological Service, the slides are prepared in a dust-free environment at 35- 40°C. Then a square measuring 18 x 18 mm is cut out of the 65 x 65 adhesive slide. This is immersed in an aqueous immersion oil with a refractive index of 1.43. Particle analysis takes place automatically under a transmitted light microscope with 20x magnification. In total, an area of 10 mm^2 is scanned. The 100 obtained images are analysed by a PC-supported image analysis program and the particles are measured.



The particle data obtained in this way are used to calculate the particle number deposition rate and the mass concentration in five size classes in accordance with VDI Guideline 2119-4.

Thanks to the contrast of the particles under the optical microscope, the coarse dust can be divided into two particle classes: transparent and opaque particles.

5 Results

5.1 Coarse dust

The sites show different levels of dust pollution. Sisseln shows the highest and Hallwyl the lowest pollution level.

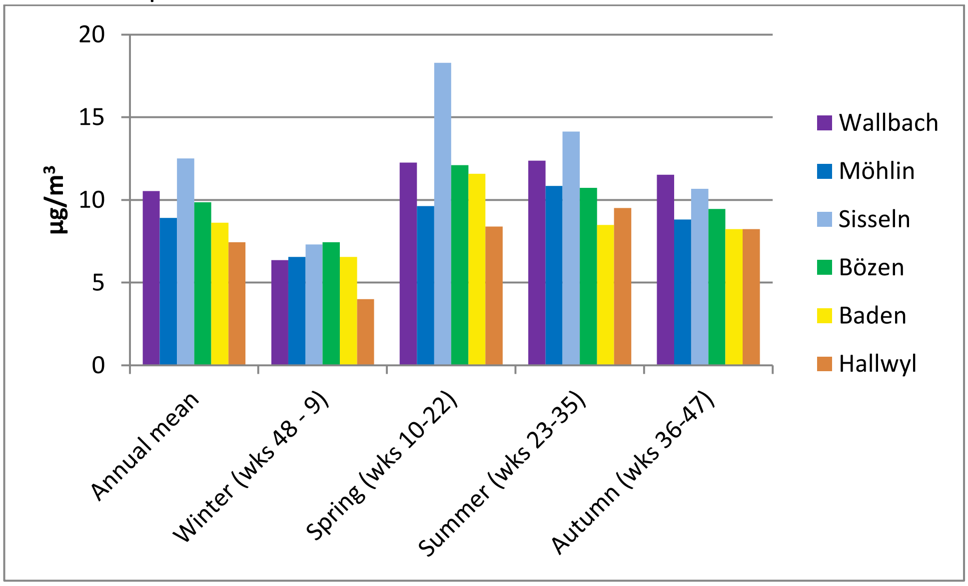


Figure 3: Coarse dust pollution (2.5-80 µm) at the various sites, by season

Coarse dust is the dust fraction that contributes most to the visual soiling of surfaces (coarse particles). Figure 3 shows clearly that the coarse dust pollution varies considerably from season to season. This depends not only on weather conditions (dust is stirred up to a lesser extent on moist, covered or frozen surfaces), but also to a relevant extent on activities (agriculture, construction industry etc.). Because coarse dust settles relatively quickly, the measurement site and its proximity to sources of coarse dust is also an important factor that influences the dust concentration.

It is clearly visible in Figure 4 that the opaque particle class, in contrast to the transparent particle class, remains at a stable concentration level of approx. 2 µg/m^3 throughout the year. As one would expect, the lowest readings are to be found in the summer quarter.

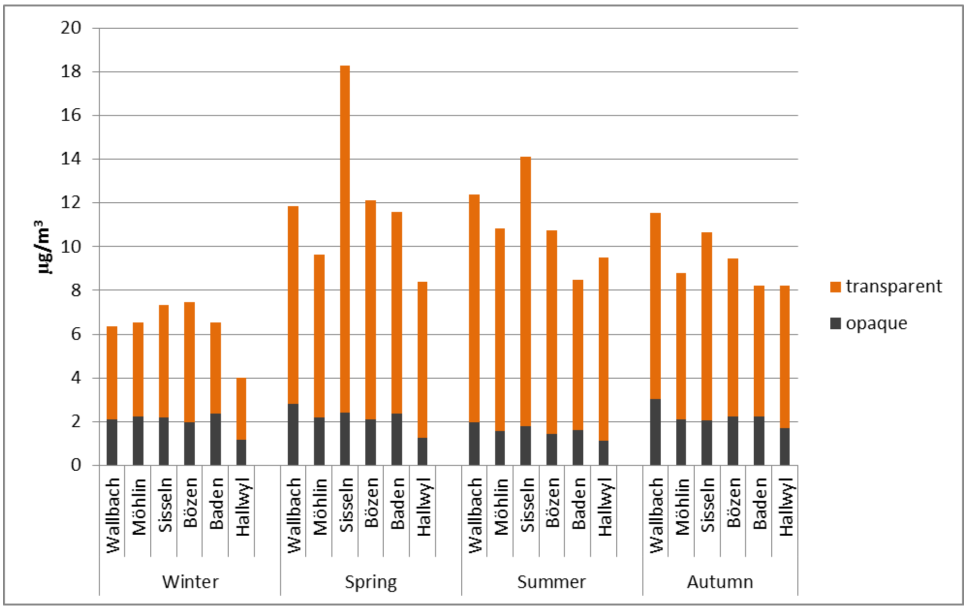


Figure 4: Proportions of opaque and transparent particles (2.5-80 µm) at the different sites, by season

5.2 Transparent particle class

The transparent particle class mainly comprises mineral and biological components (plant residue and spores[1]). These may originate from a wide variety of sources, such as wind erosion, agriculture, part of stirred-up mineral road dust, the construction industry, Sahara dust events, volcanic emissions etc. On average throughout the year, the transparent class accounts for between 75 and 85% of coarse dust.

One typical characteristic of this particle class is the low concentration levels in the winter. This is due to the wet, snow-covered and in some cases frozen surfaces which therefore release fewer stirred-up particles. This effect is magnified by the fact that less intensive construction and agricultural activity (land cultivation) is carried out in the winter.

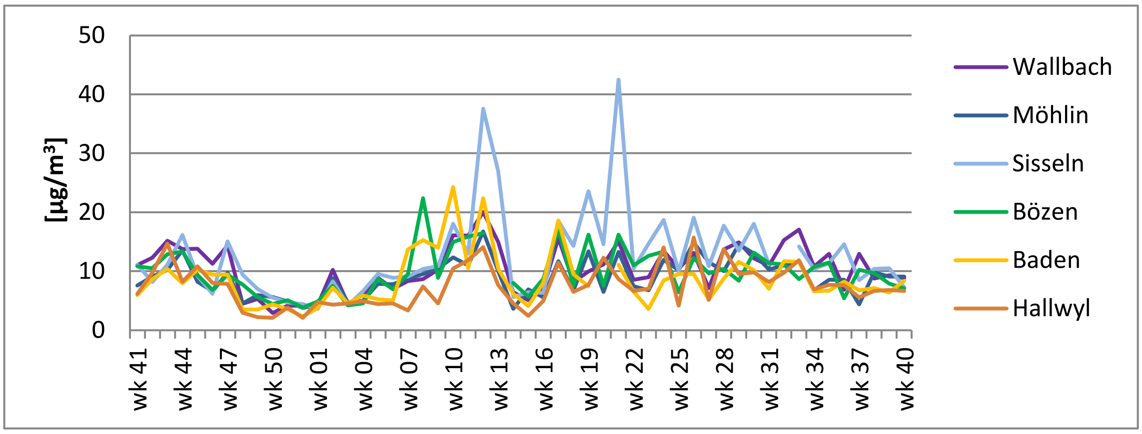


Figure 5: Transparent particle class readings over time (2.5-80 µm)

[1] Pollen is selected by staining and is disregarded

The transparent particle class readings over time show various events that took place at one or more sites simultaneously. If they did not occur or occurred to a lesser extent at the other sites, they are regarded as local or regional events (e.g. at the Upper Rhine sites).

5.3 Opaque particle class

The opaque particles are composed mainly of soot, tyre particles and metal oxides. In terms of health, these are more relevant than the transparent particles, with the exception of asbestos and quartz dust. Contrary to expectations, no increased concentration is evident in the winter quarter. During this period, the readings for Wallbach and Hallwyl are below the annual mean. This suggests that the sources of opaque particles may in some cases be absent in the winter at these sites, or that they then generate additional pollution in the summer.

It should be noted that this method is unable to register primary soot particles with a diameter of 30 nm and their agglomerates with a diameter of up to approx. 1 µm from wood-fired systems. The opaque particles generated by traffic are also stirred up to a lesser extent as a result of the aforementioned moist, snow-covered and frozen surfaces and are thus absent to some extent in the winter readings.

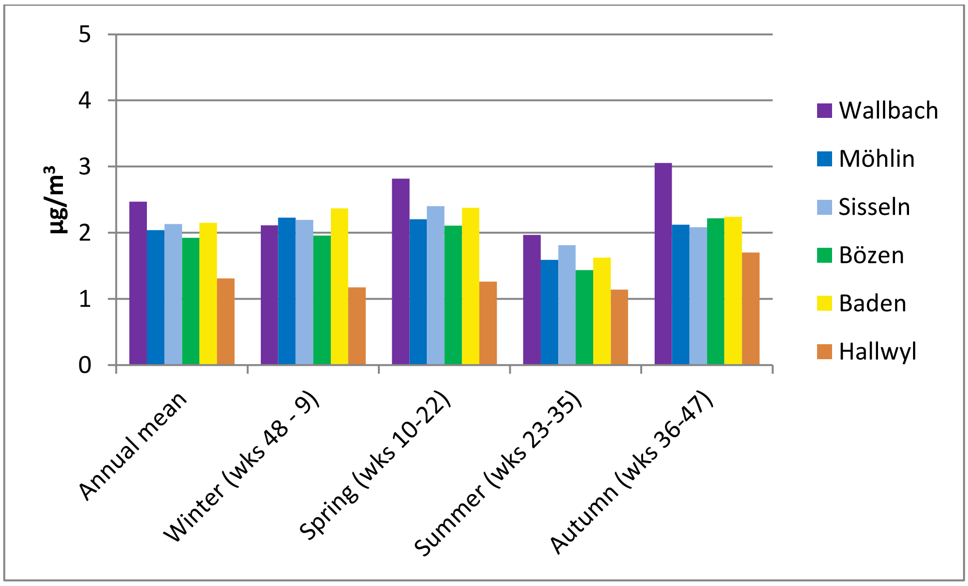


Figure 6: Opaque particle pollution (2.5-80 µm) at the different sites, by season

In contrast to the transparent particle class, the opaque particle class exhibits greater variability between the sites. This means that the pollution comes about as a result of different sources and source intensities.

Unlike the other sites, the Wallbach site shows non-homogeneous readings over time, and tends to be the most polluted site. The period between weeks 41 and 47 stands out: during this period, the opaque concentration at Wallbach is nearly twice as high as at the other five sites. It is not possible to identify the cause from the data available. Possibly the readings are due to a source that was later shut down.

Between weeks 7 and 12, Baden shows atypically high opaque concentrations. This is probably due to repairs that were carried out on the road that runs alongside the

measurement site. The optical microscopy method is unable to reveal whether the concentrations in question result from soot particles from vehicle engines, asphalt particles or metallic particles.
In week 8, Bözen likewise shows an increased opaque concentration, in parallel to the also increased transparent particle class.
The sharp drop in concentrations between weeks 48 and 52 is also clearly visible; this is due to weather fronts passing through and bringing turbulent conditions. The inversion phase in early 2012 can also be seen clearly.

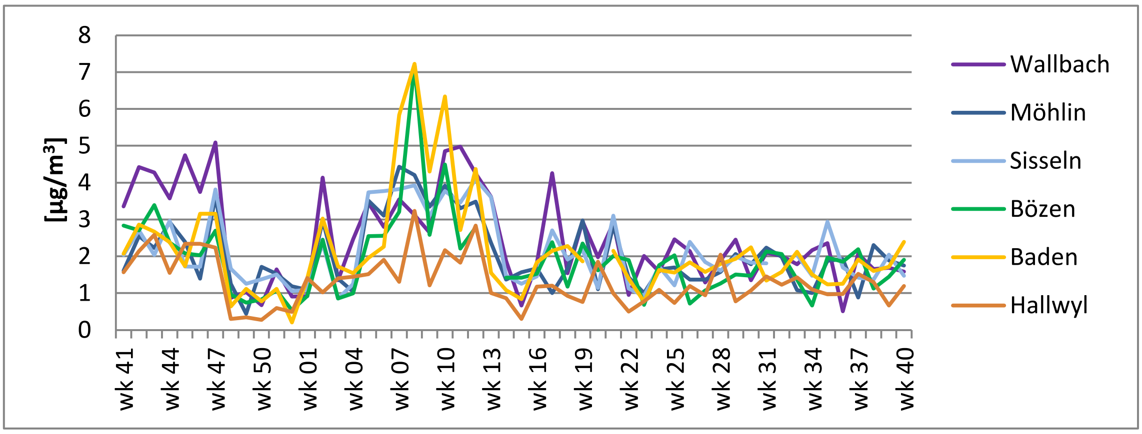


Figure 7: Opaque particle class readings over time (2.5-80 µm)

5.4 Particle number deposition rate

Considerable seasonal differences at the sites can also be observed when looking at the particle number deposition rate. Sisseln exhibits twice as many particles in the spring as in the winter. Hallwyl shows the lowest number of deposited particles of all the sites in the winter (see Figure 8).

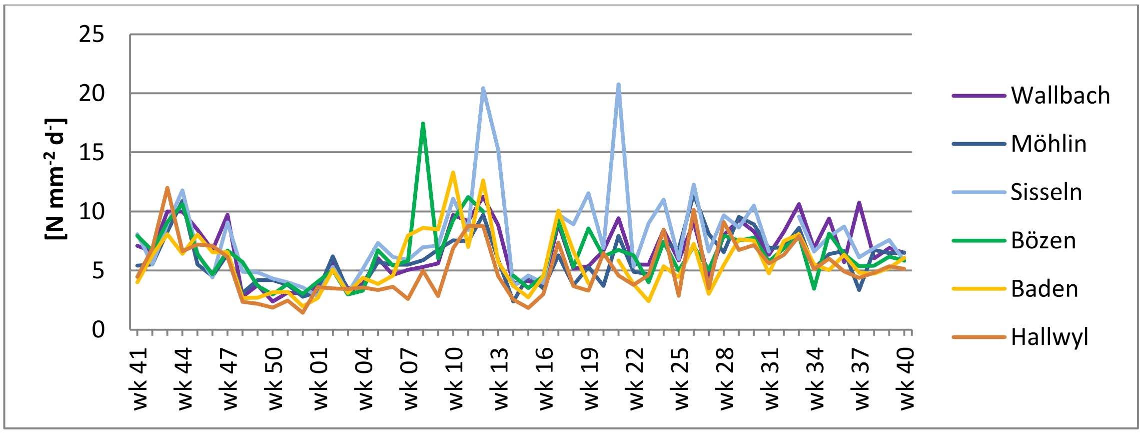


Figure 8: Particle number deposition rate (PM2.5-80) at the different sites

A reading of one particle per square millimetre per day corresponds to a particle number deposition rate of 1,000,000 particles per square metre per day.

The peak readings in weeks 12 and 21 in Sisseln are between 80 and 90% in the transparent particle class. The deposited particles, in other words, are mainly mineral or organic in nature (plant remains and spores).

The particle number deposition rate can also be used as a guideline measurement[2] of surface soiling, however, and could therefore serve in future as a yardstick when complaints are made about dust.

Table 3 shows particle pollution levels in comparison to Hallwyl, an example of a site with low pollution levels (pollution level in Hallwyl = 100%)

Site	Additional pollution Hallwyl = 100%	Particle number deposition rate N/m^2 and Tag
Hallwyl	100%	5,110,000
Baden	111%	5,680,000
Möhlin	117%	5,970,000
Bözen	125%	6,410,000
Wallbach	130%	6,640,000
Sisseln	152%	7,760,000

Table 3: Relative and absolute comparison of the particle number deposition rate for the six sites

[2] This can serve only as a guideline because the deposition conditions in the Sigma-2 housing differ from the normal outdoor conditions because the air flow is reduced inside the housing

6 Comparison with PM10

Sampling with the Sigma-2 followed by microscopic analysis allows coarse dust pollution by particles with a diameter of between 2.5 and 80 µm to be assessed. Because each particle is analysed separately, their size can also be classified.

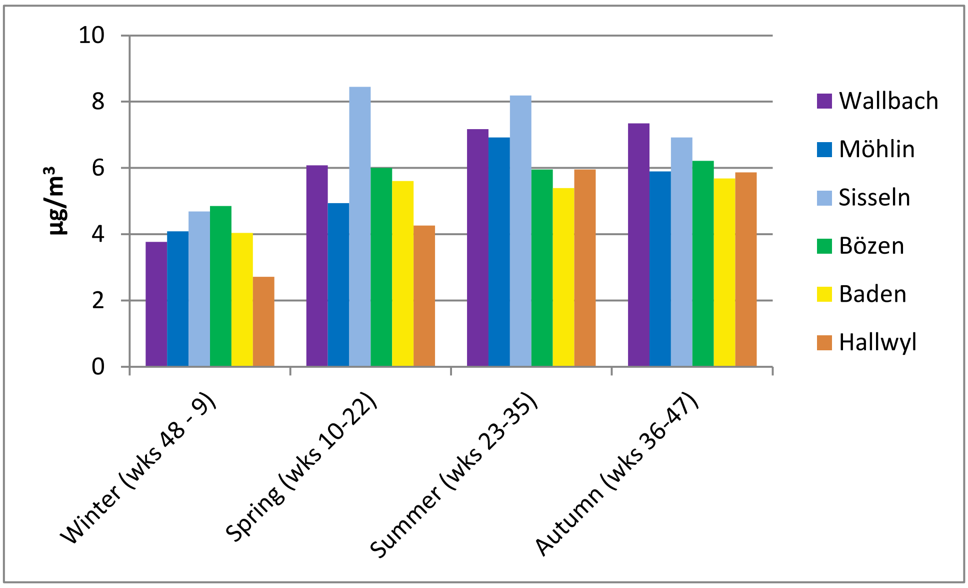


Figure 9: Coarse mode concentration (PM10–2.5) at the different sites, by season

Thus the coarse mode concentration (PM10-2.5) can also be determined, an assessment criterion that is used above all in the USA. Since this size range is part of the PM10, a comparison between the coarse mode and the PM10 concentration can be performed.

For the purposes of the project in question, the Baden and Sisseln sites, at each of which one TEOM FDMS is stationed, was also equipped with a Sigma-2 sampler so that the coarse mode concentration can be compared with the PM10 concentration at these two sites.

The PM2.5 proportion can also be calculated using the following equation:

$$PM2.5_{calc.conc.} = TEOM_{PM10\ conc.} - Sigma\text{-}2_{coarse\ mode\ conc.}$$

Site	PM10 (TEOM) annual mean [µg/m³]	Coarse mode (Sigma-2) annual mean [µg/m³]	PM2.5 (calculated) annual mean [µg/m³]
Baden	18.6	5.5	13.1
Sisseln	20.4	6.9	13.5

Table 4: Calculated PM2.5 annual mean values in Baden and Sisseln

For Baden, this results in a PM2.5 to PM10 ratio of 0.70 and for Sisseln a ratio of 0.66. In 2011, the following ratios were calculated on a random sample basis in the NABEL measurement network:

Measurement site	PM2.5/PM10 ratio
Bern Bollwerk	0.76
Lugano-Università Zürich-Kaserne	0.73 0.73
Basel-Binningen Dübendorf-Empa	0.71 0.72
Härkingen-A1	0.71
Payerne Magadino-Cadenazzo	0.67 0.70
Rigi-Seebdenalp	0.76

Table 5: PM2.5/PM10 ratios at NABEL sites in 2011

The PM2.5/PM10 ratios calculated for Baden and Sisseln correspond very well to the readings calculated in the NABEL measurement network. Although Sisseln is at the lower limit, this fact can be explained by the high proportion of coarse dust in the spring.

Calculating the PM2.5 from the TEOM data and the coarse mode data from the Sigma-2 sampling provides additional information about the development of the PM2.5 over time.

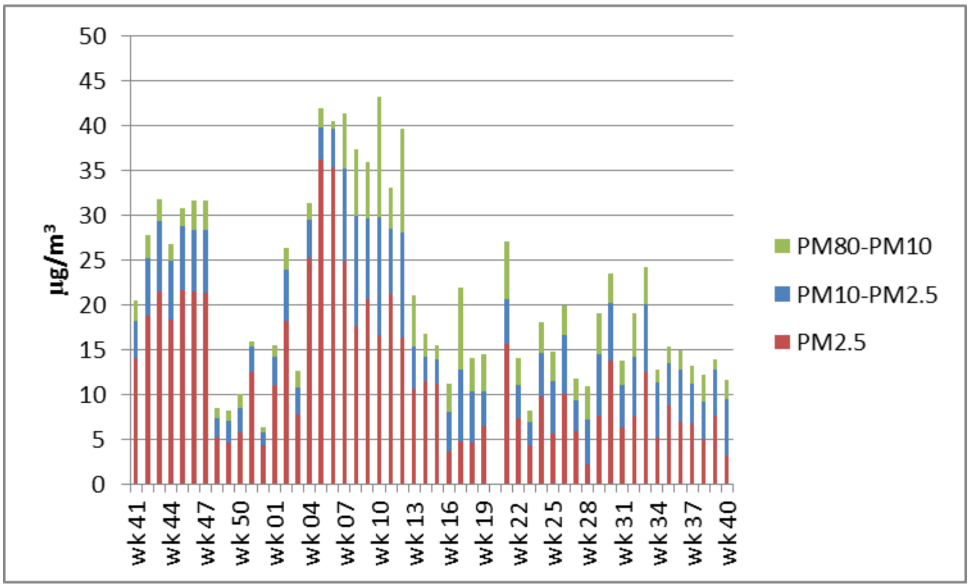


Figure 10: Development of the various size fractions at the Baden measurement site over time. The increased PM2.5 pollution level in the colder months is clearly visible (with the exception of the period of bad weather between weeks 47 and 52)

7 Conclusion

7.1 Economic aspects

The Sigma-2 sampler offers a simple method of conducting passive sampling of particles. The task of replacing the acceptor slides can easily be entrusted to non-professionals. Thanks to its compact size, the system can also be used at difficult sites where there is little space. Overall, in terms of initial outlay and running costs, this method is a more cost-efficient sampling and analysis system than conventional particle measurement systems.

Local management of the Sigma-2 passive sampling system allowed costs to be kept low and a very favourable costs-benefit ratio to be achieved. Thanks to the independent work done by the research partners involved, it was nonetheless possible to obtain some new knowledge. In this context, the collaboration between the canton and the external partners allowed an optimal outcome to be achieved.

7.2 Technical aspects

As a supplementary system, the Sigma-2 can offer additional benefits when it comes to addressing various issues in an existing immission measurement network.

Dust complaints and lawsuits
Using the Sigma-2, it is quickly possible to determine whether the coarse dust pollution level (relevant aspect of surface soiling) is increased or not. Because the sample can also be analysed under a scanning electron microscope, there is a chance that the source(s) can be identified.

Sigma-2 passive sampling measurement network used analogously to NO_2 passive sampling measurement network
It has always been the case in immission monitoring that NO_2 has been measured using analysers and passive samplers. Passive sampling measurements are still conducted to the present day, albeit to a lesser extent, despite the fact that the answers to most questions relating to NO_2 have been known for years and measurements serve “only” trend monitoring purposes.
At a number of selected sites, a Sigma-2 passive sampler could also be used in parallel. This would allow the various immission categories to be better assessed with respect to the still unresolved particle questions. If local personnel were used for management at these sites, the resources that would be freed up as a result could be used to handle the additional work involved in the Sigma-2 measurement.

Additional information about conventional mass concentration determination

There is a general consensus amongst experts that the PM10 measurement alone will not be sufficient in future (see the working group of the Federal Commission for Air Quality [EKL] on the subject). Even today, it is still unclear whether other PM variables such as the particle number concentration, main chemical substances etc. should be measured. Using continuous analysers to determine these substances would entail very high investment costs running into tens of thousands of Swiss francs per measured variable. At the same time, the operating costs would also increase.

The results of the project under review show that use of the Sigma-2 alone is already able to provide a great deal of information in the form of orientation measurements and calculations. One significant advantage of the new methods is that the results of the simple basic analysis allow a decision to be taken about a possible subsequent analysis.

The example of the Sigma-2 measurements in Baden and Sisseln reveals that the existing TEOM PM10 measurement and the parallel Sigma-2 measurement allow conclusions to be drawn about the PM2.5 concentration. This may not appear to be particularly beneficial at first glance. However, this reveals that the primary particles play a subordinate role in an inversion situation and that the secondary particles that are formed from combustion processes account for the main mass proportion. Although this information is not new, it involved a great deal of work and costs in the past to verify it or prove it in laboratory tests. The new measurement method allows the dust composition dynamics to be tracked without excessive effort or cost. This can be an advantage when planning measures and monitoring success.

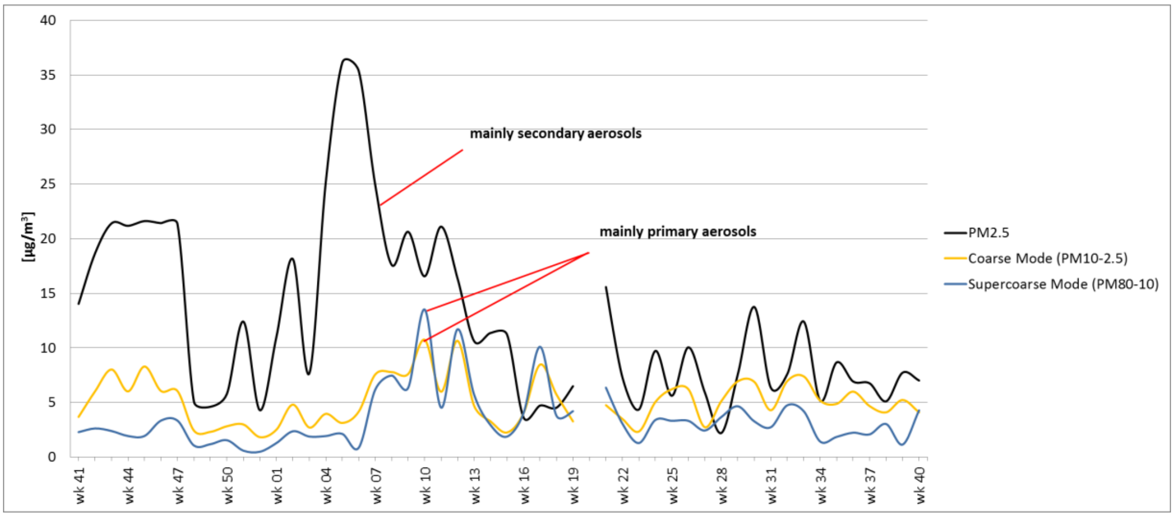


Figure 11: Concentration of the PM2.5, coarse mode (PM10-2.5) and supercoarse mode fraction (greater than PM10) in Baden over time

8 Methodological insights

8.1 PM10 calculation

As already mentioned in Chapter 6, the level of PM2.5 is fairly constant at most of the sites. This means that the PM10 pollution level can be calculated from coarse mode (PM10-2.5) at any site with the aid of a PM2.5 reference concentration.
In the following section, the PM10 pollution level in Sisseln is calculated using the following input data:

- coarse mode concentration from the Sigma-2 measurement in Sisseln
- PM2.5 concentration from Baden as a reference site

Figure 12 compares the calculated and measured PM10 concentrations for the Sisseln site. The X axis shows the calculated PM10 concentration as calculated from the coarse mode concentration in Sisseln and the PM2.5 value calculated from Baden. The Y axis shows the TEOM concentration measured in Sisseln.
There may be various factors which explain the section of the axis at 3 µg/m^3, e.g.:

- measurement uncertainty of TEOM measurements in Baden and Sisseln
- measurement uncertainty of the coarse mode concentration measurement
- more different PM2.5 annual means than shown by the NABEL measurements

The available data are as yet unable to explain the cause.

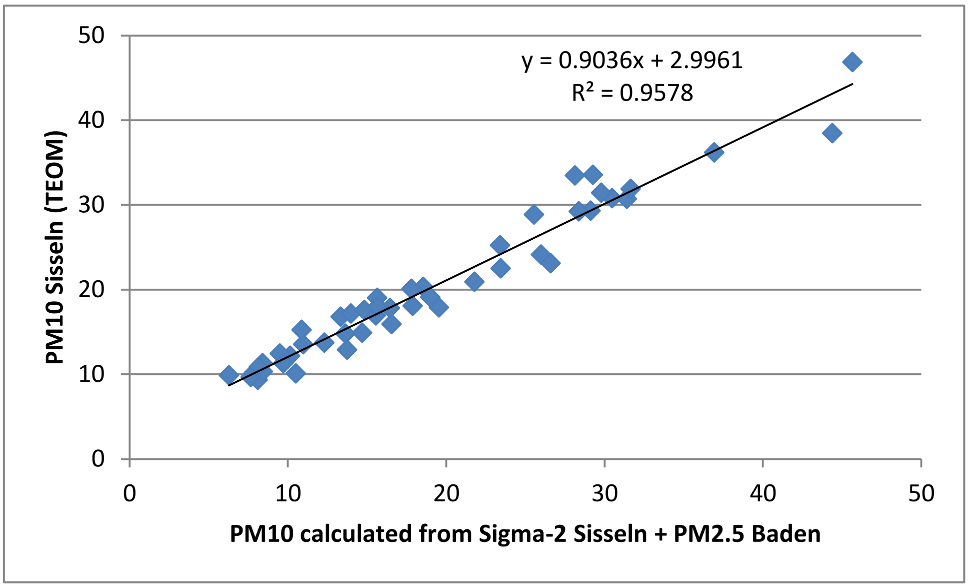


Figure 12: X/Y plot for Sisseln illustrating the Sigma-2 measurement week-by-week in comparison to the TEOM measurement (week 41/2011 – week 40/2012)

Based on the assumption that the annual mean PM2.5 concentration is identical for all sites, the PM10 concentration for all 6 Sigma-2 sites can be calculated from the annual mean coarse mode value using the following equation:

PM10 $conc_{cal.}$ = coarse mode $conc_{meas.}$. + $PM2.5_{conc.cal.}$

The mean value from the Baden and Sisseln measurement stations was used as the PM2.5 reference value for the PM10 calculation in Table 6.

Site	**Coarse mode concentration**	**PM2.5 calculated ($\bar{x}$ Baden, Sisseln)**	**PM10 calculated**	**PM10 measured (TEOM)**
	µg/m³	µg/m³	µg/m³	µg/m³
Wallbach	6.4	13.3	19.7	
Möhlin	5.7	13.3	19.0	
Sisseln	6.9	13.3	20.2	20.4
Bözen	5.7	13.3	19.0	
Baden	5.5	13.3	18.8	18.6
Hallwyl	4.6	13.3	17.9	

Table 6: Results of the guideline PM10 calculation for the 6 Sigma-2 sites and a comparison between the TEOM PM10 measurement and the PM10 calculated from the coarse mode concentration

Showing a deviation of 2% between the calculated and measured annual mean PM10 value for Baden and Sisseln, the correspondence between the two can be described as very good.
This means that the Sigma-2 passive sampler is also suitable for use for guideline PM10 measurements.

8.2 SEM-EDX analysis

The SEM-EDS analysis on the DWD’s adhesive slides has proven to be not ideal because the adhesive layer is too thick and smaller particles sink into it. This can result in focusing problems, and thus in too many “false” particles during the individual particle analysis, which drastically increases the analysis or may even render it impossible.
As a result of this finding, carbon adhesive tabs have additionally been used during sampling since week 25/2011. This allows samples that show anomalies to be subjected to a detailed morpho-chemical analysis without the aforementioned problems.