

## 8 Synthesis

The aim of this thesis is to assess how patterns in the geochemical soil composition can be used to distinguish natural variability from anthropogenic impact both in attribute and geographical space. The patterns found in this study are related to processes determining variability in soil composition, including variability caused by human influence. The many soil quality indicators presented can be used as a first initiative for sustainable management of soil quality.

In the following I will evaluate the methods used, the achievements of this study, and give an overview of the implications of the results, together with possible future applications.

### 8.1 Methods used

The methods used in this study are in general based on state of the art analytical methods combined with well established statistical techniques from the field of geochemical surveying and exploration. With the use of X-ray fluorescence spectrometry (XRF) and inductively coupled plasma mass spectrometry (ICP-MS) a broad range of elements could be analysed on many samples. For future comparison with data obtained from regular environmental Dutch soil surveys, the ICP-MS samples were leached using a hot aqua-regia solution. Both multi-element techniques were specifically chosen to broaden the analytical focus with major components, trace elements, and the rare earth elements (REE). This wide range of components can be obtained with minimal extra effort compared to a smaller selection such as environmental priority metals. This resulted in a better understanding of not only the human effect but also of the natural composition and relations in the soil.

By using both techniques some overlap between measured components occurred, which was useful to obtain extra information about analytical quality and bias. Due to the different approaches of the techniques, partial aqua-regia leach for ICP-MS and total analysis for XRF, some basic mineralogical information was also obtained.

Using well established statistical techniques for data analysis has the advantage that possibilities and limitations are well known and that many examples are available. This makes the techniques more transparent to the researcher and easily explainable to users. However, more complex methods have not been avoided, such as robust principal components analysis, so long as they provide a true advantage over standard techniques.

The methodology of this study incorporated several techniques to obtain information about the accuracy, variability and bias on analytical and sampling scales and, if applicable, methods to reduce noise and bias. It is essential to be able to distinguish the contribution of geochemical soil variability from that due to chemical analysis and sampling.

## 8.2 Achievements of this study

It was expected from the history of Zeeland that the soil contained slightly elevated concentrations of so called “heavy metals” (Cd, Cu, Pb, and Zn) compared to pristine background concentration levels. Also persistent organochlorine pesticide residues were expected to be present. This is confirmed in this study.

It was shown that the spatial variability of the heavy metals, Fe, and calcite related components, has a relatively large contribution of local scale variance. This means that for these components small scale variability is an important source of the total variation and compositing for these elements is considered useful. For most other elements the regional variance predominates, reducing the need for compositing.

The main part of this study comprises the geochemical characterisation of the province. The main hypotheses were that 1) based on insight into the natural variability the anthropogenic imprint could be recognised and 2) the subsoil is suitable for obtaining a geochemical baseline as a reference to calculate the anthropogenic impact. An additional question studied was if the imprint results in spatially distinct areas where specific human processes have had a relevant contribution to the soil composition.

To test these hypotheses a dataset of 31 chemical components was selected from the overall dataset of 83 components, based on precision, accuracy and completeness. For this dataset a measurement was available for each variable at each location and for each depth, topsoil and subsoil. The dataset still comprised major, minor, and trace elements, providing a reference for a wide range of components. It formed the core for the geochemical assessment of the soils of Zeeland.

To determine the anthropogenic imprint it is necessary to have a reference, or baseline, comprising the natural soil composition. For the Netherlands it was believed that no good baseline existed (van der Meent et al., 1990), and reference values like the legal threshold values are not considered representative for natural soil composition. When a true baseline is available, deviations from this baseline can be attributed to separate processes. Human processes, for example the local application of fertilisers or atmospheric deposition of contaminants from remote areas, will in general lead to increased concentration levels, or enrichment, of certain components. Using a baseline, such enrichment can be determined and the extent of the influence of the associated process can be assessed.

A good method for many elements to obtain a baseline is the use of the relation with  $\text{Al}_2\text{O}_3$ . The correlation of an element with  $\text{Al}_2\text{O}_3$  can indicate if the element is uninfluenced by human processes. Overall higher correlations with  $\text{Al}_2\text{O}_3$  in the subsoil in Zeeland confirm the hypothesis that the subsoil can be used as a pristine, non-influenced, reference. Using

a regression model based on the subsoil relation of an element with  $\text{Al}_2\text{O}_3$ , the extent of the enrichment of the topsoil can be quantified both in absolute terms and as ratio with the subsoil baseline. The enrichment can be assessed using individual enrichments of soil components or by the average enrichment, called the contamination index, of all components with elevated concentration. Despite the presence of enrichments for many elements the legal threshold level is not exceeded, thus the soils may be regarded as legally clean with regard to inorganic components. This implies also that the legal threshold values are not a suitable reference for natural background soil concentrations. Compared to all other techniques illustrated in chapter 5, the subsoil regression baseline method gives the best insight into 1) the geological/pedological pattern of geochemical variability, 2) random natural variation, and 3) the anthropogenic imprint on Zeeland soils.

The use of the above described baseline provides enrichments for individual elements based on the covariability with only one other variable. To fully understand the soil factors that determine the variability in soil composition a more multidimensional space must be considered. Multi-element covariability (principal component analysis) revealed four main factors that can be related to clay, calcite, silt and anthropogenic processes. The natural clay and calcite related processes comprise the largest part of the overall soil variability. The anthropogenic factor explains only a small part of the overall variability emphasising that these anthropogenic processes are really an imprint on the overall natural variability. Yet, for individual elements such as Sn, the anthropogenic factor may explain up to about 40% of the variance.

The regional patterns of both the contamination index and the scores on the anthropogenic factor show many similarities. However, since the anthropogenic factor explains the variability caused by human processes based on the total soil composition while the contamination index is an average of ratios from a few bivariate relations, I prefer the former. Nevertheless, enrichments or the contamination index can be quantified as concentrations of individual components or average of more components, making it easier to compare them with normative values and values from other areas.

When the samples in attribute space are grouped, i.e. based on their similarity with respect to interactions between the processes distinguished, no spatial regions emerge where samples are discriminated solely on the basis of anthropogenic enrichment. This suggests that the extent of the human imprint on the Zeeland soil, is not related to certain regions but rather is distributed diffusely over the province.

Using the techniques and experience from the former chapters, chapter 7 gave an assessment of DDT residue levels based on separate datasets provided by the local authorities. Compared with other areas, the DDT residue concentrations in Zeeland are such that there is no reason to believe that the DDT contamination is more severe than can be expected of an agricultural area. The extent of the values compared to uninfluenced areas is about a factor 2 to 10 higher, indicating that values for Zeeland must be regarded as enriched compared to the non-natural ubiquitous DDT residue background.

Unfortunately, a regional assessment of DDT residue values is hampered by the large contribution of small scale (sampling) variance. This means that for a regional overview the

observed variance is largely influenced by local scale noise. Together with the clear differences between the sub datasets, this implies that the regional variability should be judged with caution, despite the fact that large scale features are still visible.

### **8.3 Implications**

The collection of soil quality data over time is a prerequisite of sustainable management. With a dynamic assessment the direction and magnitude of change can be evaluated (Wienhold et al., 2004). This study showed that with the use of state of the art analytical techniques and well established methods from geochemistry, a profound insight can be obtained in a single survey into the natural soil composition and how it is influenced by human processes. This both provides methods to obtain a reference for natural soil composition and to quantify the anthropogenic imprint. It is shown in this study that the framework of legal limits does neither provides such a reference nor is able to detect subtle anthropogenic enrichment. This, in a way, is remarkable considering that this framework is also used for the building material decree and related to soil pollution risk maps that should prevent spreading of contaminants. I therefore suggest that references for soil composition should be derived from (local) baselines rather than legal limits. When obtaining such a baseline, quantification of the variability due to sampling and analytical procedures, and methods to reduce or compensate bias between sub-surveys are as important as the baseline itself. If multi-element techniques are used, as most laboratories do, focus should not only be on environmental priority components but also on components which might provide information about natural geochemical soil composition and variability.

Soil pollution risks appear to be, as defined in the Dutch normative framework, mainly determined by DDT residues. Unfortunately, the on average low concentrations in soil, complex analytical determination, and high small-scale variability result in large uncertainties concerning the regional variability of this component. Consequently, to obtain a more trustworthy georeferenced overview of DDT residue levels, more effort aimed at reduction of variance and bias should be put in the assessment of this component.

The question remains what “soil quality” should mean for the Dutch situation and which indicators to use. The definition of soil quality as given by Karlen et al. (1997): “the capacity of a specific kind of soil to function, within natural or managed boundaries, to sustain plant and animal productivity, maintaining or enhance water and air quality, and support human health and habitation”, clearly implies not only a characterisation, but an evaluation against certain criteria as well. The so called “soil quality maps”, as used in the Netherlands to facilitate spatial development, clearly do not provide information about soil quality as envisaged by Karlen et al. (1997), despite their name. Therefore, one of the goals to reach sustainable soil management should be a reconsideration of what is understood by “soil quality”. Only then it is possible to define which soil quality indicators should be assessed. The reference data and methods provided in this study could be a starting point for obtaining such indicators, finally leading towards sustainable soil management.