Advances in long-term soil-pollution monitoring of Switzerland§

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Abstract

The Swiss soil-monitoring network (NABO) was launched in 1984 and comprises currently 105 observation sites covering all characteristic land-use types across Switzerland. So far, the sampling periodicity was 5 y, and the fifth sampling campaign will be accomplished by end of 2009. The concentrations of Cd, Zn, Pb, Cu, Hg, Ni, Cr, Co, and F were measured. The major results and conclusions are: (1) Even topsoils in remote areas are to some extent polluted, mainly by Pb, Cu, and Cd. However, elevated concentrations can also be of natural origin, *e.g.,* for F, Ni, Cr, and Cd. (2) Land use alone is often a rather unreliable indicator to discriminate soil pollution. (3) After the 2nd campaign positive, negative, or no temporal concentration changes were measured. From the 3rd campaign onwards, nonmonotonous (zigzag) evolutions were frequently observed. Therefore, no certified trends can be stated after three measurement campaigns during a period of 10 y. (4) Soil monitoring is an environmental time-series problem. The only way to detect reliable signals and trends earlier is to improve the overall measurement quality (precision and bias) and to shorten the measurement periodicity. (5) The causes of temporal soil concentration changes are complex and result from natural processes, anthropogenic processes, and methodological artifacts. Hence, not all soil concentration changes are due to anthropogenic inputs. Based on the state-of-art of our experience, some basic methodological requirements and recommendations can be supported for a "good soil-monitoring practice": (1) Assurance of long-term continuity and consistency under changing boundary conditions as site conditions, methodologies, *etc.* (2) Implementation of a scientifically and politically appropriate spatial and temporal measurement resolution. (3) Long-term assessment of reliability (quality assurance) by adequate quantification of precision, bias, and confidence intervals along the whole measurement chain. (4) Documentation of all potentially relevant boundary conditions by suitable metadata. Only soil-monitoring results meeting these requirements are fit to support political decisions.

Key words: soil monitoring / temporal change / time series / soil pollution / inorganic pollutants / trace metals

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1 Introduction

The concern of beneficial as well as detrimental human impacts on biogeochemical cycles in the environment is the basic motivation and legitimation of long-term chemical soil monitoring, defined here as the measurement of temporal chemical changes in soil. Compared to the environmental media like air, water, and organisms, the residence time of chemicals in soils is generally by far the longest, and this may lead to ecotoxic relevant accumulations in soils. Since soils are not only chemical sinks but also sources of nutrients and pollutants for the food chain, soils are an essential environmental monitoring media for sustainable food production and food safety. Despite all possible measures to impede or reduce soil-pollution sources there will always remain uncontrolled emissions. The achievements of political and technical measures to reduce pollutant inputs in soils can finally only be correctly assessed by reliable direct soil monitoring. "Monitoring soils over medium- to long-term is the only way in which the magnitude and direction of changes in soil properties arising form human activity and natural pedogenic processes can be measured" (*Billet*, 1996).

The history of chemical soil monitoring begun in the 19th century by long-term agro-ecosystem experiments (*Rasmussen* et al., 1998). Their initial purpose was to increase and maintain productivity, later combined with sustainability. Around 1960, it shifted progressively to environmental impacts of pollutants—not at least due to sewage-sludge application (*e.g., McBride*, 2003; *McGrath* et al., 1995). Currently species adaptation to climate change is becoming an issue (*e.g., Aitken* et al., 2008; *Jentsch* et al., 2007). A fundamental change was initiated when soil monitoring remained not only confined to controlled field experiments but soil-monitoring networks were established with spatially scattered sets of observation sites under different environmental and management conditions. This was especially the case when forest decline became an issue in the early 1980s (*e.g., Johnson* and *Siccama*, 1983; *Krause* et al., 1986). It was probably the concern that after the forest dieback the soils might "die", which gave at least implicitly rise to many regional and national soil-monitoring networks mainly in Europe (*Winder*, 2003; *Morvan* et al., 2008). But most existing national soil-

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monitoring networks have undertaken only a single sampling campaign so far and thus remain soil-status inventories at present (*Saby* et al., 2008). Screening the literature, we found several publications of temporal trace-metal soil concentration changes from two observations between 1 and 40 y (*Siccama* and *Smith*, 1980; *Billet* et al., 1991; *Friedland* et al., 1992; *Pichtel* et al., 1997; *Egli* et al., 1999; *Meneses* et al., 1999; *Lark* et al., 2006). Longer and/or more intense time series are known from long-term agro-ecosystem experiments (see *Rasmussen* et al., 1998) and related to seasonal cycles (*Heuvelink* and *Webster*, 2001) of soil temperature (*e.g.,* Forbes, 1846, in *Heuvelink* and *Webster*, 2001), soil moisture (*e.g., Robock* et al., 2000) and soil solution (*e.g., Graf Pannatier* et al., 2005) at selected sites. At our knowledge—additionally supported by the 35 contributions on soil monitoring at the EUROSOIL 2008 conference (*Blum* et al., 2008)—the Swiss soil-monitoring network (NABO) is the only one to have published concentration changes of inorganic pollutants from three different time observations on a national scale so far (*Desaules* et al., 2006). Furthermore, a complementary study of time series with up to six measurements analyzing the causes of the concentration changes was conducted on a subset of the NABO sites (*Desaules* et al., 2004).

The main objective of the present paper is to trace the advances in inorganic soil-pollution monitoring of the Swiss soil-monitoring network illustrated by selected results over 20 y (1984–2004) with recommendations for less advanced soil-monitoring networks. Emphasis is put on the step-by-step evolution of findings. Soil-monitoring data of organic pollutants are not included in this paper, since up-to-date only status studies for PAH, PCB, and PCDDF (*Bucheli* et al., 2004; *Schmid* et al., 2005; *Desaules* et al., 2008) have been published for the NABO network.

2 Outline of the Swiss soil-monitoring network (NABO)

In Switzerland, the environmental legislation (*SAEFL*, 2001) demands the running of a reference national soil-monitoring network—known as NABO ("NAtionale BOdenbeobachtung")—to evaluate the status and long-term trends of soil pollution concentrations due to inputs of anthropogenic chemicals. Focus is given to early detection and forecasting as a warning system to reduce risks in decision making for natural-resource management and as a tool to assess the achievements of political and technical measures to reduce soil pollution. The NABO network was launched in 1984 and comprises 105 long-term-observations sites of the major land-use types throughout Switzerland (Fig. 1) which were resampled every 5 y so far (*Desaules* and *Studer*, 1993; *Desaules* and *Dahinden*, 2000; *Desaules* et al., 2006;). Comprehensive information about NABO and further publications can be found under www.nabo.admin.ch

2.1 Site selection and characteristics

The basis for the selection and distribution of the initially 102 and 105 observation sites since 1995 was a matrix of 20 soil geographical regions and 13 land-use types weighed according to hypothetical pollutant inputs. The land use of the sites

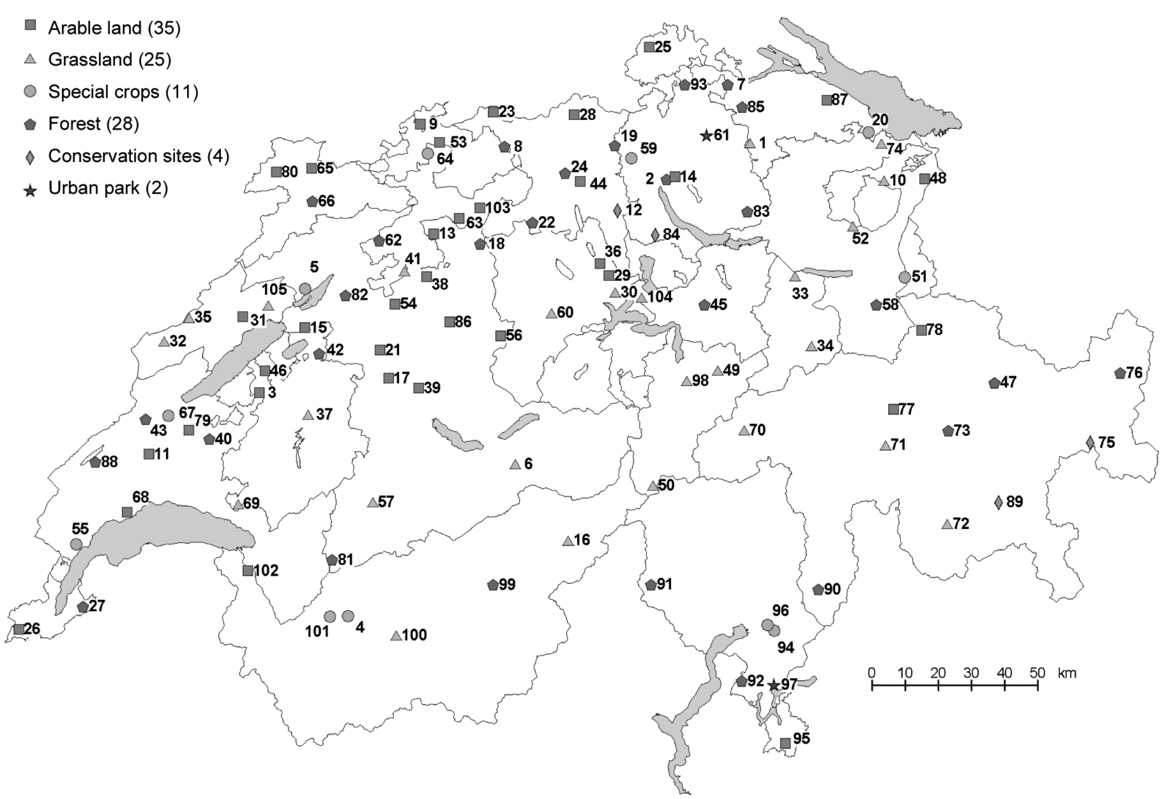

Figure 1: Geographical distribution and land use of the 105 long-term soil-pollution observation sites of the Swiss soil-monitoring network.

are: arable land 35, vineyard 4, orchard 3, horticulture 4, intensive grassland 8, extensive grassland 17, conservation sites 4, deciduous forest 12, coniferous forest 16, and urban park 2. The range of altitude is between 210 and 2400 m asl (median 565 m asl). The mean annual temperature and precipitation range from –1.6°C to 11.4°C (median 7.9°C) and from 550 to 2330 mm (median 1128 mm). Topsoil pH (0.001 M $CaCl_2$) ranges from 2.9 to 7.6 (median 5.3) and total organic C from 1% to 43%.

2.2 Soil sampling, physical sample preparation, and storage

At each site—from a located sampling surface area of 10 m × 10 m—four replicate composite soil samples of 25 stratified increments each were taken by a steel gouge auger with an inside diameter of 3 cm from the surface down to 20 cm soil depth. The samples were transported by car at ambient temperature to the laboratory, oven-dried at 40°C, separated by hand from stones and other nonsoil material, passed through a jaw crusher (Retsch®) and sieved through a 2 mm nylon-mesh sieve. The samples were stored in glass containers in the dark at temperatures < 20°C. The stored laboratory samples were mechanically mixed in a Turbula® immediately before taking test portions for chemical analysis. A manual provides a full description of the procedure (*Hämmann* and *Desaules*, 2003). The duration of a regular sampling campaign to visit all sites of the NABO reference network is 4 y and the sampling periodicity of the sites 5 y. In consequence, the dates of the initial four measurement campaigns were 1985/89, 1990/94, 1995/99, and 2000/2004.

2.3 Chemical analysis

The eight trace metals Cd, Zn, Pb, Cu, Hg, Ni, Cr, and Co were detected in the filtrate of 10 g test portion boiled 2 h in 2 M HNO_3 solution with a weight ratio of soil test portion to solvent of 1:10. At the first campaign (1985/89), Pb, Cu, Zn, Ni, and Cr were detected by FAAS and Cd, Co by GAAS with Zeeman compensation. During the second campaign (1990/94), Pb detection changed to GAAS with Zeeman compensation in 1993. At the third campaign (1995/99), all trace metals except Hg were detected by ICP-MS. Hg was detected by cold-steam AAS with D_2 underground compensation in an external laboratory. The element-specific determination limits were defined by the lowest used standard multiplied by the dilution factor and were not stable over time.

The analytical results were expressed in mg (kg dw [dry weight])$^{-1}$ with reference to sample aliquots dried at 105°C.

2.4 Measurement quality

The quality-control and quality-assurance measures along the measurement chain were the following:

(1) Soil-sampling and physical-sample-preparation protocols with detailed information on location, sampling plan, land use and management, environmental conditions, sampling device, soil depth, sampling support, increment materialization (completeness), estimated soil characteristics, packing and transport conditions, drying, crushing, sieving, splitting, storage containers, and a follow-up of sample weights at different conditions. A detailed description with forms are given in *Hämmann* and *Desaules* (2003) and can be downloaded (www.nabo.admin.ch > Quality > Sampling monitoring forms).

(2) Chemical analysis: The internal analytical quality as precision and stability were based on long-term control charts of two internal-standard soil samples with contrasting properties and replicate analysis of roughly 10% of the soil samples within and between batches. To cope with the temporal inconsistencies of the chemical analysis, the samples of the first campaign (site reference samples) were reanalyzed simultaneously with the corresponding samples of each following campaign. The original analytical results were corrected on the basis of the corresponding reanalyzed samples of the first campaign. This should allow for avoiding analytical drift and ensuring analytical stability.

The external analytical quality as comparability and bias were assessed by the regular participation at a proficiency testing program—adapted to the legislation of soil protection in Switzerland (*SAEFL*, 2001)—which comprises 12 samples per year (www.nabo.admin.ch > Quality > Proficiency testing).

(3) Site measurement-repeatability precision: It includes the site- and element-specific precision under staggered repeatability conditions (n = 4) for the whole measurement chain from soil sampling, over physical sample preparation to chemical analysis.

2.5 Data presentation and evaluation

For reasons of relative and ecotoxic comparability and evaluation, the results of trace-metal concentrations and their temporal changes were normalized in percent of the respective guide values (GV) set as 100%. Guide values (Tab. 1) are legally based soil-quality benchmarks, which can be considered as general precautionary upper limits of negligible risks (*SAEFL*, 2001).

3 Results and discussion

3.1 Baseline measurements of the first campaign (1985/89)

An overview of baseline trace-metal concentrations of the NABO network is given in Tab. 1 together with the corresponding guide values.

The frequent concentrations (10th to 90th percentile) of the NABO-network baseline measurements are in good agreement with the compilation of frequent concentrations from thousands of other available data from Switzerland (*Keller* and *Desaules*, 2001; *Desaules* et al., 2006). View to the limited number of exceeded guide values, the frequent concentrations up to the 90th percentile are considered to be of negligible risk. This is in agreement with the purpose of the long-term soil-monitoring reference network. The reasons for

Table 1: Baseline trace-metal topsoil (0–20cm) concentrations (2 M–HNO_3 extraction) of the Swiss soil-monitoring network with 102 observation sites of different land use.

Trace metals	Min. / mg kg^{-1}	10th perc. / mg kg^{-1}	Median / mg kg^{-1}	90th perc. / mg kg^{-1}	Max. / mg kg^{-1}	Guide values[a] / mg kg^{-1}	Exceeded guide values / *n*
Pb	11	16	24	38	133	50	6
Cu	3	6	18	35	860	40	6
Cd	0.06	0.11	0.23	0.49	1.83	0.8	5
Zn	22	35	53	89	145	150	0
Hg	0.02	0.06	0.10	0.19	0.41	0.5	0
Ni	5	6	22	40	324	50	5
Cr	6	13	25	38	534	50	1

[a] precautionary upper limits of negligible risks (*SAEFL*, 2001)

exceeded guide values may be attributed mainly to traffic and urbanization (Pb), agrochemicals (Cu), long-range atmospheric pollution (Pb, Cd), and parent material (Ni, Cr, Cd, Pb) (*Desaules* and *Studer*, 1993; *Desaules* et al., 2006). Relatively high Ni and Cr concentrations are related to parent materials containing ultramafic rocks as serpentinite (*Gasser* et al., 1995), and high geogenic levels of Cd are mainly related to oolithic limestones (*Benitez Vasquez*, 1999).

The example of the geographical distribution of the baseline topsoil and subsoil Pb concentrations of the NABO network (Fig. 2) gives evidence of the generally higher Pb concentrations in the topsoil compared to the subsoil. This is an indication of soil pollution, provided bulk density is similar which is not the case for soils with an organic layer under forest (sites no. 45, 83, 84, 88, 92). Other sites with exceeding guide values are two urban parks (no. 61, 97), permanent grassland near to a road (no. 33), pasture land also used as an army-training site (no. 52) and somehow astonishingly, a remote site in a national park (no. 75) at an altitude of 2400 m asl. The initial hypothesis of long-range transboundary soil pollution from N Italy could only partly be confirmed. A subsequent isotopic study did find a geogenic and pedogenic evidence (*Hansmann* and *Köppel*, 2000). The apparent tendency of relatively high Pb concentrations in NE Switzerland and S of the Alps should not be overinterpreted.

The site classification by land use (Fig. 3) reveals that within-class variation is generally larger than between-class variation—except for the Cu concentrations in the four vineyard sites from 223 to 860 mg kg^{-1}—and thus, land use alone is often not a reliable indicator of soil pollution.

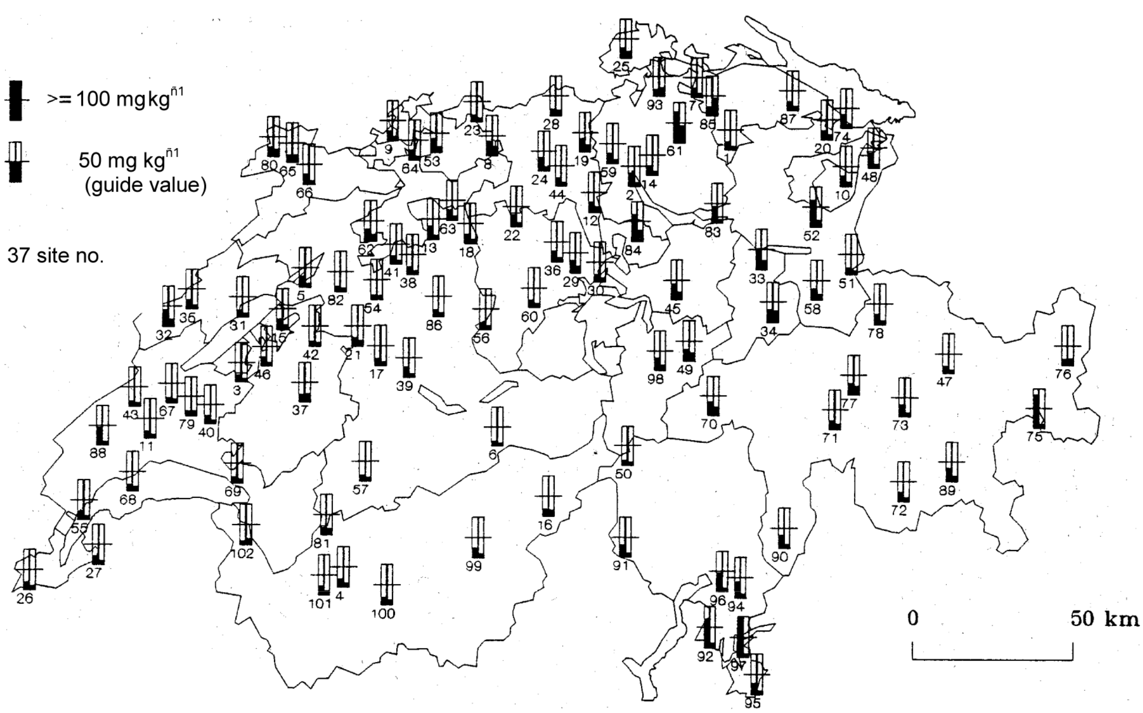

Figure 2: Geographical distribution of the baseline Pb topsoil (0–20 cm) and subsoil concentrations of the Swiss soil-monitoring network relative to the guide value (*SAEFL*, 2001: precautionary upper limit of negligible risks).

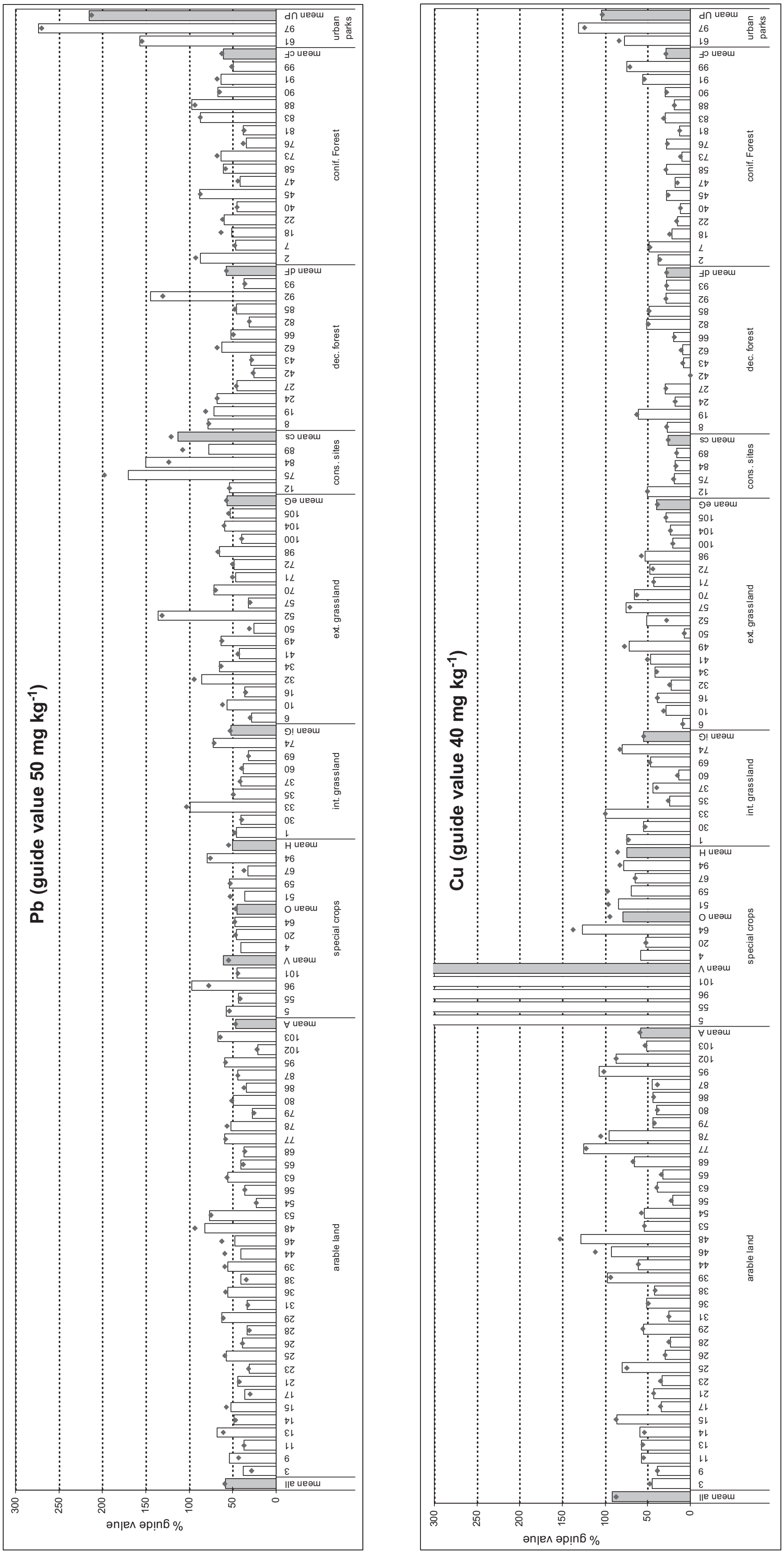

Figure 3: Changes of normalized Pb and Cu topsoil (0–20 cm) concentrations after 5 years of the Swiss soil-monitoring network sites grouped by land use (gray bars are mean values).

3.2 Measurements of the second campaign (1990/94) after 5 years

The concentration changes of Pb and Cu after 5 y (Fig. 3) show, that at site level besides no changes, positive as well as negative changes occurred which practically outweighed the mean changes.

After 5 years, 87% of the sites showed already at least one statistically significant positive or negative concentration change for one of the nine investigated elements (*Desaules* and *Dahinden*, 2000). But the summary presentation for all sites and the no-till sites sampled at different soil depths (Fig. 4) confirm that the changes were mostly < 5% of the respective guide value and quite symmetric. It is worth to note that no clear evidence of greater changes could be observed at the no-till sites as it was expected due to less mechanical pedoturbation and related dilution effects of pollutant inputs on these sites. An exception was Pb at the shallow 0–5 cm sampling depth which is not robust due to the surface ruggedness. With regard to positive as well as negative concentration changes after the second measurement campaign within 5 y, it is argued that pollutant inputs cannot be the only cause of measured changes.

The guide value–normalized concentration changes after 5 y for all investigated elements on nine selected sites (Fig. 5) demonstrate clearly that quite different patterns of changes occur even for the same land-use and management types and support the hypothesis that other factors than only land use may considerably influence soil concentration changes.

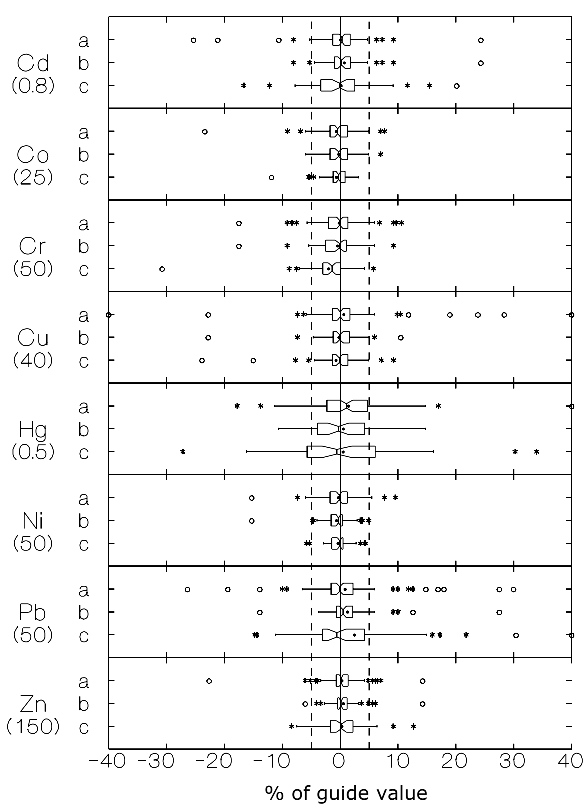

Figure 4: Changes of normalized topsoil concentration of eight trace metals after 5 years of the Swiss soil-monitoring network: (a) for all sites 0–20 cm (n = 100), (b) for no-till sites 0–20 cm (n = 54), and (c) like (b) but 0–5 cm; (0.8) = guide value in mg kg^{-1}.

3.3 Measurements of the third campaign (1995/99) after 10 years

The third measurement campaign revealed that nonmonotonous (zigzag) evolutions of temporal concentration changes are common, indicating that the dynamics of changes is not linear and, thus, reliable trend statements are not yet possible. The examples of Pb and Cu (Fig. 6) exhibit obvious soil concentration changes only for some land-use types. But a closer look at the number of sites (n) by category and implausible evolutions of the temporal changes as for extensive grassland and coniferous forest (Pb), special crops (Cu), and urban parks (Pb, Cu) ask for closer investigation. The more the sites are pooled, the more the positive and negative concentration changes neutralize each other, raising the question whether the expression of mean concentration changes make sense in soil monitoring.

3.4 Time series with more than three measurements

At first sight, implausible significant decreases of soil concentrations after 5 y (*Desaules* and *Dahinden*, 2000), poor agreements between directly measured concentration changes and indirect balances (*Desaules* et al., 2003) and nonmonotonous (zigzag) temporal evolutions of concentration changes (*Desaules* et al., 2006) gave rise to the already mentioned complementary time-series study to investigate the causes of soil concentration changes (*Desaules* et al., 2004). The results of Pb and Cu on a grassland site in Fig. 7 show that the range of the more frequent measurements of the complemantary study over 3 y (n = 6) can be even more important than for the regular NABO measurements (n = 3) over 10 y. This astonishing findings suggest that important short-term causes (sudden effects or events) as measurement bias especially due to sampling under different soil conditions (moisture and/or compaction) and soil dynamics (*e.g.,* pedoturbation, erosion) must be important artifacts.

Short and extensive soil-monitoring time series can hardly confirm significant trends, they can only be identified and certified after sufficiently long and intense measurement series (Fig. 8). The longer the measurement periodicity, the longer it takes to detect a significant trend. It is therefore a contradiction to declare early detection as a major purpose of soil monitoring to decision makers and to recommend a measurement periodicity of 10 y or more. The measurement noise of environmental studies is generally considerable and the noise-to-signal ratio small (*Beard* et al., 1999). To reliably quantify the noise, the measurement periodicity has to be shortened to generate numerous measurements under different soil conditions in the shortest time possible. To reduce the noise, the measurement quality starting from soil sampling to chemical analysis has to be improved. Within the noise, no interpretation of single measurements is possible.

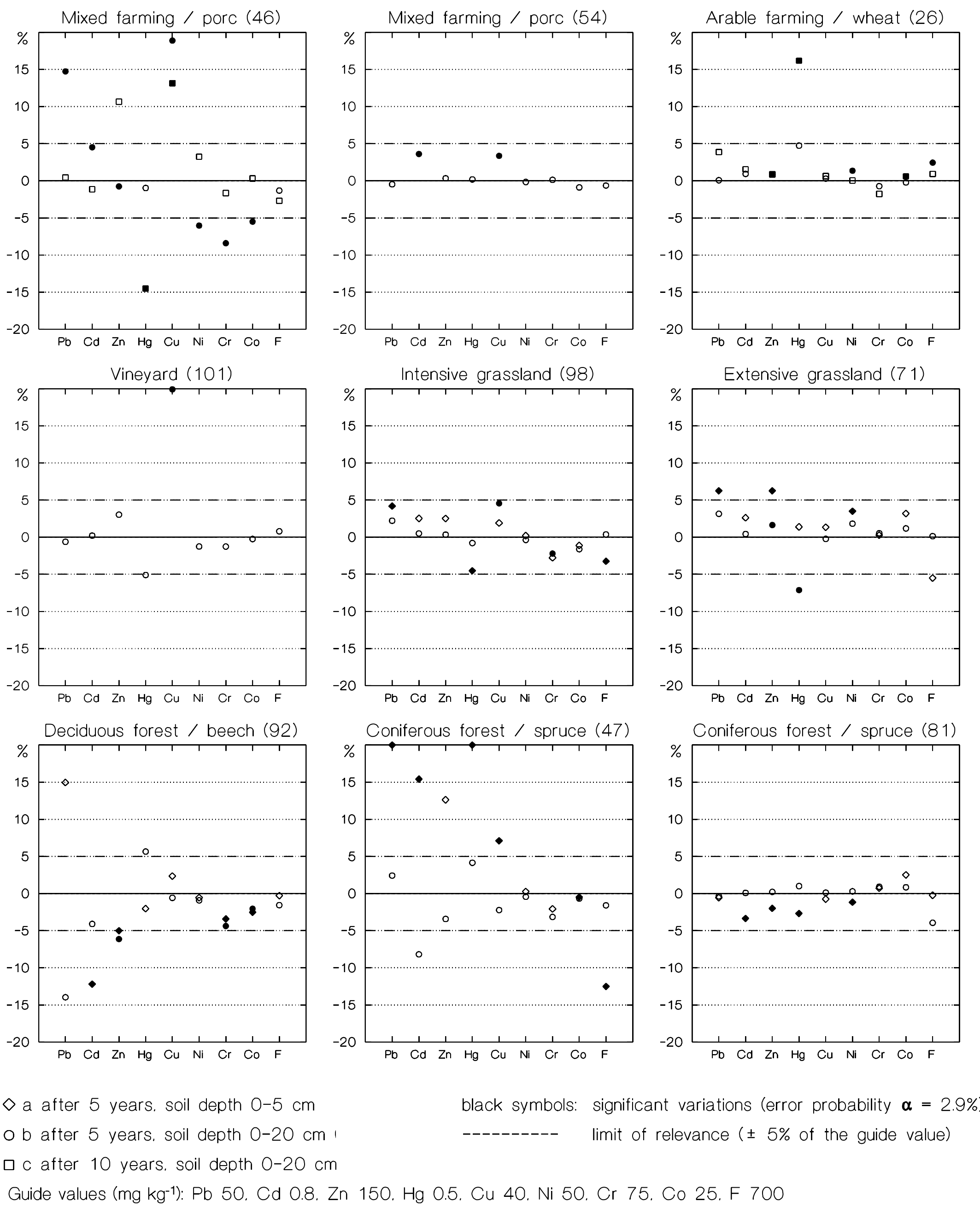

Figure 5: Pattern of normalized concentration changes for nine inorganic pollutants on selected observation sites of similar and different land use.

It may be instructive to discuss the reasons why it took us 10 y to recognize that soil monitoring is basically an environmental time-series problem as, *e.g.,* climate change, and has to be treated as such. The answer is that with a chosen measurement periodicity of 5 y we had to wait 10 y for the third measurements to become aware that nonmonotonous (zigzag) evolutions were common. But why didn't we realize the problem earlier by analogy with other already existing environmental time series? Here, the embarrassing answer is that we were dazzled as obviously all other regional and national monitoring networks known to us. How could this happen? We erroneously rejected the analogy with other environmental time series, because they were basically all cyclic by nature, and we overestimated the measurement accuracy looking only at precision and not considering bias properly, as it is still very common in field experiments and environmental studies. And where finally may be the reason of these fundamental errors? We believe to have found it in the historic evolution of soil monitoring which roots in controlled field experiments. This hypothesis is supported by the fact that statistical methods developed for controlled field experiments are commonly used in soil monitoring for which they are often not suitable due to lack of control.

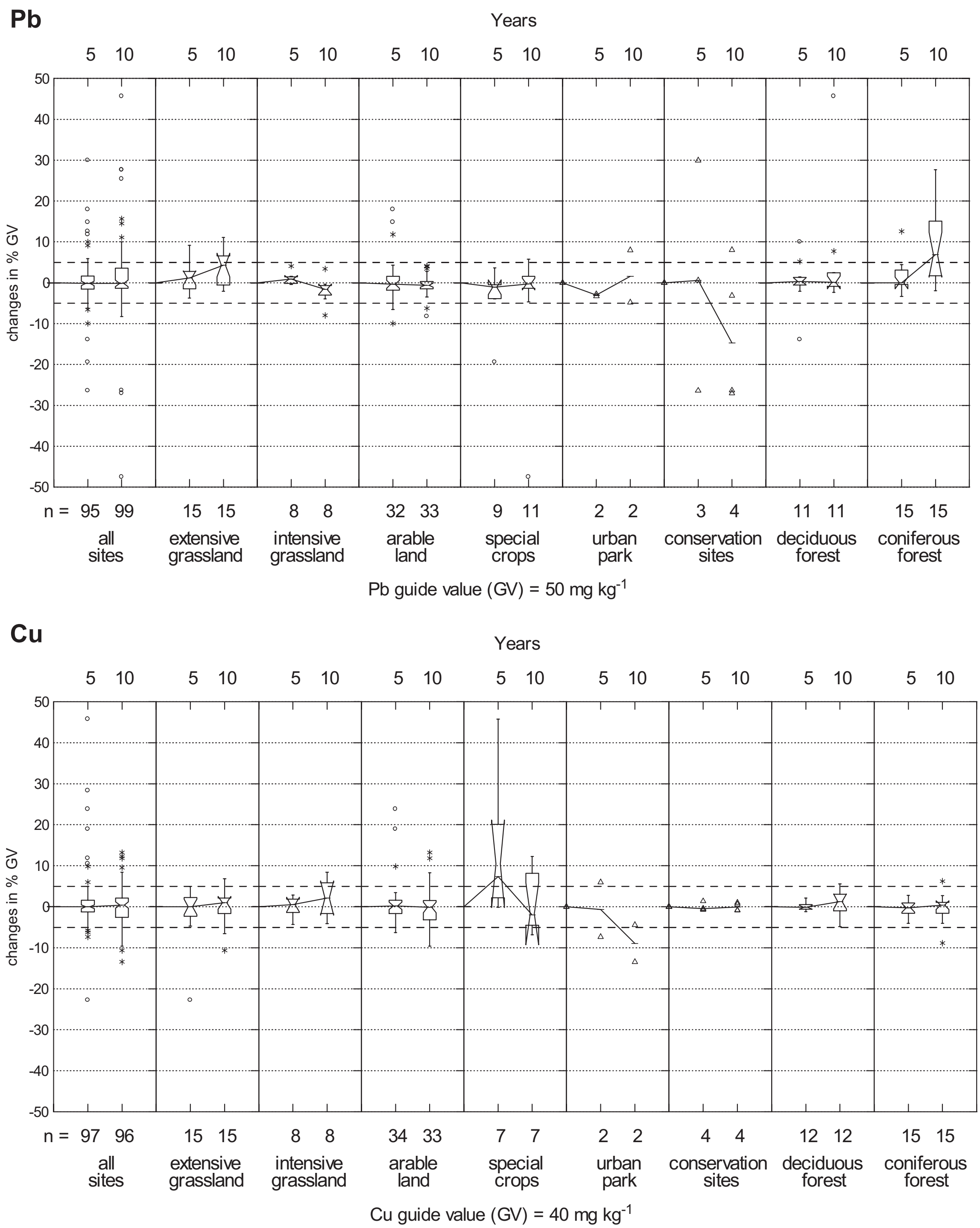

Figure 6: Changes of normalized Pb and Cu topsoil (0–20 cm) concentrations after 5 and 10 years of the Swiss soil-monitoring network grouped by land use.

4 Conclusions and recommendations

The soil-monitoring findings and conclusions are closely related to the number of soil-measurement campaigns and are summarized as follows:

(1) So far the representativity of the NABO network can be considered as satisfying since the frequent concentrations (10th to 90th percentile) of the baseline measurements are in good agreement with the compilation of frequent concentrations from other available data from Switzerland (*Keller* and *Desaules*, 2001). The concentrations in mineral soils are widely dominated by their geogenic background, and land use alone is therefore generally not a reliable indicator for elevated concentrations except for Cu in old vineyards.

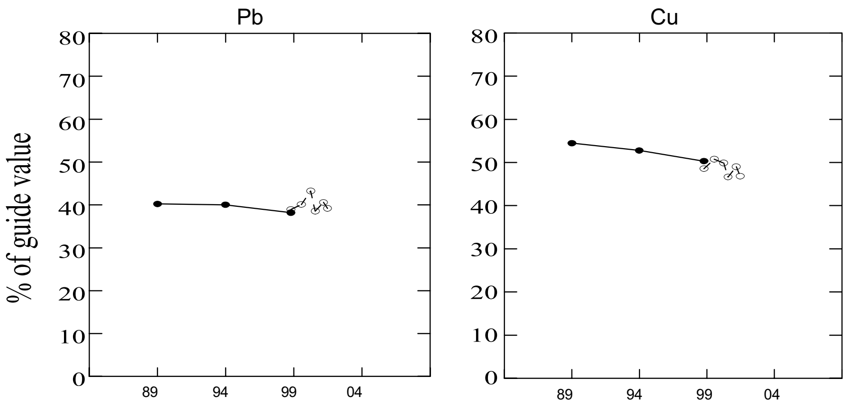

Figure 7: Comparison of normalized Pb and Cu topsoil (0–20 cm) concentration changes on the grassland site (no 30) of the NABO network with three measurements (full line) and a complementary study of three years with six measurements (dashed line).

(2) The second measurements after 5 y revealed a high proportion of observation sites (87%) showing statistically significant concentration changes, however, they were not yet considered as relevant. This applies to both investigated soil depths of 0–20 cm and 0–5 cm. The positive and negative changes indicate that there must be other causes involved than only pollutant inputs. The more the sites are pooled the more the mean changes become small. Pooling of observation sites with different environmental and management conditions is a considerable source of artifacts. For this reason, statistical methods which are successful for well-controlled field experiments are often not appropriate for soil-monitoring sites embracing a great variety of environmental and land-use conditions.

(3) The third measurement campaign after 10 y confirmed the findings of the second campaign and revealed furthermore that nonmonotonous (zigzag) evolutions of temporal concentration changes were common. This is an indication that the dynamics of changes is not linear and thus, reliable trend statements are not yet possible after three measurements.

(4) Examples and scenarios with more than three measurements reveal that soil monitoring is an environmental time-series problem and has to be treated as such. This means that possible trends can only be identified and certified after sufficiently long and intense measurement series. The only way to detect reliable signals and trends earlier, is to improve the data quality (precision and bias) along the whole measurement chain and to shorten the measurement periodicity to reduce and quantify the noise with acceptable reliability.

(5) Measured concentration changes can only be correctly assessed and evaluated if their processes (causes and effects) are sufficiently understood. Soil monitoring is thus a basic tool in ecosystem research and process understanding and far off from a simple routine exercise.

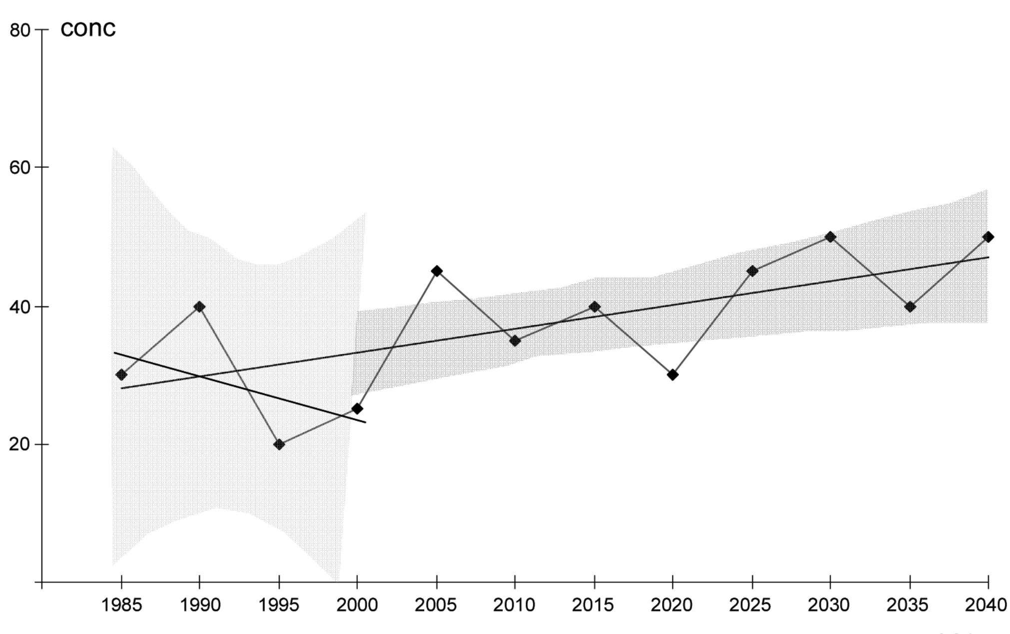

Figure 8: Soil concentration time series and linear regression lines with 95%-confidence intervals of four measurements extended to a scenario of 12 measurements.

To be reliable and politically credible, soil monitoring should be based on a scientifically sound "good soil-monitoring practice". Based on the state-of-art of our experience, we can contribute the following major requirements and recommendations:

– Assurance of long-term continuity and consistency under changing boundary conditions as site conditions, methodologies, *etc*.

– Implementation of a scientifically and politically adequate spatial and temporal measurement resolution.

– Long-term assessment of reliability (quality assurance) by appropriate quantification of measurement bias, variance, and confidence intervals.

– Documentation of all potentially relevant boundary conditions by appropriate metadata.

Political decisions are a delicate balancing act to maximize economic innovations and simultaneously minimize the risks for the population and the environment (*Harremöes* and *Gee*, 2002). An important scientific and political challenge of soil monitoring is to quantify and minimize measurement uncertainty as much as possible, in order that precautionary measures—as a buffer for risks—are neither too relaxed nor too severe. View to the enormous possible ecological damages and economical costs due to false monitoring signals, a reliable soil monitoring is a very cost-saving public investment in the long term. The required degree of reliability is determined by the politically accepted risks. Environmental-monitoring networks which are not able to adequately quantify their degree of reliability are therefore not a reliable support for political decisions.

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