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Interpretation of integrated monitoring data gathered at the Lheebroekerzand in the Netherlands.

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This investigation has been performed by order and for the account of the Directorate General for Environmental Protection, Division Climatical Change and Industry and the Director Environmental Risks and Safety Division, within the framework of project 607165 Integrated Monitoring Area Lheebroekerzand (formerly 673710, 259101 and 607160), that is part of the UN-ECE International Cooperative Programme on Integrated Monitoring on Air Pollution Effects.

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67-68	Library RIVM
69	Bureau Reports Registration
70-100	Copies for participants ICP-IM
101-110	Bureau Reports Sales

Contents

ABSTRACT

In 1993 the Dutch monitoring station Lheebroekerzand became part of the network of the International Co-operative Programme on Integrated Monitoring on Air Pollution Effects. The contribution of the Dutch monitoring activities was set up according to the Manual for Integrated Monitoring and was executed under supervision of the Laboratory for Ecotoxicology of the National Institute for Public Health and the Environment. From 1993 to 1999 a database was build up with data collected in the Lheebroekerzand. Data from studies carried out in the Lheebroekerzand before 1993 were obtained from co-operating organisations and institutes or found in literature.

The first objective of this study is to detect and estimate trends in the extensive amount of monitoring data of the chemical, physical and biological variables through time. The second objective is to determine whether correlations between biological and chemical monitoring data are present. Only biological variables showing a trend through time, and chemical variables related to the environmental themes acidification, eutrophication, desiccation and the presence of heavy metals are examined.

Results of the trend analysis of the biological variables show that there is no indication for a degradation of the bird, leafminer, epiphyte or macrofauna communities in the Lheebroekerzand. However the decrease in the total number of observed butterflies can be marked as an alarming situation. The condition of the trees is improving through time. The chemical concentrations of most pollutants indicate a downward trend in the different compartments of the ecosystem, with exception of ammonium, nitrate and sulphate in lakewater, the acidity of the lakewater and the concentration of lead in leaves and needles.

Correlations between biological and chemical variables are seen for the presence of butterflies with cadmium, lead and sulphur content in air and deposition and defoliation of the trees is correlated with sulphur, nitrogen and acidity in air and deposition. Substances responsible for acidification, eutrophication, desiccation and the presence of heavy metals in the ecosystem do not have a marked influence on the presence of birds, leafminers, epiphytes and macrofauna in the Lheebroekerzand.

If the reduction in emissions in Europe will be continued in future years, it is to be expected that the amount of most chemical compounds in the compartments lakewater, leaves and needles will diminish in future as well.

SAMENVATTING

In 1993 is het monitoring gebied in het Lheebroekerzand ingericht en opgenomen in het internationale netwerk van monitoring sites van het "International Co-operative Programme on Integrated Monitoring on Air Pollution Effects". De monitoring werd uitgevoerd volgens de "Manual for Integrated Monitoring" onder supervisie van het Laboratorium voor Ecotoxicologie van het Rijksinstituut voor Volksgezondheid en Milieu. Van 1993 tot en met 1999 werd een database gevuld met resultaten van het eigen onderzoek. Vervolgens werd deze database aangevuld met resultaten van onderzoek uitgevoerd in het Lheebroekerzand voor 1993. Deze gegevens werden verkregen van samenwerkende organisaties en instituten of werden gehaald uit de literatuur.

De beschikbaarheid van de vele chemische, fysische en biologische gegevens verzameld in het Lheebroekerzand, biedt de mogelijkheid om na te gaan of er trends in de tijd en eventuele relaties tussen de variabelen zijn waar te nemen.

Het eerste doel van dit onderzoek is om na te gaan of er met de beschikbare monitoringgegevens trends in de tijd statistisch zijn aan te tonen. Het tweede doel is om, van de biologische variabelen waarvoor statistisch een trend in de tijd is aangetoond, na te gaan of er correlaties te vinden zijn met chemische variabelen. De correlatieanalyse is beperkt tot chemische variabelen die gerelateerd zijn aan de milieubeleidsthema's verzuring, vermesting, verdroging en de aanwezigheid van zware metalen.

Het biologische gedeelte van het monitoringprogramma bestond uit een regelmatige inventarisatie van vogels, korstmossen, vlinders, bladmineerders en de vegetatie, evenals inventarisaties van de macrofauna aanwezig in het ven Kliplo en observaties aan naaldbomen ter bepaling van de vitaliteit. Het chemisch-fysische deel van het programma bestond uit meteorologische variabelen zoals temperatuur, luchtvochtigheid, hoeveelheid neerslag en instraling samen met chemische analyses van lucht, neerslag, bladeren, naalden, mossen, bodem, bodemwater, grondwater en venwater.

Resultaten en conclusies betreffende de trends in de tijd zijn:

- ✓ Van de 69 voorkomende vogelsoorten vertoonden 8 soorten een afname in de tijd en 5 soorten een toename in de tijd. Het aantal soorten, het totaal aantal waargenomen individuen en de Shannon Wiener Diversity Index veranderden niet in de tijd.
- ✓ Van de 24 waargenomen soorten vlinders vertoonden 4 soorten een afname in de tijd en geen van de soorten een toename. Het aantal soorten veranderde niet, maar het totaal aantal waargenomen individuen nam af in de tijd. De Shannon Wiener Diversity Index vertoonde een afnemende trend in de tijd.
- ✓ Van de bladmineerders vertoonde één van de 18 soorten een afname in de tijd; de Shannon Wiener Diversity Index en het aantal soorten namen eveneens af in de tijd, maar het totaal aantal individuen veranderde niet.
- ✓ Het aantal soorten korstmossen nam toe in de tijd.
- ✓ Voor het voorkomen van macrofauna in het ven Kliplo geldt dat van de 256 soorten er slechts

twee een afname in de tijd vertoonden en vijf een toename te zien gaven. Het aantal soorten, het totaal aantal organismen en de Shannon Wiener Diversity Index veranderden niet.

- ✓ In het monitoring programma “Forest Damage” is alleen een afnemende trend in de tijd waargenomen voor de variabele ontbladering.
- ✓ Er is geen reden tot ongerustheid betreffende de aanwezigheid van vogels, bladmineerders, korstmossen en macrofauna in het Lheebroekerzand.
- ✓ De afname van het totaal aantal geobserveerde vlinders geeft aanleiding tot verder onderzoek.
- ✓ De conditie van de bomen verbetert in de tijd.
- ✓ Een groot deel van de chemische stoffen vertonen een afname in de tijd in de verschillende compartimenten van het ecosysteem.
- ✓ Gedurende de gemeten periode vertonen concentraties van ammonium, nitraat en sulfaat in het venwater een toename in de tijd. Ook de zuurgraad vertoont een toenemende trend.
- ✓ In bladeren en naalden is de afnemende trend voor lood zoals waargenomen in de overige compartimenten niet geconstateerd.
- ✓ Als de afname in emissies in Europa gecontinueerd wordt in de komende jaren is het te verwachten dat ook de hoeveelheid anthropogene verbindingen in het venwater, de bladeren en de naalden gaan afnemen.

Resultaten en conclusies betreffende mogelijke correlaties tussen biologische en chemische variabelen zijn:

- ✓ Stoffen verantwoordelijk voor verzuring, vermesting, verdroging en de aanwezigheid van zware metalen in het ecosysteem hebben, in de aanwezige concentraties in het Lheebroekerzand, geen aantoonbare invloed op de aanwezigheid van vogels, bladmineerders, korstmossen en macrofauna.
- ✓ De aanwezigheid van de gemeten concentraties van cadmium, lood en zwavel in het Lheebroekerzand lijkt invloed te hebben op het voorkomen van vlinders.
- ✓ Voor de variabele ontbladering gemeten in het monitoring programma “Forest Damage” is een correlatie gevonden met zwavel en stikstof in lucht en in regenwater, en met de zuurgraad van het regenwater. Dit kan er op wijzen dat verzuring en vermesting invloed hebben op de conditie van de bomen in het Lheebroekerzand.

Aanbevelingen:

- ✓ Om meer overlappende data van biologische en chemische variabelen en langere tijdseries te verkrijgen moet de database uitgebreid worden met gegevens uit onderzoek voor 1993.
- ✓ Indirecte correlaties tussen alle gemeten variabelen kunnen onderzocht worden.
- ✓ De resultaten uit dit onderzoek kunnen nagerekend worden met data aanwezig in de

internationale database van het ICP-IM.

- ✓ Indien meer data beschikbaar zijn kan een multivariate analyse uitgevoerd worden.
- ✓ De afname van de vlinders dient nader onderzocht te worden.
- ✓ Om de verbetering van het ecosysteem te kunnen blijven volgen in de tijd is monitoring in de toekomst noodzakelijk.

De resultaten van dit onderzoek bevestigen dat de genomen emissie beperkende maatregelen in Nederland effectief zijn geweest. Het herstel van het ecosysteem heeft daarentegen nog niet volledig plaatsgevonden.

SUMMARY

In 1993 the Dutch monitoring station Lheebroekerzand became part of the network of the International Co-operative Programme on Integrated Monitoring on Air Pollution Effects. The contribution of the Dutch monitoring activities was set up according to the Manual for Integrated Monitoring and was executed under supervision of the Laboratory for Ecotoxicology of the National Institute for Public Health and the Environment. From 1993 to 1999 a database was build up with data collected in the Lheebroekerzand. Data from studies carried out in the Lheebroekerzand before 1993 was obtained from co-operating organisations and institutes or found in literature.

The availability of chemical, physical and biological data gathered at the Lheebroekerzand from 1993 to 1999, supplemented with data from studies from previous years, gives the opportunity to analyse and characterise the data for possible trends and correlations.

The first objective of this study is to detect and estimate trends in the extensive amount of monitoring data of the chemical, physical and biological variables through time. The second objective is to determine whether correlations between biological and chemical monitoring data are present. Only biological variables showing a trend through time, and chemical variables related to the environmental themes acidification, eutrophication, desiccation and the presence of heavy metals have been taken into account.

The biological monitoring activities consisted of a regular inventory of breeding birds, epiphytes, butterflies, leafminers and vegetation in the monitoring area, as well as inventories of aquatic macrofauna in the moorland pool Kliplo and observations on the performance of pine trees. The chemical-physical part included meteorological parameters like temperature, humidity, the amount of precipitation and irradiation, together with chemical analysis of organic and inorganic compounds in air, wet and dry deposition, mosses, leaves and needles, lake water, soil, soilwater and groundwater.

Results and conclusions concerning the trends through time are:

- ✓ For birds, 8 species showed a downward trend against 5 species an upward trend of the total of 69 observed species. The number of species, the total number of organisms and the Shannon Wiener Diversity Index did not change through time.
- ✓ From the total of 24 observed species of butterflies, 4 species showed a downward trend in time and none showed an upward trend. The number of species did not change but the total number of organisms showed a decreasing trend. The Shannon Wiener Diversity Index showed an increasing trend through time.
- ✓ For leafminers one of the 18 species showed a downward trend; the Shannon Wiener Diversity Index and the number of species were decreasing through time but the total number of organisms did not change.
- ✓ The number of species of epiphytes is increasing through time.
- ✓ From the total number of 256 species of macrofauna, 2 species showed a downward trend against

5 species an upward trend. The number of species, the total number of organisms and the Shannon Wiener Diversity Index did not change.

- ✓ For the monitoring programme Forest Damage the only variable with a downward trend was defoliation.
- ✓ There is no indication for degradation in the presence of birds, leafminers, epiphytes and macrofauna in the Lheebroekerzand.
- ✓ The decrease in the total number of organisms of butterflies can be marked as an alarming situation.
- ✓ The condition of the foliage is improving in time.
- ✓ A substantial part of the chemical pollutants indicate a downward trend in the different compartments of the ecosystem. However concentrations of ammonium, nitrate and sulphate in lakewater are still showing an upward trend. Also the acidity of the lakewater is not diminished during the monitored period.
- ✓ The observed downward trends in chemical variables are not detected for lead in leaves and needles.
- ✓ If the reduction in emissions in Europe will be continued in future years, it is to be expected that the amount of most chemical compounds in the compartments lakewater, leaves and needles will diminish in future as well.

Results and conclusions concerning the correlations between biological and chemical monitoring variables are:

- ✓ Substances responsible for acidification, eutrophication, desiccation and the presence of heavy metals in the ecosystem do not have a marked influence on the presence of birds, leafminers, epiphytes and macrofauna in the Lheebroekerzand.
- ✓ For the presence of butterflies it seems that there is a correlation with the cadmium, lead and sulphur content in air and deposition.
- ✓ The defoliation measured in the monitoring programme Forest Damage demonstrates a correlation with the sulphur, nitrogen and acidity in air and deposition. This could mean that acidification and eutrophication have influence on the foliage of the trees in the Lheebroekerzand.

Recommendations:

- ✓ More data of previous years has to be collected to obtain more overlapping data of biological and chemical variables and to ensure longer time-periods.
- ✓ Indirect correlations between variables should be studied.
- ✓ Trend and correlation analysis should be verified with the data of the international database of ICP-IM.

- ✓ With more data involved, it is possible to carry out multivariate analysis.
- ✓ The decrease of the presence of butterflies in the Lheebroekerzand needs to be studied.
- ✓ For tracing the recovery of the ecosystem, more years of monitoring is necessary.

The results of this study confirm that the emission reduction policy in the Netherlands has been effective. However, reduced exposure to pollutants is not yet fully reflected in ecosystem recovery.

1. INTRODUCTION

1.1. Background and framework

In 1979 several monitoring programmes started within the UN-ECE Convention on Long-range Transboundary Air Pollution (CLRTAP) with the purpose of monitoring and assessing effects of air pollutants on the environment. The International Co-operative Programme on Integrated Monitoring on Air Pollution Effects (ICP-IM) started in 1989 with a three year Pilot Programme with 36 monitoring sites participated grossly distributed over Europe and Canada. In 1992 ICP-IM has been established. From 1993 to 1999 the Netherlands' site the Lheebroekerzand was part of the network of ICP-IM. The most important objective of this integrated effect-directed programme is to determine and predict the state and change of terrestrial and freshwater ecosystems in a long-term perspective with respect to the impact of air pollutants.

In 1999 22 countries carried out the integrated monitoring programme with 70 sites in Europe and Canada.

Studies about the effects of exposure to air pollutants acting on particular receptors indicate the need of an integrated approach in order to understand the underlying mechanisms for the observed ecological effects. Special attention is needed to investigate the ecological impact of critical exposure. This can be accomplished by putting more emphasis on studies concerning the biological effects and the relationship with local chemical exposure levels.

The availability of chemical, physical and biological data gathered at the Lheebroekerzand from 1993 to 1999, supplemented with data from studies from previous years, gives an opportunity to detect and characterise possible causal relationships by means of trend- and correlation analysis.

1.2. The RIVM project: Integrated Monitoring Area Lheebroekerzand

Since 1989 the site at Lheebroekerzand, in the province of Drenthe, was gradually developed. Lheebroekerzand was chosen as a monitoring site because it most presumably represents the background concentration levels of many pollutants in the Netherlands. The relative "clean" ecological situation of the Lheebroekerzand and its biological function is expected to be quite vulnerable to the impact of significant changes in chemical compositions in different environmental compartments. Monitoring these changes is from a scientific view therefore very interesting and important. The area is located in a forest and nature reserve. Many studies have already been carried out in this area and many are still going on.

The co-ordination of the monitoring activities and the collection of data were carried out by the Laboratory for Ecotoxicology from the National Institute for Public Health and the Environment (RIVM-ECO). RIVM-ECO is the National Focal Point of the Netherlands for ICP-IM. In 1993, fieldwork started according to the Manual for Integrated Monitoring Programme Phase 1993-1996 (UN-ECE, 1993). Data of previous studies carried out in the Lheebroekerzand were collected and made available. For the subprogramme on Lakewater chemistry, the available data dated back to as early as

1924. In addition to the parameters in the subprogrammes in the Manual, additional biological parameters were added to the national programme. The biological monitoring activities consisted of regular inventories of breeding birds, epiphytes, butterflies, leafminers and vegetation in the monitoring area, as well as inventories of aquatic macrofauna in the moorland pool Kliplo and observations on the health and performance status of pine trees. The chemical-physical part included meteorological parameters such as temperature, humidity, the amount of precipitation and irradiation, with chemical analysis of organic and inorganic compounds in air, wet and dry deposition, mosses, leaves and needles, lakewater, soil, soilwater and groundwater.

Many national institutes, organisations and volunteers made it possible to carry out this ambitious monitoring programme.

Since the start of the monitoring programme in 1993 there has been pressure on continuity. Every year, the importance of integrated monitoring in the Netherlands had to be proven. Many internal RIVM-discussions and presentations were passed. In 1997 the Director Environmental Research of RIVM concluded that the project did not provide the information that was required for the national tasks of RIVM. Still the decision to stop the monitoring task was postponed to enable the identification of topics that may be of national interest. In June 1999 this was successful. Co-operation was found with several projects within RIVM and requests for data for the validation of modelling results were regularly posed.

Unfortunately, in July 1999, the Dutch Minister of the Environment decided to put an end to the Dutch contribution to the ICP on Integrated Monitoring. This decision was taken in view of budget restrictions for national environmental research. In January 2000 the research equipment was removed from the site at Lheebroekerzand.

The history, the objectives of the programme, the surveys of the monitoring data, as well as the organisational aspects of the monitoring site the Lheebroekerzand are described in several RIVM reports published by Mathijssen-Spiekman *et al.* (1994, 1995^a, 1995^b, 1996, 1998 and 2001).

1.3. Objectives of the present report

The dataset of the Lheebroekerzand, including chemical, physical and biological variables, is used to study and characterise the underlying correlations by means of trend– and correlation analysis.

The objectives of this report are:

a) to detect and estimate trends through time in the monitoring data of the chemical, physical and biological variables;

b) to determine whether direct effects of the chemical monitored parameters on the biology could be assessed by correlation analyses.

2. INTEGRATED MONITORING AREA LHEEBROEKERZAND

2.1. The site

2.1.1. General description

The Dutch monitoring station Lheebroekerzand is situated in the north-eastern part of the Netherlands, approximately 4 km from the village Dwingeloo in the province of Drenthe (Figure 1). This 95-hectare nature reserve is owned by the State Forestry Service (SBB) and is located within the national park "Dwingelderveld".

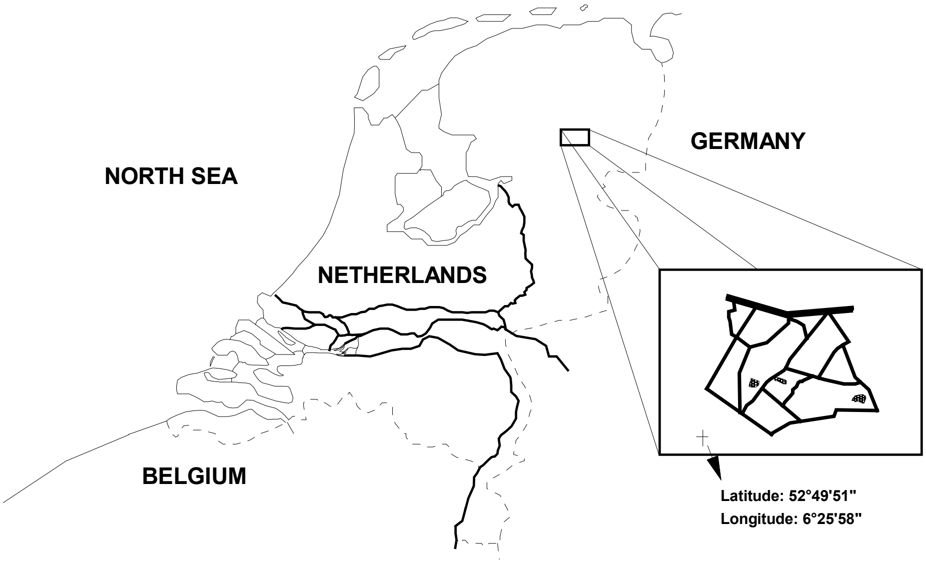


Figure 1: Location of the Lheebroekerzand in the Netherlands.

The national park, consisting of areas of forest, heath and drift sands, covers roughly 3500 hectares. The topography of the area is nearly flat and 12 m above the sea level. It has a locally temperate sea climate with a yearly average temperature of 8.6°C. The soil consists mainly of poor sands, part is humic sand or peat-moor. Table 1 presents basic information of the site.

The forest was established in 1918-1930 in a region that was originally a drift sand area. Today diverse vegetation exists in the area including trees, shrubs, heath and mosses. The site also provides suitable habitats for birds, foxes, squirrels, deer, reptiles and amphibians. Within the area a small oligotrophic lake, Kliplo, is located. This rainwater-fed lake has a surface of 0.6 hectares, aquatic biota as well as chemical features are monitored. Kliplo is selected because of its diversity of aquatic species and its well-preserved character.

Table 1: Basic information of the Lheebroekerzand.

Geographical co-ordinates	52°49'51" N 6°25'58" E
Size	95 ha (surroundings 3500 ha)
Owner	State Forestry Service
Protection status	Nature reserve
Maximum / minimum elevation	13.5 / 12.0 metres
Vegetation zone	Nemoral deciduous forest region
Long-term average precipitation (30-year period 1961-1990)	780 mm/year
Long-term average temperature (30-year period 1961-1990)	8.6° C
Climate	Temperate sea climate
Length of vegetation period (mean temperature > 5 ° C for 5 consecutive days)	200 days/year
Dominant vegetation	*Pinus sylvestris*
Dominant soil type	Sand
Anthropogenic stress	Highway, small villages, farmers

2.1.2. Geomorphology

The Lheebroekerzand area is part of a larger drift sand area, arisen in the Holocene "Formation of Kootwijk". The drift sands in the area are mainly covering eolic sediments "Formation of Eindhoven" or loamy ground moraine "Formation of Drenthe", with partially a thin layer of sand deposit "Formation of Twente" in between. A small part is oligotrophic peat-moor "Formation of Griendsveen" (Castel, 1984).

2.1.3. Hydrology

A thorough hydrological survey of the Dwingelderveld was made by T. Bakker, 1984. The hydrology of the monitoring area can be extracted from this survey. The Lheebroekerzand monitoring area contains the following pools: "Reigersplas", "Droseraveen" and "Kliplo". Only Kliplo is included in the ICP-IM-programme for water chemistry and hydrobiology. The pool probably originated as a result of compression of certain layers in the soil, thus becoming impermeable for water. About one metre under the pool a layer consisting of boulder clay is found. Due to the relative height of the impermeable layer, Kliplo appears to have a higher water table than the subsoil water table in the surrounding area. All the pools in Lheebroekerzand are hardly influenced by drainage and independent of the surrounding area. They are only fed by rainwater. Because of the glacial layer of boulder clay beneath the pools the exchange with groundwater is impossible. The sand deposit above the clay contains only water in winter when it is almost completely saturated with water. In the summer the sand deposit dries up. Only above impermeable layers the water remains. The Lheebroekerzand, as a part of the Dwingelderveld can be described as an infiltration area. The deep groundwater flux is from east to west with a maximum speed of 10 cm per year (Bakker 1984, Hoentjen 1993).

2.1.4. Vegetation

From 1908 onwards a nature reserve was established. Within this forest this specific area was developed between 1918 and 1930. Especially within the reserve area a wide variety of vegetation exists including *Pinus sylvestris* (Scots pine), *Quercus robur* (Oak), *Betula pendula* (Birch), *Juniperus communis* (Junipertree) as well as many species of heath, mosses and other plants.

2.2. The monitoring programme

From 1993 to 1999 the monitoring programme existed of the subprogrammes described in the Manual for Integrated Monitoring (UN-ECE, 1993) with additional biological monitoring programmes. The monitoring programmes executed in the Lheebroekerzand are:

Biological programmes
✓ Inventory of Birds (BB);
✓ Inventory of Butterflies (BF);
✓ Inventory of Leafminers (LM);
✓ Inventory of Plants (BV);
✓ Trunk Epiphytes (EP);
✓ Hydrobiology of Lakes (LB);
✓ Vegetation (VG);
✓ Forest Damage (FD);

Physical programme
✓ Climate (AM);

Chemical programme
✓ Air Chemistry (AC);
✓ Precipitation Chemistry (DC);
✓ Throughfall and Stemflow Chemistry (TF, SF);
✓ Metal Chemistry in Mosses (MC);
✓ Soil Chemistry (SC);
✓ Soil Water Chemistry (SW)
✓ Groundwater Chemistry (GW);
✓ Lake Water Chemistry (LC);
✓ Foliage and Litterfall Chemistry (FC, LF).

Figure 2 gives an overview of the specific locations where these subprogrammes were executed from 1993 to 1999.

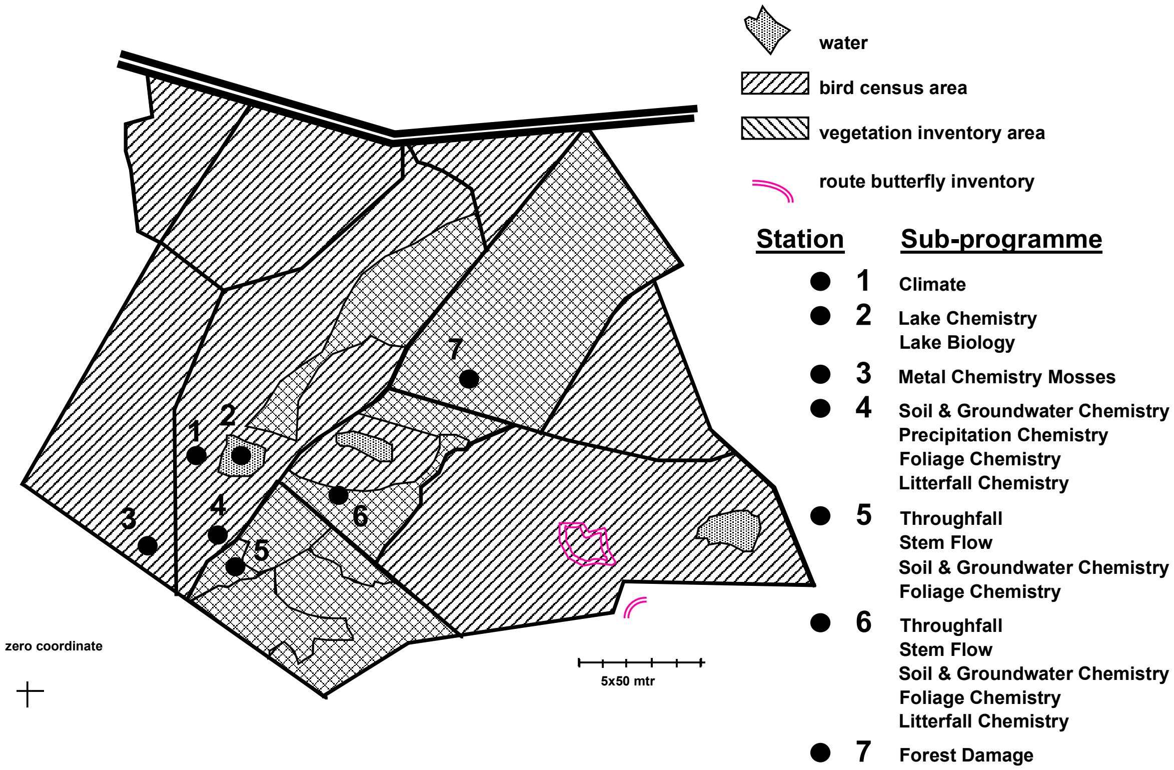


Figure 2: *Basemap Lheebroekerzand with station and activity indication.*

The programmes were carried out according the methods described in the Manual for Integrated Monitoring. Detailed descriptions are published by Mathijssen-Spiekman e*t al.* in the annual national reports (1994, 1995[a], 1995[b], 1996, 1998, 2001)

2.3. The monitoring data

From 1993 to 1999 data are collected in the Lheebroekerzand for the above mentioned monitoring programmes. Data from studies carried out in the Lheebroekerzand before 1993 were obtained from co-operating organisations and institutes or found in literature. A thorough literature research was not performed. Only data that were already collected during the years of monitoring are incorporated in this study. In Appendix I an overview is given of the complete dataset used for this study with the various sources of supply. Additionally, in Appendix II an overview is given of the available measured variables with their specific monitoring period.

In Figure 3 the availability of data of the different monitoring subprogrammes is shown. For trend analysis it is important to have long timeseries; for correlation analysis overlapping data are needed. Long timeseries are available for the monitoring programmes Lakewater chemistry, Deposition chemistry, Trunk epiphytes and Inventory of birds. Overlapping data are available; only between 1980 and 1999. Unfortunately not all data are appropriate to use for the analyses. Particularly data of the longer time series can't be used because data are not consistent over the years, mostly because of changing procedures (indicated in Figure 3).

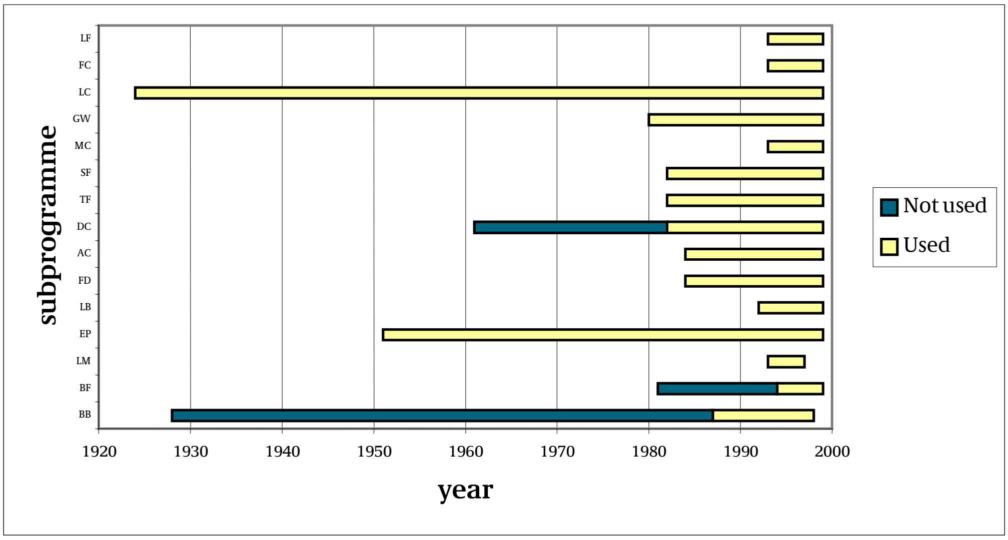


Figure 3: Data held in the Lheebroekerzand database.

All the results of field measurements and observations in the Lheebroekerzand were collected, checked and stored in the RIVM-ECO database that is constructed in DataPerfect software. The database includes relevant information for every measured value such as sampling conditions, method of analysing and value below detection limit. In Appendix III the additional information is explained.

3. TREND ANALYSIS

The first objective of this study is to detect and estimate trends in the monitoring data of the chemical-, physical- and biological variables over time. Statistical methods and graphical presentations for trend analysis give the opportunity to identify at a glance the observed variation over a relevant period of time. The detection and estimation of trends is complicated by characteristics of the monitoring data. For long-term studies, variations in the measurements can be caused by actual trends but also by changes in sampling- and analytical procedures and by seasonal or other cyclic fluctuations. Anticipating on variations possibly caused by changing sampling and analytical procedure data were checked. No attention is paid to variations caused by seasonality, because the time-series cover enough years of monitoring to make it possible to detect trends over the years.

An important requirement in trend analysis is the availability of complete datasets without missing values. The analysis is getting more complicated in situations that the period of missing values is becoming substantial. There is no uniform strategy how to handle such datasets. Helsel and Hirsch (1995) recommend using all data values when the period of missing values does not exceed a third of the total time domain.

3.1. Material and Method

3.1.1. Data preparation

The basis for the trend analysis is the database of the Netherlands of ICP-IM as described in paragraph 2.3. The physical variables are left aside because KNMI and RIVM-LLO did carry out many studies on this subject with a much more extensive dataset.

First, all datasets were checked for observations below detection limits. For these data, the values of the detection limits were divided by a factor of 2. Secondly, the units of similar variables used in the different monitoring programmes were unified.

Furthermore, the datasets were evaluated on the presence of missing values. For biological variables it was assumed that a missing value means that a species is not present. Subsequently, a zero was entered into the list of observations. For chemical variables the missing values were not filled in because the period of missing values did never exceed a third of the total time domain. All data values were used as recommend by Helsel and Hirsch (1995).

Finally, per variable and per monitoring programme the time period on which a trend analysis could be performed was evaluated. If this period was less than 4 years the data were not further used for the analysis. In the event of the programmes Inventory of Plants, Vegetation, Soil chemistry and Soilwater chemistry only data from one or two years of monitoring were available. Therefore, these data were not used for the detection of trends.

Subsequently, redundant information was removed from the data. The remaining data were coded using the subprogramme code, the variable code, the date and the value according to the next example:

BB = Subprogramme code

DRYO MAR = Variable code (species code)

8906 = Date (YYMM)

3.5 = Value

These procedures lead to a dataset that is suitable for the trend analysis.

3.1.2. Statistical method

For the trend analysis the linear regression (model I) has been used (Sokal and Rohlf, 1997). The date is chosen as the independant variable (X), the observed values are assigned the role of the dependant variables (Y). For each variable within each monitoring programme a linear regression has been calculated. If the slope of the regression line differs significantly from zero it means that a trend is determined. Table 2 shows the possible outcomes per variable.

Table 2: Possible trends in the data.

Trend	Confidence interval	Code
significant upward	99.9	+++
	99	++
	95	+
no significant trend	< 95	o
significant downward	95	-
	99	--
	99.9	---

3.2. Results and Discussion

All statistically significant trends are graphically presented in Appendix IV.

The available data made it possible to carry out 865-trend analyses; 168 significant trends were detected.

3.2.1. Biological variables

Statistically significant trends observed in the biological variables are presented in Tables 3 (downward in time) and 4 (upward in time).

Table 3: Biological variables showing a significant downward trend.
(-: $p<0.05$; --: $p<0.01$; ---: $p<0.001$)

Subprogramme	Code variable	Variable	Period	Trend
Inventory of Birds (BB), species	DRYOMAR	*Dryocopus martius*	87-98	-
	PARUCRI	*Parus cristatus*	87-98	-
	PHOEPHO	*Phoenicurus phoenicurus*	87-98	-
	STURVUL	*Sturnus vulgaris*	87-98	-
	AEGICAU	*Aegithalos caudatus*	87-98	--
	CARDCAN	*Carduelis cannabina*	87-98	--
	CUCUCAN	*Cuculus canorus*	87-98	--
	SYLVBOR	*Sylvia borin*	87-98	--
Family	STURNIDX	*Sturnidae* (1 species)	87-98	-
	AEGITHAX	*Aegithaidae (*1species*)*	87-98	--
	CUCULIDX	*Cuculidae (*1 species)	87-98	--
Order	CUCULIFS	*Cuculiformes (*1 species*)*	87-98	--
Inventory of Butterflies (BF), Species	PIERNAP	*Pieris napi*	94-99	-
	BOLOAQU	*Boloria aquilonaris*	94-99	--
	OCHLVEN	*Ochlodes venata*	94-99	--
	NRORG	Total number of organisms	94-99	--
	VACCOPT	*Vacciniina optilete*	94-99	--
Family	LYCAENIX	*Lycaenidae* (7 species*)*	94-99	-
	NYMPHALX	*Nymphalidae* (5 species)	94-99	-
	HESPERIX	*Hesperiidae* (4 species)	94-99	--
Biological Group	BF7	Species that hibernate as a small caterpillar (section group 4, 2 species)	94-99	--
	BF9	Species that hibernate as a juvenile caterpillar and have a short maturation (section group 4, 1 species)	94-99	--
	BF12	Species that hibernate as a juvenile caterpillar and have a long maturation (section group 4, 5 species)	94-99	--
Lnventory of Leafminers (LM)	SPP.Bp	Number of species on *Betula pendula*	93-97	-
	ST LUTEE.Bp	Stigmella luteella on *Betula pendula*	93-97	-
	DIX_SW.Bp	Shannon-Wiener diversity index of *Betula pendula*	93-97	--
Hydrobiology of Lakes (LB), species	FORELIL	*Forelia liliacea*	92-99	-
	LIMSUBC	*Limnephilus subcentralis*	92-99	-
Family	CORIXIDX	*Corixidae* (21 species)	92-99	-
Forest Damage (FD)	DEFO	Defoliation	84-99	---

Table 4: Biological variables showing a significant upward trend.
(+: p<0.05; ++: p<0.01; +++: p<0.001)

Subprogramme	Code variable	Variable	Period	Trend
Inventory of Birds	COLUOEA	*Columba oenas*	87-98	+
(BB), species	CORVCRO	*Corvus corone*	87-98	+
	MOTAALB	*Motacilla alba*	87-98	+
	SAXITOR	*Saxicola torquata*	87-98	++
	LULLARB	*Lullula arborea*	87-98	+++
Family	COLUMBIX	*Columbidae* (3 species)	87-98	++
	ALAUDIDX	*Alaudidae* (1 species)	87-98	+++
Order	COLUMBIS	*Columbiformes* (3 species)	87-98	++
Inventory of Butterflies (BF)	DIX_SW	Shannon-Wiener diversity index	94-99	+
Trunk Epiphytes (EP)	SPPV	Number of species	51-99	+++
Hydrobiology of	ARREAFF	*Arrenurus affinis*	92-99	+
Lakes (LB), species	ARREGLO	*Arrenurus globator*	92-99	+
	PELCAES	*Peltodytes caesus*	92-99	+
	PLEAMIN	*Plea minutissima*	92-99	+
	ARRENEU	*Arrenurus neumani*	92-99	++
Family	AESHNIDX	*Aeshnidae* (6 species)	92-99	+
	PLEIDAEX	*Pleidae* (1 species)	92-99	+
	ARRENURX	*Arrenuridae* (11 species)	92-99	++

3.2.1.1 Inventory of birds (BB)

From 1928 to 1998 131 species are observed in the Lheebroekerzand. For all these species a trend analysis is carried out. Trend analysis is also carried out on the Shannon-Wiener index, on the total number of verified species, on the total number of birds and on the family and order level of the organisms. It became obvious that trend analysis on the data of the inventories carried out before 1987 were not reliable, because the inventories were not carried out at regular time-intervals and often only the presence of the species was recorded and not the number of organisms. The presented trend analyses are the results for the period from 1987 to 1998. During this period 69 species are observed.

In Table 3 a downward trend in time is identified for 8 species *(Aegithalos caudatus, Carduelis cannabina, Cuculus canorus, Dryocopus martius, Parus cristatus, Phoenicurus phoenicuris, Sturnus vulgaris* and *Sylvia borin*), 3 families *(Aegithaidae, Cuculidae* and *Sturnidae,*) and 1 order (*Cuculiformes*). An upward trend in time is identified for 5 species (*Columba oenas, Corvus corone, Lullula arborea, Motacilla alba* and *Saxicola torquata*), 2 families (*Alaudidae* and *Columbidae*) and 1 order (*Columbiformes*) (Table 4). Trends in the Shannon-Wiener index, in the total number of verified species and in the total number of birds were not found.

Van Dijk *et al.* (1999) determined indexes for birds for natural habitat plots monitored in the Netherlands. Four of the five species with an upward trend *(Corvus corone, Columba oenas, Lullula arborea, Saxicola torquata)* were confirmed by these indexes. The index for the fifth species (*Motacilla alba)* is fluctuating during the years 1984-1997 and does not show a clear upward trend. For only two, within a total of 8 species, the downward trend is confirmed by the indexes of Van Dijk (*Carduelis cannabina* and *Sturnus vulgaris*). The other downward trends in the Lheebroekerzand could be caused by local conditions (such as tree felling) and are not generally observed in the Netherlands. Influences responsible for changes in the bird population are severe winters, drought, the number of mice and the availability of beechnuts (Van Dijk *et al.,* 1999).

3.2.1.2 Inventory of butterflies (BF)

In the period 1981 to 1990 the inventories only consisted of observations on the number of species . The number of organisms was not counted. A trend in the number of observed species could not be identified for this period. From 1994 to 1999, next to the number of species, the number of organisms was counted. For this period trend analysis is carried out on the species, the families and the orders. Trend analysis is also performed on the total number of species, on the total number of organisms, on the Shannon-Wiener index and on different biological groups. The division in biological groups is done according to F.A. Bink as described in Wijnhoff *et al.,* 1992. On the basis of the way of hibernation of the butterflies the six groups were distinguished as: 1) species that hibernate as grown-up butterflies; 2) Species that hibernate as pupae; 3) species that hibernate in the egg-stage; 4) species that hibernate as a caterpillar (this group is parted in 8 sections, each section corresponds with a way of hibernation of the caterpillar, see Table 3); 5) species that hibernate twice within one generation and 6) species that hibernate the first year as pupae and the second year as caterpillar.

A total of 24 species were recognised from 1994 to 1999.

Table 3 shows a downward trend for 4 species *(Boloria aquilonaris, Ochlodes venata, Pieris napi* and *Vacciniina optilete*), 3 families (*Lycaenidae, Hesperiidae* and *Nymphalidae*), the total number of organisms and three sections of the fourth biological group. Only the butterflies that hibernate as a caterpillar show a downward trend. On the other hand, the only variable that show an upward trend is the Shannon-Wiener diversity index (Table 4). Trends in the number of individual species are not seen.

Indexes given by Van Swaay and Ketelaar, (2000) for *Ochlodes venata* and *Pieris napi* for monitored plots in the Netherlands show the same downward trend. The other two species, *Boloria aquilonaris* and *Vacciniina optilete* are common in the Lheebroekerzand, but became scarce in other parts of the Netherlands (Veling, 1995). The downward trends for *Boloria aquilonaris* and *Vacciniina optilete* can not be confirmed by indexes of Van Swaay and Ketelaar.

3.2.1.3 Inventory of leafminers (LM)

Monitoring is carried out from 1993 to 1997. For this whole period trend analysis is possible for the species, the families and the orders. Besides, the available data of the total number of species, the total number of organisms and the Shannon-Wiener diversity index is analysed. A distinction is made for leafminers on different trees.

From the total of 18 observed leafminers species, only one species (*Stigmella luteella*) showed a downward trend in time on *Betula pendula* but not on *Quercus robur*. The number of species and the Shannon-Wiener index (only on B*etula pendula*) also had a significantly downward trend. The total number of organisms did not change significantly during the period of monitoring.

3.2.1.4 Trunk epiphytes (EP)

Since 1951, inventories on epiphytes were carried out in the Lheebroekerzand. The available data could not be made quantitative. Therefore, only a trend analysis could be carried out on the total number of observed species. This turned out to have a significant upward trend. During the period 1951 to 1999 75 species are determined in the Lheebroekerzand. Per year the number of verified species is varying from 1 to about 40 species.

Van Herk (1998) also mentions the upward trend: the variety of species has changed; acidophytic species disappeared and other species, particularly nitrophytic species appeared.

3.2.1.5 Lakewater biology (LB)

Trend analyses are done for the period 1992 to 1999 for the species, family, order, Shannon-Wiener diversity index, the number of species, total number of organisms and the amount of chlorophyll. From the 256 species only two species showed a downward trend (*Forelia liliacea* and *Limnephilus subcentralis*) against five with an upward trend (*Arrenurus affinis, Arrenurus globator, Arrenurus neumani, Peltodytes caesus* and *Plea minutissima*). Just one family (*Corixidae*) showed a downward trend and three families (*Aeshnidae, Arrenuridae* and *Pleidae*) an upward trend. Trends on the number of species, the total number of organisms, the Shannon-Wiener diversity index and the amount of chlorophyll were not statistically significant.

3.2.1.6 Forest damage (FD)

The data of the whole monitored period (1984-1999) was appropriate to carry out trend analysis on all variables with the exception of the crown height and the crown width, which were only observed in 1998. Only for defoliation a downward trend is detected (Table 3), but this means that the percentage of defoliation is decreasing, thus the condition of the crown of the tree is improving. The upward trends found for the height and the diameter of the trees only mean that the trees are growing as expected. These trends are not shown in Table 4.

3.2.2. Chemical variables

Statistically significant trends observed in the chemical variables are presented in Tables 5 (downward in time) and 6 (upward in time).

Table 5: Chemical variables showing a significant downward trend.
(-: p<0.05; --: p<0.01; ---: p<0.001)

Subprogramme	Code variable	Variable	Period	Trend
Air Chemistry (AC)	O_3	Ozone	84-99	-
	NH_4	Nitrogen ammonium	90-99	---
	NO_2	Nitrogen nitrogen dioxide in gas phase	84-99	---
	NO_3	Nitrogen nitrate in gas phase	90-99	---
	SO_2	Sulphur sulphur dioxide	84-99	---
	SO_4	Sulphur sulphate	84-99	---
Precipitation Chemistry (DC)	Cu	Copper filtered	93-99	-
	Al	Aluminium filtered	96-99	---
	Ca	Calcium filtered	82-99	---
	Cd	Cadmium filtered	93-99	---
	Cr	Chromium filtered	93-99	---
	Mg	Magnesium filtered	82-99	---
	Ni	Nickel filtered	93-99	---
	Pb	Lead filtered	93-99	---
	SO_4	Sulphate filtered	82-99	---
Throughfall Chemistry (TF) *Pinus sylvestris* (PS)	PO_4	Phosphorous phosphate filtered	82-99	-
	Zn	Zinc filtered	93-99	-
	Cr	Chromium filtered	93-99	--
	Na	Sodium filtered	82-99	--
	Ni	Nickel filtered	93-99	--
	Pb	Lead filtered	93-99	--
	Al	Aluminium filtered	96-99	---
	Ca	Calcium filtered	82-99	---
	Cd	Cadmium filtered	93-99	---
	CTY.25	Specific conductivity at 25 °C	82-99	---
	Mg	Magnesium filtered	82-99	---
	NH_4N	Nitrogen ammonium filtered	82-99	---
	NO_3	Nitrogen nitrate filtered	82-99	---
	SO_4	Sulphur sulphate filtered	82-99	---
TF, *Quercus robur* (QR)	Ca	Calcium filtered	93-99	-
	Mg	Magnesium filtered	93-99	-
	Na	Sodium filtered	93-99	--
	Pb	Lead filtered	93-99	--
	Al	Aluminium filtered	96-99	---
	Cd	Cadmium filtered	93-99	---
	Cr	Chromium filtered	93-99	---
	Ni	Nickel filtered	93-99	---
Stemflow Chemistry (SF) *Pinus sylvestris* (PS)	F	Fluoride filtered	82-85	-
	Pb	Lead filtered	93-99	-
	Al	Aluminium filtered	96-99	---
	As	Arsenic filtered	93-99	---
	Ca	Calcium filtered	82-99	---
	Cd	Cadmium filtered	93-99	---
	Cl	Chloride filtered	82-99	---
	Cr	Chromium filtered	93-99	---
	CTY.25	Specific conductivity at 25 °C	82-99	---
	Mg	Magnesium filtered	82-99	---
	Na	Sodium filtered	82-99	---
	NH_4N	Nitrogen ammonium filtered	82-99	---
	NO_3	Nitrogen nitrate filtered	82-99	---
	SO_4	Sulphur sulphate filtered	82-99	---

Table 5: Chemical variables showing a significant downward trend (continued).
(-: $p<0.05$; --: $p<0.01$; ---: $p<0.001$)

Subprogramme	Code variable	Variable	Period	Trend
Stemflow Chemistry	Mg	Magnesium filtered	93-99	-
(SF)	SO_4	Sulphur sulphate filtered	93-99	-
Quercus robur (QR)	Al	Aluminium filtered	96-99	--
	Na	Sodium filtered	93-99	--
	As	Arsenic filtered	93-99	---
	Cd	Cadmium filtered	93-99	---
	Cr	Chromium filtered	93-99	---
Metal Chemistry of	Cu	Copper	93-99	-
Mosses (MC)	Ni	Nickel	93-99	-
	Pb	Lead	93-99	-
Groundwater	Ca	Calcium dissolved/filtered	80-99	-
Chemistry (GW)	Fe	Iron dissolved/filtered	89-99	-
Level 1200 cm	Mg	Magnesium dissolved/filtered	80-99	-
	NO_3N	Nitrogen nitrate dissolved/filtered	80-99	-
	As	Arsenic dissolved/filtered	80-99	--
	Ni	Nickel dissolved/filtered	80-99	--
	Zn	Zinc dissolved/filtered	80-99	---
GW, level 3000cm	Zn	Zinc dissolved/filtered	80-98	--
Lake Water	Fe	Iron filtered	76-96	-
Chemistry (LC)	Ca	Calcium filtered	68-99	--
	CNR.N320	Colour number nonfiltered 320 nm	81-91	--
	pH	pH of liquids measured in field	24-99	--
	P_{tot}	Phosphorous total filtered	72-96	--
	Cl	Chloride filtered	58-99	---
	Wlevel	Watermark	82-98	---
Foliage Chemistry	Fe	Iron	96-99	-
(FC)	DW	Oven-dry sample weight of 100 needles/leaves	93-99	-
Pinus sylvestris (PS)				
FC, *Quercus robur* (QR)	P_{tot}	Phosphorous total	94-99	--
Litterfall Chemistry	Cu	Copper	93-99	-
(LF)	Ldep	Litterfall amount	93-99	-
Pinus sylvestris (PS)	P_{tot}	Phosphorous total	94-99	-
LF,	K	Potassium	93-99	-
Quercus robur (QR)	Ldep	Litterfall amount	93-99	-
	P_{tot}	Phosphorous total		--

Table 6: *Chemical variables showing a significant upward trend.*
(+: $p<0.05$;++: $p<0.01$;+++-: $p<0.001$)

Subprogramme	**Code Variable**	**Variable**	**Period**	**Trend**
Precipitation Chemistry (DC)	pH	pH of liquids	82-99	++
Throughfall Chemistry (TF) *Quercus robur* (QR)	PO_4	Phosphorous phosphate filtered	93-99	+
	pH	pH of liquids	93-99	+++
Stemflow Chemistry (SF) *Pinus sylvestris* (PS)	pH	pH of liquids	82-99	+++
	RR	Precipitation partial	82-99	+++
SF *Quercus robur* (QR)	K	Potassium filtered	93-99	+
	RR	Precipitation partial	93-99	+++
Metal Chemistry of Mosses (MC)	As	Arsenic	94-99	+
Groundwater Chemistry (GW) level 1200 cm	COR	Organic carbon dissolved/filtered	80-94	++
	Na	Sodium dissolved/filtered	80-99	++
GW, level 3000 cm	Ca	Calcium dissolved/filtered	80-98	+
	Mn	Manganese dissolved/filtered	90-98	+
	SO_4S	Sulphur sulphate dissolved/filtered	80-98	+
	Sr	Strontium dissolved filtered	90-98	+
	Fe	Iron dissolved/filtered	89-98	++
	K	Potassium dissolved/filtered	80-98	++
	Cl	Chloride dissolved/filtered	80-98	+++
	Mg	Magnesium dissolved/filtered	80-98	+++
Lake Water Chemistry (LC)	Al	Aluminium filtered	78-99	+
	K	Potassium filtered	68-99	+
	SO_4S	Sulphur sulphate filtered	72-99	+
	NO_3N	Nitrogen nitrate filtered	68-99	++
	CNR.N455	Colour number nonfiltered 455nm	90-99	+++
	NH_4N	Nitrogen ammonium filtered	68-99	+++
Foliage Chemistry (FC) *Pinus sylvestris* (PS)	Stot	Sulphur total	94-99	+
	Pb	Lead	93-99	+
FC, *Quercus robur* (QR)	CORT	Total organic carbon	93-99	+
	Pb	Lead	93-99	+
Litterfall Chemistry (LF) *Pinus sylvestris* (PS)	Pb	Lead	93-99	+
	Sr	Strontium	96-99	+
	S_{tot}	Sulphur total	94-99	+
	Mg	Magnesium	93-99	++
LF, *Quercus robur* (QR)	Ca	Calcium	93-99	+
	COR	Total organic carbon	93-99	+
	Pb	Lead	93-99	+

3.2.2.1 Air chemistry (AC)

The complete dataset held in the database was appropriate to carry out the trend analysis. All chemical substances in the air compartment, with the exception of ammonia, demonstrate a significant downward trend in concentration (Table 5). For the concentration of ammonia in air no trend is seen.

The decrease of most of the monitored chemical substances in the air compartment at Lheebroekerzand during the last fifteen years indicates that the policy aimed to diminish the emission of air pollutants in the Netherlands seems to be successful.

3.2.2.2 Precipitation chemistry (DC)

Analyses are carried out in rainwater collected in open collectors and with wet-only collectors. The choice is made to use the data on bulk precipitation because these are collected locally and the period of monitoring for the open collectors matches with available data of other monitoring programmes. A number of chemical substances is monitored since 1982. All available data of the bulk monitoring is used for the trend analysis.

Table 5 shows a downward trend for many organic and inorganic chemical compounds. The pH is the only variable with an upward trend. The downward trend for SO_4 is also observed in the air chemistry-monitoring programme.

Statistically significant downward trends of NH_4, NO_3 and SO_4 in bulk deposition are observed in 50% of the ICP-IM sites for the period 1988-1998 (Forsius e*t al.*, 2001). In the Netherlands monitoring area, Forsius only demonstrated a downward trend for NO_3. He didn’t detect trends for SO_4 and NH_4. The longer time series used in this study (1982-1999) actually does show a significant downward trend for $SO_{4.}$

3.2.2.3 Stemflow chemistry (SF) and Throughfall chemistry (TF)

The chemical substances in the stemflow and throughfall compartment are monitored for two different tree species, *Pinus sylvestris* and *Quercus robur*. A view of the trends is given per tree species in Table 5 and 6. The start of the period of monitoring varies from 1982 to 1996 per variable. From 1993 to 1999 data from all variables is available with exception of aluminium, which is measured from 1996 to 1999. Most of the chemical concentrations show a downward trend both for *P. sylvestris* and *Q. robur*. Only PO_4 shows an exception with an upward trend in throughfall rainwater of *Q. robur*. The amount of precipitation for stemflow for both tree species shows a clear upward trend, as does pH in stemflow of *P. sylvestris, pH* in throughfall of *Q. robur* and potassium in stemflow of *Q. robur*.

3.2.2.4 Metal chemistry of mosses (MC)

The monitoring period for mosses was relatively short (1993-1999). Nevertheless, 3 downward trends are detected (Cu, Ni and Pb) and 1 upward trend (As) in the total of 23 measured variables (Table 5 and 6).

3.2.2.5 Groundwater chemistry (GW)

Analyses in groundwater are carried out since 1980 in samples taken at two levels below ground level, namely at 1200 and 3000 cm. The number of variables analysed is enlarged in 1990. The period per variable that is used for the trend analysis is given in Table 5 and 6. In total 24 variables are monitored per level. On the level of 1200 cm, 7 downward (As, Ca, Fe, Mg, Ni, NO_3, Zn) and 2 upward (COR, Na) trends were detected. At 3000 cm just 1 downward

(Zn) and 8 upward trends (Ca, Cl, Fe, K, Mg, Mn, SO_4 and Sr) were seen. It is possible that the downward trends in concentrations of the chemicals did not yet reach the depth of 3000 cm.

3.2.2.6 Lakewater chemistry (LC)

Since 1924, samples are taken from lake Kliplo. This does not mean that all variables that are part of the current subprogramme LC are monitored since then. Nevertheless a number of data are available for a very long period. Per variable the longest possible time-period is chosen to be subject to trend analysis. From the total of 29 variables, 7 (Ca, Cl, CNR.N320, Fe, Ptot, WM, pH) showed a downward trend and 6 (Al, CNR.N455, K, NH_4, NO_3, SO_4) an upward trend (Table 5 and 6). Van Dam (1996) also identified a significant increase of SO_4, NH_4 and K in lake Kliplo from 1981 to 1994.

An increase of SO_4 and a decrease of Ca concentration levels in the lake indicate acidification processes in this compartment, which is confirmed by the decrease of the pH. Furthermore an eutrophication of the lake is likely, since ammonium and nitrate show increasing concentrations.

3.2.2.7 Foliage chemistry (FC) and Litterfall chemistry (LF)

The chemical substances of the monitored variables are analysed in leaves of the oak *Q. robur* and in needles of the pine tree *P. sylvestris*. For foliage and litterfall chemistry similar upward trends are seen for concentrations of Pb, S_{tot} and COR (Table 5 and 6). Downward trends are less equal. For Foliage chemistry only 3 out of 52 variables show a downward trend against 4 demonstrating an upward trend. For Litterfall chemistry 6 variables show a downward trend and 7 an upward trend.

3.3. Discussion on trends in different compartments

To compare the trends in the different compartments a graphical summary is given in figures 4, 5, 6, 7, 8 and 9. The monitoring programmes are put on the x-axis (for abbreviations see Appendix VI) and the significance of the trends on the y-axis (see Table 2). No indicator in the figures means that these variable is not included in this specific programme.

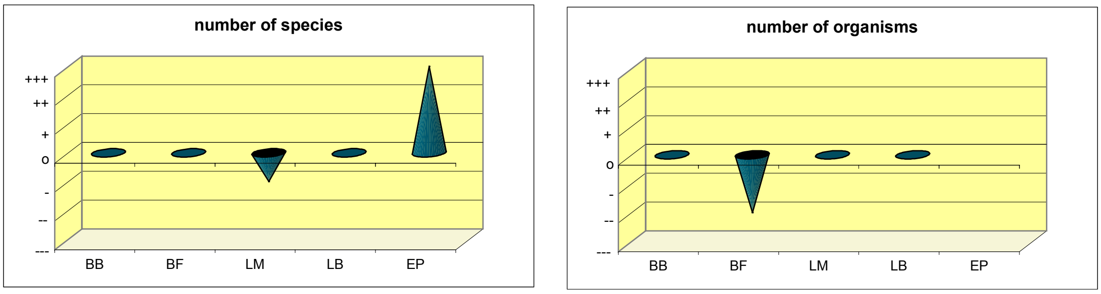


Figure 4: Trends in biological inventories

The number of species in the different biological groups show no clear similarities. The number of species of birds, butterflies and waterorganisms are not changing significantly in time. More epiphyte species are observed, but less leafminers.

For the number of organisms only the decrease of butterflies is significant.

In order to gain a better view of the effects of these changes more data of monitoring in the future is necessary.

For the chemical variables the sequence of the compartments on the x-axis corresponds to the exposure route for the chemical substances in the area. This route can be roughly divided in the input- (AC, DC), catchment- (TF, SF, MC, FC, LF, LC) and output route (GW). For the comparison only compounds are chosen that are of importance for the environmental problems acidification, eutrophication and presence of heavy metals.

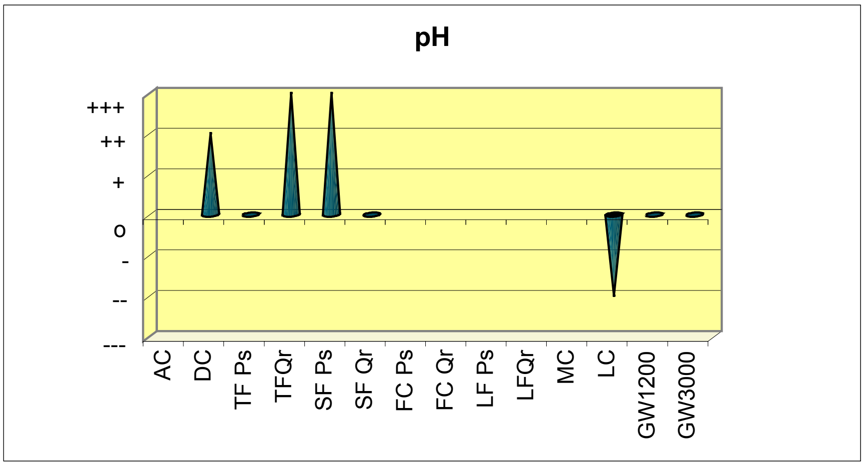
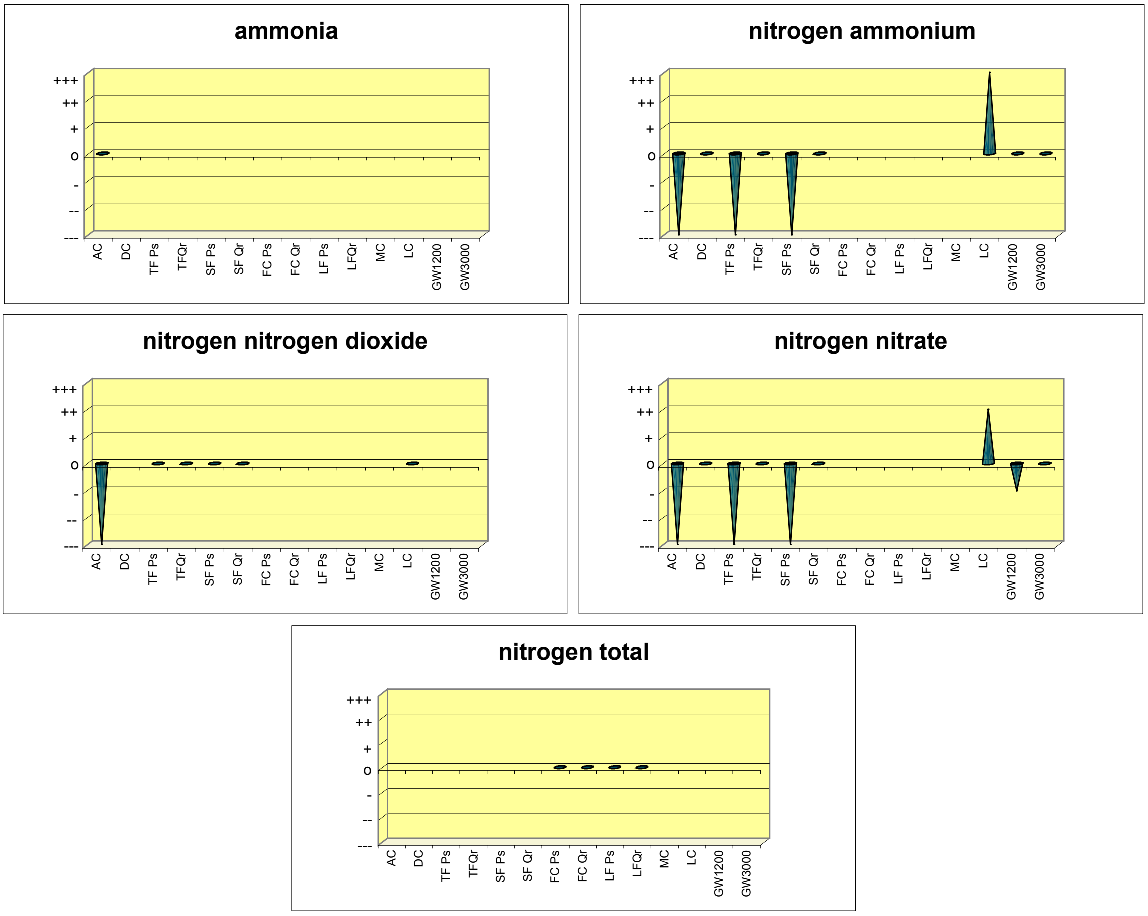


Figure 5: Trend of pH in the different compartments

For pH an upward trend is observed in rainwater (Figure 5). However the trend in pH in lakewater of the lake Kliplo is significantly downward.


ammonia

nitrogen ammonium

nitrogen nitrogen dioxide

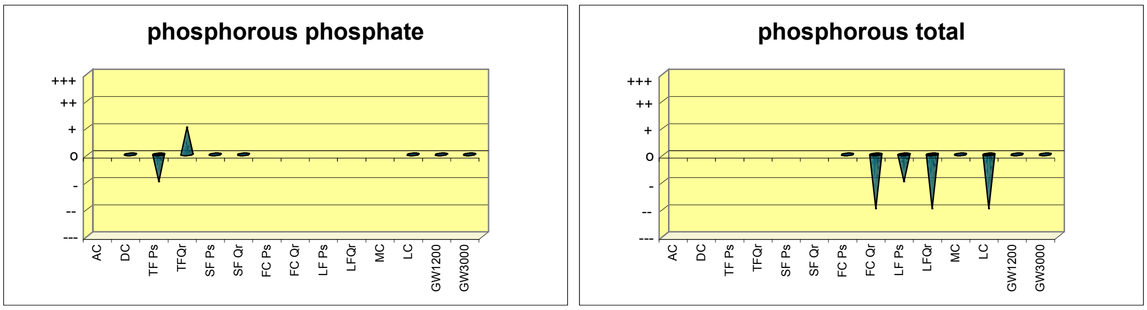
nitrogen nitrate

nitrogen total

Figure 6: Trends of nitrogen in the different compartments.

Compounds with nitrogen are compared in Figure 6. For the analysed amount of ammonium the trends for most compartments vary between strongly negative and zero. In contrast to the decreasing trend in the input of NO_3 and NH_4 in air (AC) a statistically significantly increasing trend of these substances are observed in lakewater.

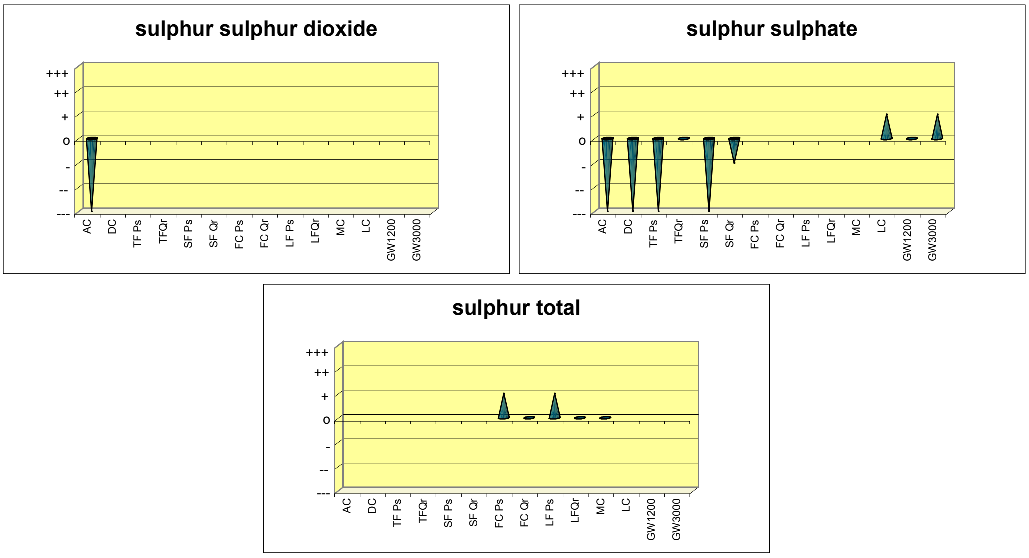
For phosphorous compounds only a weak upward trend is observed in throughfall rainwater of the oak, whereas an equally weak downward trend is observed in throughfall in pine stands (Figure 7). In the other compartments no trends are detected.


phosphorous phosphate

phosphorous total

Figure 7: Trends of phosphorous in the different compartments.

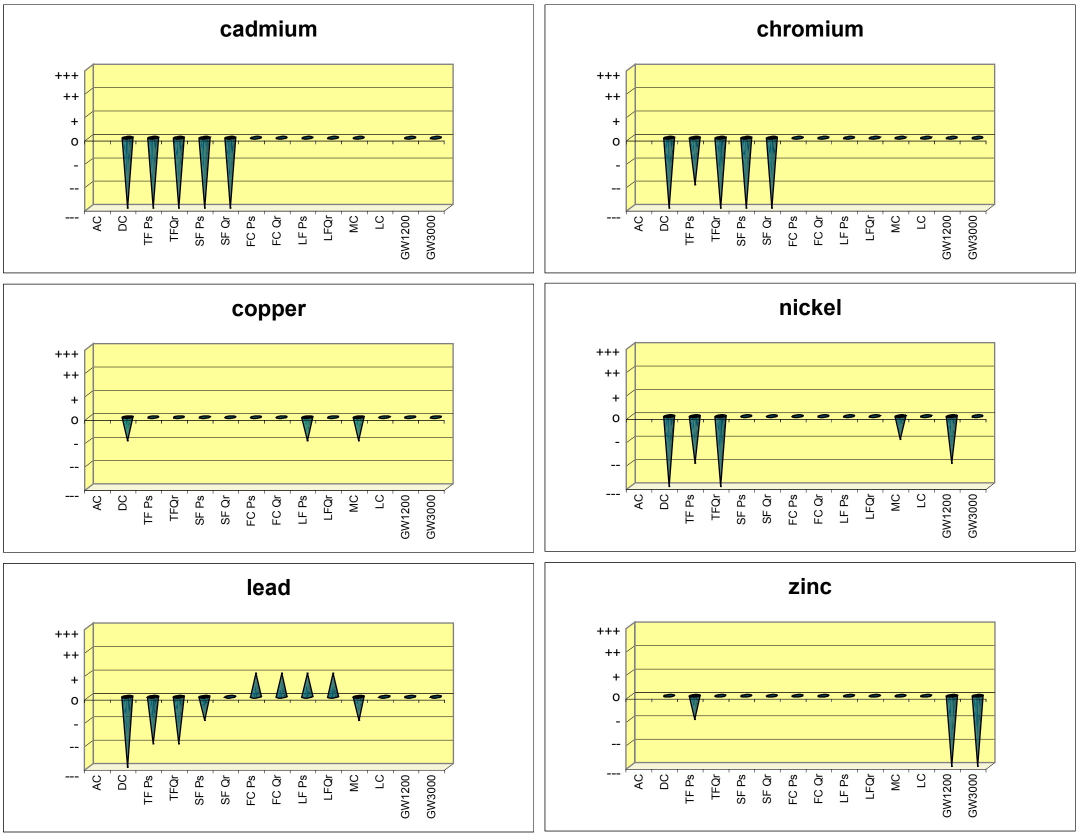
The decreasing trend for SO_4 in the input (AC, DC) is reflected in TF and SF chemistry, but not in lakewater and groundwater. For the total sulphur concentration in needles an increase is observed (Figure 8), but not for the amount of sulphur in leaves and mosses. The decreases in European emissions of sulphur can be clearly deducted from the results obtained from the Lheebroekerzand ecosystem.


sulphur sulphur dioxide

sulphur sulphate

sulphur total

Figure 8: Trends of sulphur in the different compartments.


cadmium

chromium

copper

nickel

lead

zinc

Figure 9: Trends of heavy metals in different compartments .

Heavy metals show a significant decrease in a number of compartments (Figure 9). Only for lead upward trends are observed, i.e. in the leaves and needles of both *Quercus robur* and

Pinus sylvestris.

In a relative clean area as the Lheebroekerzand a tentative conclusion on the basis of the available data can be that the concentrations in the different compartments of the ecosystem are decreasing. Changes in lakewater and groundwater appear to be less closely related to changes in deposition.

4. CORRELATION ANALYSIS

The second objective of this study is to relate possible causes and effects by determination of correlations between biological and chemical monitoring variables. Only biological variables with significant trends in time were used for this correlation study.

4.1. Material and Method

4.1.1. Data preparation

The dataset as prepared for the trend analysis, consisting of the subprogramme code, the variable code, the date and the value, is used for the correlation analysis.

Correlation analysis is applied to those variables that are important for the objectives of the integrated monitoring programme of ICP-IM causative themes.

The following data are used for the analysis:

1. biological variables with a significant trend in time.
2. yearly averages of the chemical variables with the restriction that for monthly monitored variables a minimum of 9 values is needed and for quarterly measurements 3.
3. data with a minimum of 5 years of overlapping values for the biological and chemical variables.
4. chemical variables related to environmental problems as acidification, eutrophication, desiccation and the presence of heavy metals (Table 7).
5. chemical subprogrammes that are supposed to have a direct influence on the biological subprogramme (Table 8).

Table 7 Chemical substances related to the environmental problems.

Environmental problem	Chemical
Acidification	SO_2, SO_4, NH_3, NH_4, NO_2, pH, S_{tot}, N_{tot}
Eutrophication	NO_2, NO_3, NH_3, NH_4, PO_4, N_{tot}, P_{tot}
Desiccation	RR, WM, WL
Heavy metals	Cd, Cr, Cu, Ni, Pb, Zn

Table 8: Combination of subprogrammes chosen for the correlation analysis.

Subprogramme	Air Chemistry (AC)	Precipitation Chemistry (DC)	Throughfall/ Stemflow Chemistry (TF/SF)	Lake Water Chemistry (LC)	Foliage/ Litterfall Chemistry (FC/LF)	Ground Water Chemistry (GW)
Inventory of birds (BB)	X	X	X		X	
Inventory of butterflies (BF)	X	X	X		X	
Trunk Epiphytes (EP)	X	X	X		X	
Inventory of leafminers (LM)	X	X	X		X	
Lakewater Biology (LB)				X		
Forest Damage (FD)	X	X	X		X	X

4.1.2. Statistical method

A linear correlation between variables was calculated using the product-moment correlation coefficient (Sokal and Rohlf, 1997). In case of significant correlations ($p \leq 0.05$), linear regression lines with 95% confidence intervals were plotted in the scatterplots. Three uncertainty levels were taken into account i.e. 5 %, 1% and 0.1%.

If there was not any significant correlation between a biological and a chemical variable the lines were left out in the scatterplots. The plots with significant correlations are marked as follows: white $p<0.05$; yellow $p<0.01$; green $p<0.001$.

4.2. Results and Discussion

In Appendix V, scatterplots with regression lines are presented of the selected variables.

The results of the scatterplots in Appendix V are not univocal. Positive and negative correlations are found for the same chemical variables. To get an overview of the results, the number of significant correlations is expressed as a percentage of the biological variables entered in the analysis. In Table 9, an overview is presented of the negative correlations and in Table 10 of the positive correlations.

Table 9: *Percentage of variables in each biological programme having a significant negative correlation with a chemical variable. (Combinations with only one variable are indicated with an asterisk; - : not calculated)*

Monitoring Programme	Variable	Inventory of Birds (BB)	Inventory of Butterflies (BF)	Trunk Epiphytes (EP)	Leafminers (LM)	Lake Water Biology (LB)	Forest Damage (FD)
Air	NH_3	5	8	0	0	-	0
Chemistry	NH_4	10	0	0	0	-	0
(AC)	SO_2	30	8	0	0	-	0
Deposition	Cr	5	8	0	0	-	0
Chemistry	Ni	0	8	0	0	-	0
(DC)	pH	0	25	0	0	-	0
	RR	10	0	0	0	-	0
Throughfall	Cd	0	8	0	0	-	0
Chemistry	Cr	2.5	4	0	0	-	0
(TF)	Ni	2.5	8	0	0	-	0
(*Pinus* and	pH	0	21	0	0	-	0
Quercus)	PO_4	7.5	13	0	0	-	0
	RR	5	0	0	0	-	0
Stemflow	Cd	0	4	0	0	-	0
Chemistry	Cr	2.5	8	0	0	-	0
(SF)	pH	2.5	21	0	0	-	100*
(*Pinus* and	PO_4	0	4	0	0	-	0
Quercus)	RR	2.5	8	0	0	-	0
Lakewater	NH_4	-	-	-	-	17	-
Chemistry	WM	-	-	-	-	8	-
(LC)							
Foliage	Cd	0	54	0	0	-	0
Chemistry	Cu	7.5	0	0	0	-	0
(FC)	Pb	10	0	0	0	-	0
(*Pinus* and	S_{tot}	0	67	0	0	-	0
Quercus)			50	0	0	-	0
Litterfall	Cd	2.5	67	0	0	-	0
Chemistry	Cu	0	4	0	0	-	0
(LF)	Pb	10	67	0	0	-	0
(*Pinus* and	P_{tot}	0	8	0	0	-	0
Quercus)	S_{tot}	0	42	0	0	-	100*
	Zn	2.5	0	0	0	-	0
Groundwater Chemistry (GW)	pH	-	-	-	-	-	50

Table 10: *Percentage of variables in each biological programme having a significant positive correlation with a chemical variable.*
(Combinations with only one variable are indicated with an asterisk; - : not calculated)

Monitoring Programme	Variable	Inventory of Birds (BB)	Inventory Of Butterflies (BF)	Trunk Epiphytes (EP)	Leafminers (LM)	Lake Water Biology (LB)	Forest Damage (FD)
Air Chemistry (AC)	NH_3	5	8	0	0	-	0
	NH_4	45	25	0	67	-	0
	NH_{4tot}	15	0	0	0	-	0
	NO_2	5	0	0	0	-	100*
	NO_{3tot}	20	50	0	67	-	0
	SO_2	30	0	0	0	-	100*
	SO_4	25	50	0	33	-	100*
Deposition Chemistry (DC)	Cd	0	42	0	0	-	0
	Cr	10	58	0	0	-	0
	Ni	0	33	0	0	-	0
	NO_3	0	33	0	0	-	0
	Pb	0	8	0	0	-	0
	SO_4	0	0	0	0	-	100*
	Zn	5	17	0	0	-	0
Throughfall Chemistry (TF) (*Pinus* and *Quercus*)	Cd	10	58	0	83	-	0
	Cr	5	38	0	0	-	0
	NH_4	0	0	0	0	-	100*
	Ni	5	58	0	0	-	0
	Pb	0	8	0	0	-	0
	pH	10	0	0	0	-	0
	SO_4	0	0	0	0	-	100*
Stemflow Chemistry (SF) (*Pinus* and *Quercus*)	Cd	10	21	0	0	-	0
	Cr	7.5	54	0	0	-	100*
	SO_4	0	0	0	0	-	100*
Lakewater Chemistry (LC)	PO_4	-	-	-	-	25	-
	SO_4	-	-	-	-	8	-
Foliage Chemistry (FC) (*Pinus* and *Quercus*)	Cd	0	8	0	0	-	0
	P_{tot}	0	42	0	0	-	0
	S_{tot}	0	8	0	0	-	0
	Zn	0	0	0	33	-	0
Litterfall Chemistry (LF) (*Pinus* and *Quercus*)	Cd	0	8	0	0	-	0
	Cu	7.5	13	0	0	-	0
	P_{tot}	0	58	0	0	-	0
	S_{tot}	0	8	0	0	-	0
Groundwater Chemistry (GW)	NO_2	-	-	-	-	-	50

The results in Tables 9 and 10 are discussed in the paragraphs below for each biological monitoring programme.

4.2.1. Inventory of birds (BB)

In the Figures 1 to 10 in Appendix V the correlations between the number of breeding pairs of individual bird species and chemical substances in air, deposition and foliage, as specified in Table 8, are shown. The correlations between the measured chemicals on the presence of the birds is not clear. Most of the correlations found are positive; this means that for these species the number of birds is increasing with an increasing amount of the chemical. Only one species demonstrates a negative correlation with NH_3 in air, Two species show negative correlations with NH_4 in air, and six species are negatively correlated with SO_2 in air (5%, resp. 10% and 30% in Table 9). The other negative correlations in Table 9 are less or equal to 10% of the variables. Worth mentioning is that the negative correlations are from birds that showed a positive trend in time, for example for SO_2 in air *Columba oenas, Corvus corone* and *Lullula arborea* (Figure 1, Appendix IV; Figure 1, Appendix V). Influence of the chemical substances in the different compartments on the presence of birds can not be explained by these results. Desiccation and the presence of heavy metals are not correlated to the presence of birds in the Lheebroekerzand.

4.2.2. Inventory of butterflies (BF)

In the Figures 11 to 20 in Appendix V the correlations between the presence of butterflies and chemical substances in air, deposition and foliage, as specified in Table 8, are shown. Positive and negative correlations are found for butterflies (Table 9 and 10). The meaning of the positive correlations is not clear. The negative correlations found for concentrations of cadmium in leaves and needles (54-67% of the species), lead (67% of the species) and sulphur (50-42% of the species) could explain the declining trend of the number of butterflies in the Lheebroekerzand. The fact is that in leaves and needles an increase of the concentrations of lead and sulphur is found in time (Figure 14, 15 in Appendix IV). The available concentrations of the monitored chemicals in the compartments air (AC) and rainwater (DC, TF, SF) are not correlated with the presence of butterflies. Eutrophication and desiccation do not bother butterflies, but acidification of rainwater (DC, TF and SF) and the availability of sulphur and heavy metals as cadmium and lead in foliage (FC, LF) are correlated with the presence of butterflies.

4.2.3. Inventory of leafminers and Trunk Epiphytes (LM and EP)

The positive correlations for ammonium, nitrate and sulphate in air, cadmium in throughfall and zinc in foliage can not be explained for leafminers (Table 10, Figure 21-30 in Appendix V). No negative correlations for leafminers are found. For epiphytes no correlations are existing (Table 9-10; Figure 31-40 in Appendix V). None of the environmental themes have shown correlations with the presence of leafminers and epiphytes.

4.2.4. Lakewater biology (LB)

Negative correlations with ammonium in lake water are observed for the species *Arrenurus neumani* and *Peltodytes caesus* (Figure 41, Appendix V). A positive correlation with phosphate is calculated for the species *Peltodytes caesus* and *Plea minutissima* and the family *Pleidae*. With the presence of 256 species in Kliplo it can be stated that the concentrations of the monitored chemical substances in the lakewater are not correlated with the species composition of the macrofauna.

4.2.5. Forest damage (FD)

The positive correlations for the variable defoliation are of importance. The defoliation is expressed in percentages, so an increasing defoliation could be related to an increasing concentration of the chemical. In Figures 42 to 47 in Appendix V positive correlations are shown for NO_2, SO_2 and SO_4 in air, SO_4, NH_4 and Cr in rainwater, and NO_2 in groundwater. The negative correlations found for the pH in stemflow and in groundwater could indicate the influence of acidification on defoliation of trees. Acidification and eutrophication caused by nitrogen and sulphur do seem to have influence on the defoliation.

5. CONCLUSIONS AND RECOMMENDATIONS

With the data gathered in the Lheebroekerzand it was possible to detect and estimate trends in the monitoring data in time and to find correlations between biological and chemical variables.

5.1. Conclusions trends

To draw conclusions about trends in biological variables the next summarising remarks are of importance:

1. For birds 8 species showed a downward trend against 5 species an upward trend of the total of 69 observed species. The number of species, the total number of organisms and the Shannon Wiener Diversity Index did not change in time.
2. From the total of 24 observed species of butterflies 4 species showed a downward trend in time and none showed a statistically significant upward trend. The number of species did not change but the total number of organisms showed a decreasing trend. The Shannon Wiener Diversity Index showed an increasing trend in time.
3. For leafminers one of the 18 species showed a downward trend; the Shannon Wiener Diversity Index and the number of species were decreasing in time but the total number of organisms did not change.
4. The number of species of epiphytes is increasing in time.
5. From the total number of 256 species of macrofauna 2 species showed a downward trend against 5 species an upward trend. The number of species, the total number of organisms and the Shannon Wiener Diversity Index did not change.
6. For the monitoring programme Forest Damage the only variable with a downward trend was defoliation.

The conclusion concerning the biological variables is that there is no need to worry about the effects opf pollutants on birds, leafminers, epiphytes and macrofauna in the Lheebroekerzand, but the decrease in the butterfly community can be marked as an alarming indicator. The condition of the foliage is improving in time.

After summarising the reported results concerning the analysis of trends for chemical variables in different environmental compartments, it can be concluded that a substantial part of the monitored and analysed pollutant/compartment combinations indicate a downward trend. On the other hand concentrations of ammonium, nitrate and sulphate in lakewater are still showing an upward trend. Also the acidity of the lakewater did not decrease during the monitoring period.

The observed downward trends in chemical variables are also not yet detected for lead in the compartments of leaves and needles.

If the reduction in emissions in Europe will be continued in future years, it is to be expected that the amount of most chemical compounds in the compartments lakewater, leaves and needles will diminish in future as well.

5.2. Conclusions correlations

Concerning the determination of possible correlations between biological and chemical monitoring variables a careful conclusion can be that substances responsible for acidification, eutrophication, desiccation and the presence of heavy metals do not seem to have a significant influence on the presence of birds, leafminers, epiphytes and macrofauna. However, for the presence of butterflies it seems that there is a correlation with cadmium, lead and sulphur. For defoliation measured in the monitoring programme Forest Damage a correlation is found with sulphur, nitrogen and acidity. This could mean that acidification and eutrophication still have influence on the foliage of the trees in the Lheebroekerzand.

5.3. Recommendations

✓ More data of previous years has to be collected to obtain more overlapping data of biological and chemical variables and to ensure longer time-periods.

✓ Indirect correlations between variables should be studied.

✓ Trend and correlation analysis should be done with data of the international database of ICP-IM.

✓ With more data involved, it is possible to carry out a multivariate analysis.

✓ The decrease of the presence of butterflies in the Lheebroekerzand needs to be studied.

✓ To follow the recovery of the ecosystem more years of monitoring is necessary.

REFERENCES

Bakker, T.W.M., 1984: Het Dwingelderveld – Deelrapport: Geohydrologie, Nederland: Staatsbosbeheer, Vereniging tot Behoud van Natuurmonumenten, Universiteit van Amsterdam, Fysisch Geografisch en Bodemkundig Laboratorium.

Castel, I.I.Y., 1984: Het Dwingelderveld – Deelrapport: geologie, geomorfologie en bodemgesteldheid, Nederland: Staatsbosbeheer, Vereniging tot Behoud van Natuurmonumenten, Universiteit van Amsterdam, Fysisch Geografisch en Bodemkundig Laboratorium.

De Groot, A.C., W.J.G.M. Peijnenburg, M.A.G.T. van den Hoop, R. Ritsema and R.P.M. van Veen, 1998: Heavy metals in Dutch field soils: an experimental and theoretical study on equilibrium partitioning, Bilthoven, the Netherlands: National Institute for Public Health and the Environment (RIVM). Report no. 607220001

De Wit, T., 1976: Epiphytic Lichens and Air Pollution in the Netherlands. Leersum, The Netherlands: Research Institute for Nature Management (RIN). Verhandeling 8.

De Zwart, D., 1997: Ordination of the intergrated monitoring data gathered under auspices of ICP-IM (UN-ECE Convention on Long-Range Transboundary Air Pollution*): 1989-1994.* Bilthoven, the Netherlands: National Institute of Public Health and the Environment (RIVM). Report no. 259101006.

Dijkstra, A., H. Dekker, B. Takman, 1992: Dagvlinders in Drenthe. Assen, Nederland: Provincie Drenthe.

Forsius, M., S. Kleemola, J. Vuorenmaa and S. Syri, 2001: Fluxes and trends of nitrogen and sulphur compounds at integrated monitoring sites in Europe. Water, Air and Soil Pollution 130: 1641-1648.

Helsel, D.R., and R.M. Hirsch, 1995: Statistical methods in water resources. Amsterdam, The Netherlands: Elsevier Science B.V., ISBN 0444814639.

Hoentjen, B., 1993: Drentse vennen: betekenis, bedreigingen en beheer. Achtergrond, aanleiding en samenvatting van een onderzoek aan Drentse vennen, uitgevoerd in 1991, Assen, Nederland: Provincie Drenthe.

KNMI, 1961-1980: Maandelijks overzicht der weersgesteldheid. De Bilt, Nederland.

KNMI, 1981-1992: Maandoverzicht van het weer in Nederland. De Bilt, Nederland.

Masselink-Beltman, H.A., 1978: Korstmossen in Drenthe (inclusief verspreidingskaartjes van ruim 30 soorten). Assen, Nederland: Provinciale Waterstaat Drenthe.

Mathijssen-Spiekman, E.A.M., R. Erven and H.A.M. de Kruijf, 1994: Development of the integrated Monitoring Area Lheebroekerzand – The Netherlands, data of 1990, 1991 and 1992. Bilthoven, the Netherlands: National Institute of Public Health and the Environment (RIVM). Report no. 673710001.

Mathijssen-Spiekman, E.A.M. and R. Erven, 1995[a]*:* The Integrated Monitoring Area Lheebroekerzand - The Netherlands (data of 1993), Bilthoven, the Netherlands: National Institute of Public Health and the Environment (RIVM). Report no. 673710002.

Mathijssen-Spiekman, E.A.M. and D. de Zwart, 1995[b]: The Integrated Monitoring Area Lheebroekerzand - The Netherlands (data of 1994). Bilthoven, the Netherlands: National Institute of Public Health and the Environment (RIVM). Report no. 673710003.

Mathijssen-Spiekman, E.A.M, 1996*:* The Integrated Monitoring Area Lheebroekerzand - The Netherlands (data of 1995). Bilthoven, the Netherlands:-National Institute of Public Health and the Envrionment (RIVM). Report no. 673710004.

Mathijssen-Spiekman, E.A.M. and D. de Zwart, 1997: Evaluation of the Integrated Monitoring Activities executed by RIVM in the framework of the UN-ECE Convention on Long-Range Transboundary Air Pollution. Bilthoven, the Netherlands: National Institute of Public Health and the Environment (RIVM). ECO-note 97-08.

Mathijssen-Spiekman, E.A.M. and M.A.H. Wolters-Balk, 1998*:* The Integrated Monitoring Area Lheebroekerzand - The Netherlands (data of 1996). Bilthoven, the Netherlands: National Institute of Public Health and the Environment (RIVM). Report no. 259101008.

Mathijssen-Spiekman, E.A.M. and M.A.H. Wolters-Balk, 2001: The Integrated Monitoring Area Lheebroekerzand – The Netherlands (data of 1997-1998-1999). Bilthoven, the Netherlands: National Institute of Public Health and the Environment (RIVM). Report no. 607165001.

Sokal, R.R. and Rohlf, F.J., 1997: Biometry. The Principles and practice of statistics in biological research. 3rd edition. New York, United States of America: Freeman and Company.

UN-ECE, 1993: Manual for integrated monitoring, programme phase 1993-1996. Helsinki, Finland: National Board of Waters and the Environment, Environment Data Centre, ICP-IM Programme Centre.

Van Dam, H., 1987: Monitoring of chemistry, macrophytes and diatoms in acidifying moorland pools. Leersum, The Netherlands: Research Institute for Nature Management (RIN). Report 87/19.

Van Dam, H., 1996: Partial recovery of moorland pools from acidification: indications by chemistry and diatoms. Netherlands Journal of Aquatic Ecology 30 (2-3): 203-218.

Van Dijk, A.J., and E.V. Koopman, 1988: Dwingelderveld Avifauna, ‘s Graveland, Nederland: Vereniging tot behoud van Natuurmonumenten.

Van Dijk, A.J., A. Boele, F. Hustings, D. Zoetebier and R. Meijer, 1999: Broedvogel Monitoring Project Jaarverslag 1996-97, Beek-Ubbergen, Nederland: Samenwerkende Organisaties Vogelonderzoek Nederland (SOVON). Monitoringrapport 1999/03.

Van Dobben, H.F., J. Mulder, H. van Dam and H. Houweling, 1992: Impact of acid atmospheric deposition on the biogeochemistry of moorland pools and surrounding terrestrial environment. Wageningen, The Netherlands: Pudoc Scientific Publishers

Van Herk, C.M., 1993: Korstmossen en zure depositie in Drenthe en Friesland. Ecologische atlas, Assen, Nederland: Provincie Drenthe.

Van Herk, C.M., 1998: Korstmossen van het Lheebroekerzand (Drenthe*).* Soest, Nederland:

Lichenologisch Onderzoekbureau Nederland (LON).

Van Swaay, C. A.M. and R. Ketelaar, 2000*:* Dagvlinders en libellen onder de meetlat: jaarverslag 1999. Wageningen, Nederland: De Vlinderstichting. Rapport VS2000.06.

Veling, K., 1995: Dagvlindermonitoring Lheebroekerzand, 1994. Wageningen, Nederland. De Vlinderstichting. Rapportnr 95.04.

Verspui, K., 1991: Dwingelderveld: libellen en dagvlinders. Pesse, Drenthe-zuid, Nederland: Staatsbosbeheer (SBB) en ‘s Graveland, Nederland: Vereniging tot behoud van Natuurmonumenten.

Wesselink, L.G., 1994: Time trends and mechanisms of soil acidification. PhD thesis. Wageningen, The Netherlands: Agricultural University of Wageningen, ISBN 9054852895.

Wijnhoff, I., J. van der Made and C. van Swaay: 1992. Dagvlinders van de Benelux. Wageningen, Nederland: Stichting Uitgeverij KNNV en De Vlinderstichting. ISBN 90-5011-034-7.

Zegers, P.M., 1974: Waardering van het landelijk gebied aan de hand van broedvogeltellingen in Zuidwest-Drenthe. Assen, Nederland: Provinciale Planologische Dienst van Drenthe.

APPENDIX I AVAILABLE DATA WITH THEIR SOURCE

Monitoring Programme	Year	Organisation	Contact-person/ Researcher	Literature
Inventory of birds (BB)	1928-1987			A.J.van Dijk (1988)
	1974			P.M. Zegers (1974)
	1987-1998	SOVON/SBB	A.J. van Dijk/ P. Kerssies	
Inventory of butterflies (BF)	1981-1988	Vlinderstichting		A. Dijkstra (1992)
	1990			K. Verspui (1991)
	1994-1999		K. Veling M. Scheper K. van Eerde J. Kleine	
Inventory of leafminers (LM)	1993-1997	TINEA	J.H. Kuchlein	
		Insectenonderzoek bureau Donner	J.H. Donner	
Inventory of plants (BV)	1988, 1999	Alterra	A. van Hees	
			S. Klerks	
Trunk Epiphytes (EP)	1951-1968	WUR	Biologisch station Wijster	C.M. van Herk (1998)
	1971-1976			T. de Wit (1976)
	1978			H.A. Masselink-Beltman (1978)
	1989-1999	LON	C.M. van Herk	C.M. van Herk (1993)
Hydrobiology of lakes (LB)	1992-1998	Zuiveringsschap Drenthe	G. Duursema	
	1999	Waterschap Reest en Wieden	M. Fagel	
Vegetation (VG)	1988, 1999	Alterra	A. van Hees	
			S. Klerks	
Forest Damage (FD)	1984-1999	SBB	P. Kerssies	

Monitoring Programme	Year	Organisation	Contact-person/ Researcher	Literature
Climate (AM)	1961-1980	KNMI		KNMI, 1961-1980
	1980-1992			KNMI, 1981-1992
	1993-1999	RIVM-ECO	R. van Veen	
			G.J. van Dijk	
			M. Wolters	
Air Chemistry (AC)	1984-1999	RIVM-LLO	E. Rentinck	
			A. Stolk	
Precipitation Chemistry (DC)	1961-1981	KNMI		KNMI, 1961-1980
				KNMI, 1981-1992
	1982-1985			H.F. van Dobben (1992)
	1990-1992	RIVM-LLO	E. Rentinck	
			A. Stolk	
	1993-1999	RIVM-ECO	R. van Veen	
			M. Wolters	
Throughfall and Stemflow Chemistry (TF, SF)	1982-1985			H.F. van Dobben (1992)
	1993-1999	RIVM-ECO	R. van Veen	
			M. Wolters	
Metal Chemistry in Mosses (MC)	1993-1999	RIVM-ECO	M. Wolters	
Soil Chemistry (SC)	1993	RIVM-LBG		
	1997	RIVM-ECO	A. C. de Groot	A.C. de Groot (1998)
Soil Water Chemistry (SW)	1997	RIVM-ECO	A. C. de Groot	A.C. de Groot (1998)
Groundwater Chemistry (GW)	1980-1999	RIVM-LBG	P. Lagas	
			H. Prins	
Lake Water Chemistry (LC)	1924-1995	AquaSense-TEC	H. van Dam	H. van Dam (1987)
	1996-1999	RIVM-ECO	M. Wolters	
Foliage and Litterfall Chemistry (FC, LF)	1993-1999	RIVM-ECO	M. Wolters	

APPENDIX II DATA HELD IN THE DATABASE OF THE LHEEBROEKERZAND

PARAMETER	BB	BF	LM	BV	EP	LB	VG	FD	AM	AC	DC	TF	SF	MC	SC	SW	GW	LC	FC	LF
Al											96-99	96-99	96-99	96-99		97	87-99	78-99	96-99	96-99
ALK																		72-99		
ANF								84-99												
As											93-99	93-99	93-99	94-99	97	97	80-99	96-99	96-99	96-99
Ba														96-99			90-99		96-99	96-99
Ca											82-99	82-99	82-99	93-99		97	80-99	68-99	93-99	93-99
Cd											84-99	93-99	93-99	93-99	93, 97	97	90-99		93-99	93-99
CEC_E															93					
Cl											82-99	82-99	82-99			97	80-99	58-99		
CNR.N320																		81-91		
CNR.N455																		90-99		
Co														96-99					96-99	96-99
COR														93	93,97	97	80-94	78-98		
CORT																			93-99	93-99
COVE				88, 99			88, 99													
CP						92-99														
Cr											93-99	93-99	93-99	93-99	93, 97	97	90-99	96-99	96-99	96-99
CTY.25											82-99	82-99	82-99			97	80-92	68-99		
Cu											84-99	93-99	93-99	93-99	93, 97	97	90-99	96-99	93-99	93-99
DAM								84-99												
DBH							88, 99	84-99												
DEFO								84-99												
DISC								87-95												
DIX_SW						92-99														
F											82-96	82-85	82-85							
Fe											84-96			93-99	93, 97	97	89-99	76-96	96-99	96-99
HCROW								99												
HEIG							88, 99	93-99												
Hg														96					96	96
HH									61-99											
K											82-99	82-99	82-99	93-99		97	80-99	68-99	93-99	93-99
Ldep																				93-99
Mg											82-99	82-99	82-99	93-99		97	80-99	72-99	93-99	93-99
Mn											84-92			93-99	93	97	90-99		93-99	93-99
Mo														96					96	96
Na											82-99	82-99	82-99	93-99		97	80-99	68-99	93-99	93-99
NH_3										92-99										
NH_4										90-99	82-99	82-99	82-99				80-99	68-99		
NH_4tot										92-99										
Ni											84-99	93-99	93-99	93-99	97	97	80-99	96-99	96-99	96-99
NO_2										84-99	93-94	93-94	93-94					78-95		
NO_3										90-99	82-99	82-99	82-99			97	80-99	68-99		
Ntot														93	97				93-99	93-99
NUM							88, 99													
O_2																		75-99		
O_3										84-99										
Pb											84-99	93-99	93-99	93-99	93, 97	97	90-94	96-99	93-99	93-99
pH											82-99	82-99	82-99		93, 97	97	80-99	24-99		
PO_4											82-99	82-99	82-99			97	84-93	73-99		
Ptot														96-99	93		80-99	72-96	94-99	94-99
RET																			93-99	
RR											61-99	82-99	82-99							
Sb														96-99					96-99	96-99
SECS								84-99												
SiO_2														96-99			84	81-99	96-99	96-99
SO_2										84-99										
SO_4										84-99	82-99	82-99	82-99			97	80-99	72-99		
SOL_G									94-99											
SOL_U									92-96											
SPPV	28-98	81-99	93-97		51-99	92-99														
Sr														96-99			90-99		96-99	96-99
Stot														96-99					94-99	94-99
T									61-99									75-99		
Ti														96-99					96-99	96-99
V											84-92			96-99					96-99	96-99
WCROW								98-99												
WL																	90-99			
WM (NAP)																		82-98		
Zn											89-99	93-99	93-99	93-99	93, 97	97	80-99	96-99	93-99	93-99

APPENDIX III INFORMATION STORED IN THE DATABASE

The biological values are all stored with the following information (exemplary):

BB = Subprogramme code (Inventory of Birds)

NL01 = Country code and area number (the Netherlands, Lheebroekerzand)

SBB = Institution code (State Forestry Service)

8606 = Date (YYMM = year and month)

95 = Spatial pool (size of area in hectare used for inventory)

AEGI CAU = Species code (standard abbreviation code (see Manual))

A1 = List (species list ICP-IM)

0.1 = Value (number of observed species per hectare)

\- = Quality flag (codes for extra information, for example V=species verified but value not given)

Physical and chemical values are stored with the extra information (exemplary):

DC = Subprogramme code (Precipitation chemistry)

NL01 = Country code and area number (the Netherlands, Lheebroekerzand)

VM = Institution code (RIVM)

001 = Station code

BULK = Medium code (bulk precipitation)

IM = List (variables list ICP-IM)

150 = Level (sampling height or depth)

9402 = Date (YYMM = year and month)

5 = Spatial pool (number of samples)

PO4P _F = Parameter code ICP-IM (includes way of sampling and analysing)

DA = Variables list ICP-IM (includes unit)

5.25 = Observed value

W = Status flag = codes to identify value (X = arithmetic mean, W = weighed mean, S = sum, M = mode)

\- = Quality flag codes for extra information (E=estimated from measured value; L=less than detection limit)

APPENDIX IV GRAPHS OF TRENDS

Figure 1: *Trends in number of birds per species (subprogramme BB).*

Figure 2: *Trends in number of butterflies per species or biological group (subprogramme BF).*

Figure 3: *Trends in number of leafminers and the Shannon-Wiener diversity index (subprogramme LM).*

Figure 4: *Trends in number of species of epiphytes (subprogramme EP).*

Figure 5: *Trends in number of organisms per species of the macrofauna (subprogramme LB).*

Figure 6: *Trend in defoliation for trees (subprogramme FD).*

Figure 7: *Trends in chemical variables for air (subprogramme AC).*

Figure 8: *Trends in chemical variables for deposition (subprogramme DC).*

Figure 9: *Trends in chemical variables for throughfall precipitation for Pinus sylvestris (PS) and Quercus robur (QR, subprogramme TF).*

Figure 10: *Trends in chemical variables for stemflow precipitation for Pinus sylvestris (PS) and Quercus robur (QR, (subprogramme SF).*

Figure 11: *Trends in chemical variables for mosses (subprogramme MC).*

Figure 12: *Trends in chemical variables for groundwater, depth 1200 cm and 3000 cm below ground level (subprogramme GW).*

Figure 13: *Trends in chemical variables for lake-water (subprogramme LC).*

Figure 14: *Trends in chemical variables for living needles (PS) and leaves (QR, subprogramme FC).*

Figure 15: *Trends in chemical variables for dead needles (PS) and leaves (QR, subprogramme LF).*

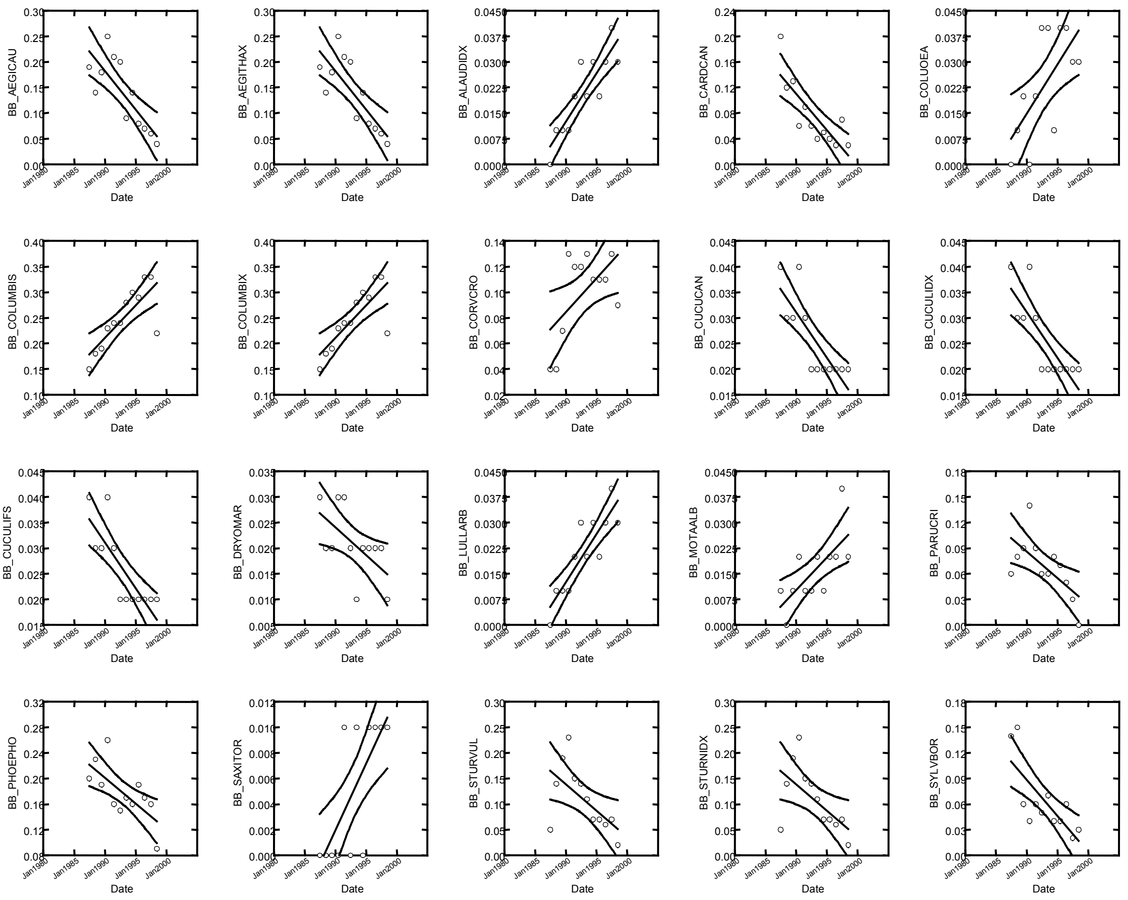


Figure 1: Trends in number of birds per species (subprogramme BB).

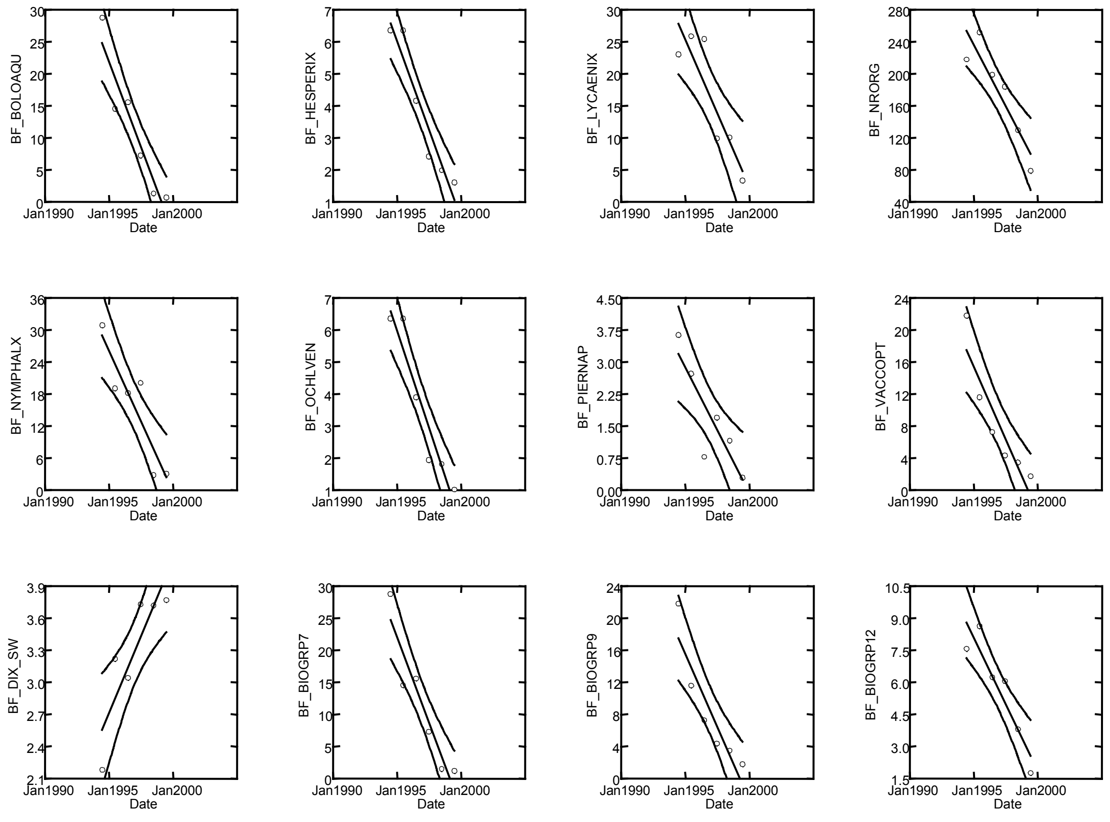


Figure 2: Trends in number of butterflies per species or biological group (subprogramme BF).

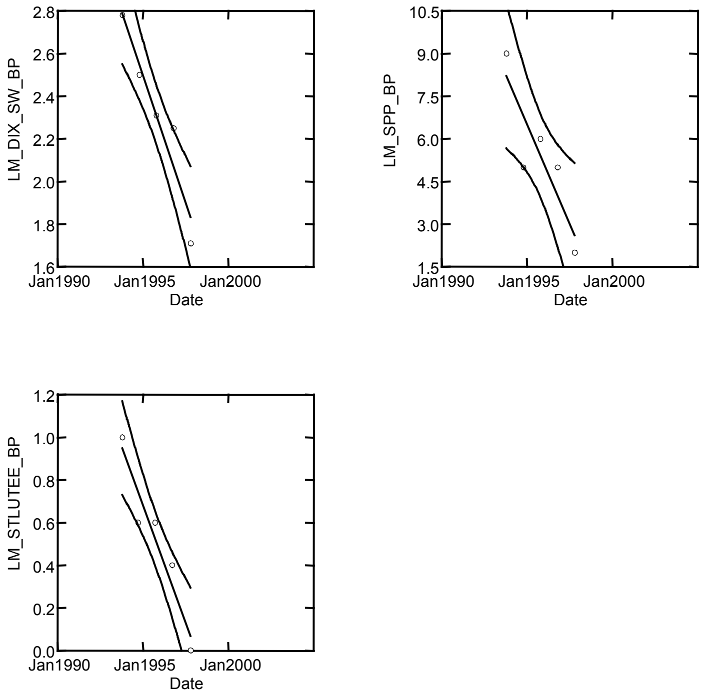


Figure 3: *Trends in number of leafminers and the Shannon-Wiener diversity index (subprogramme LM).*

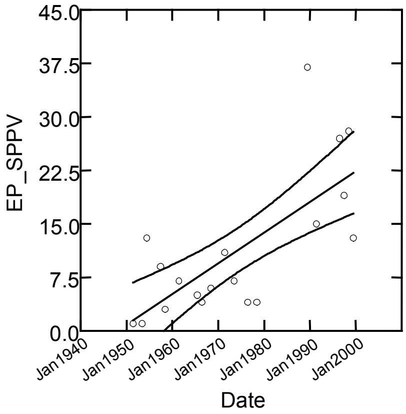


Figure 4: *Trends in number of species of epiphytes (subprogramme EP).*

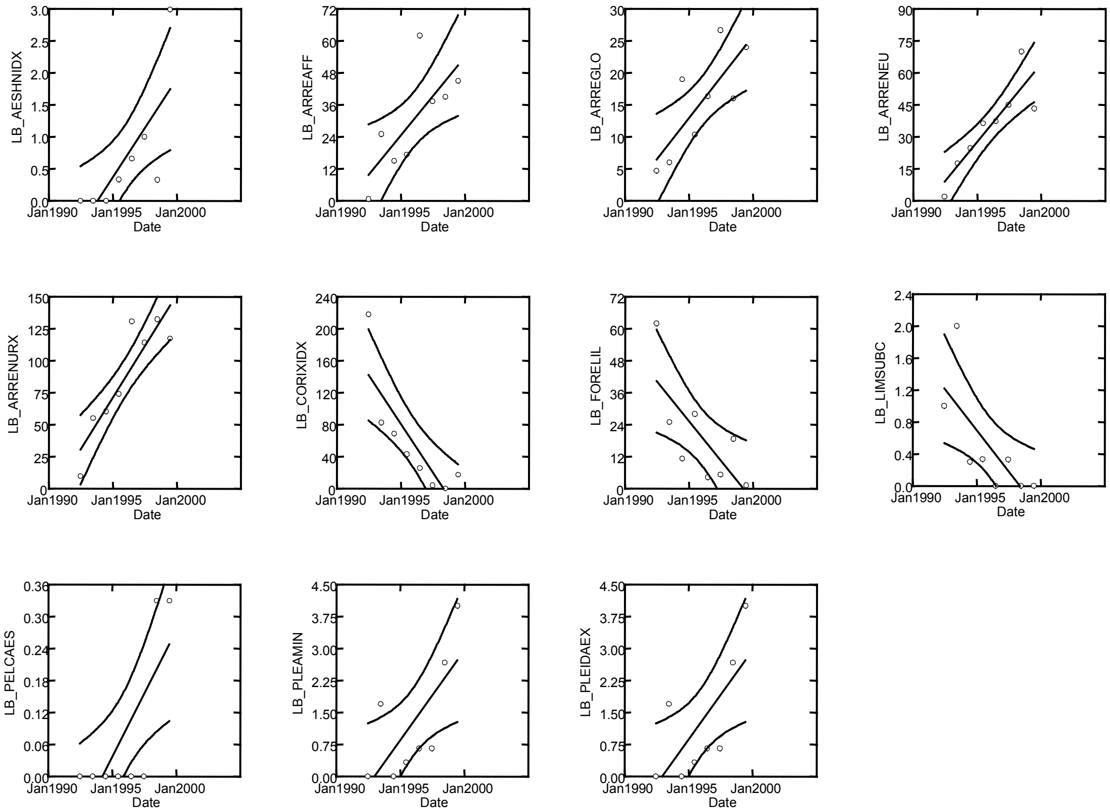


Figure 5: *Trends in number of organisms per species of the macrofauna (subprogramme LB).*

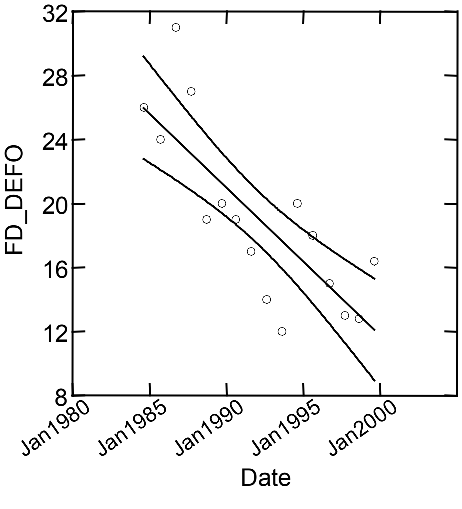


Figure 6: *Trend in defoliation for trees (subprogramme FD).*

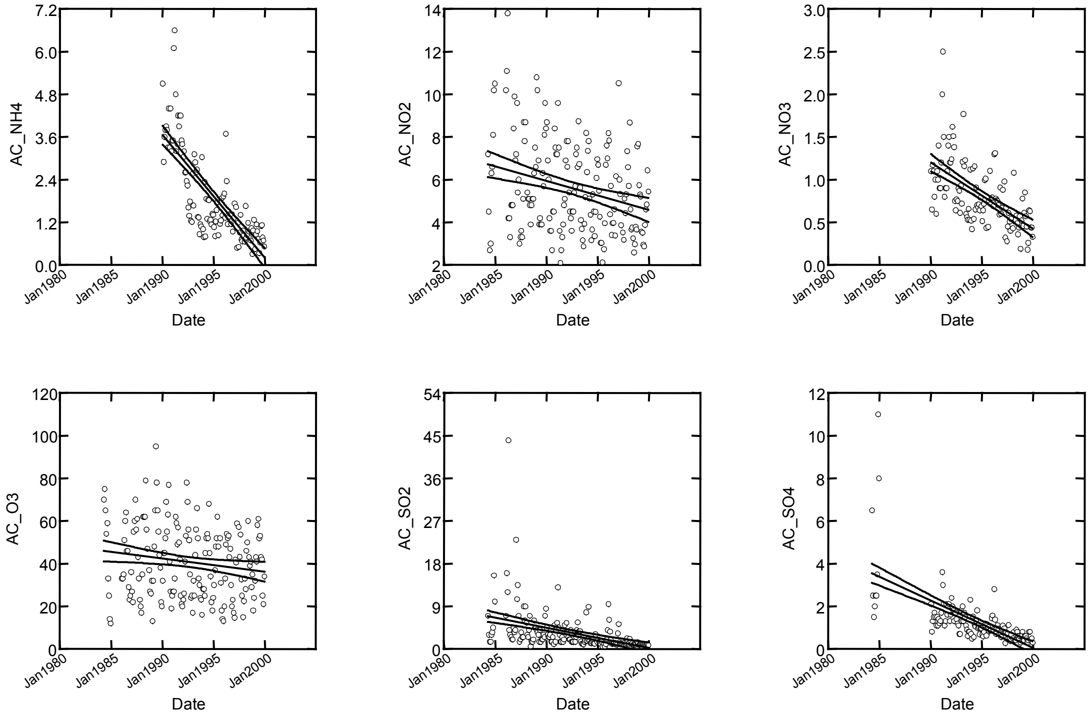


Figure 7: Trends in chemical variables for air (subprogramme AC).

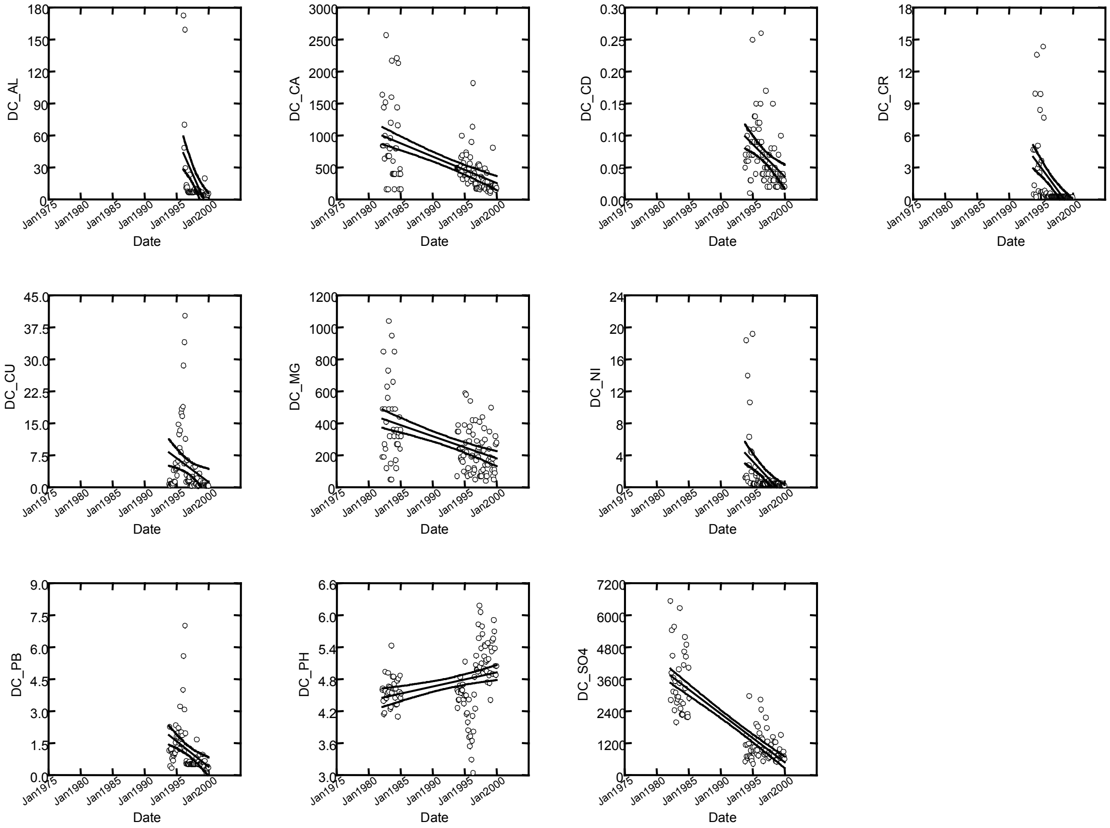


Figure 8: Trends in chemical variables for deposition (subprogramme DC).

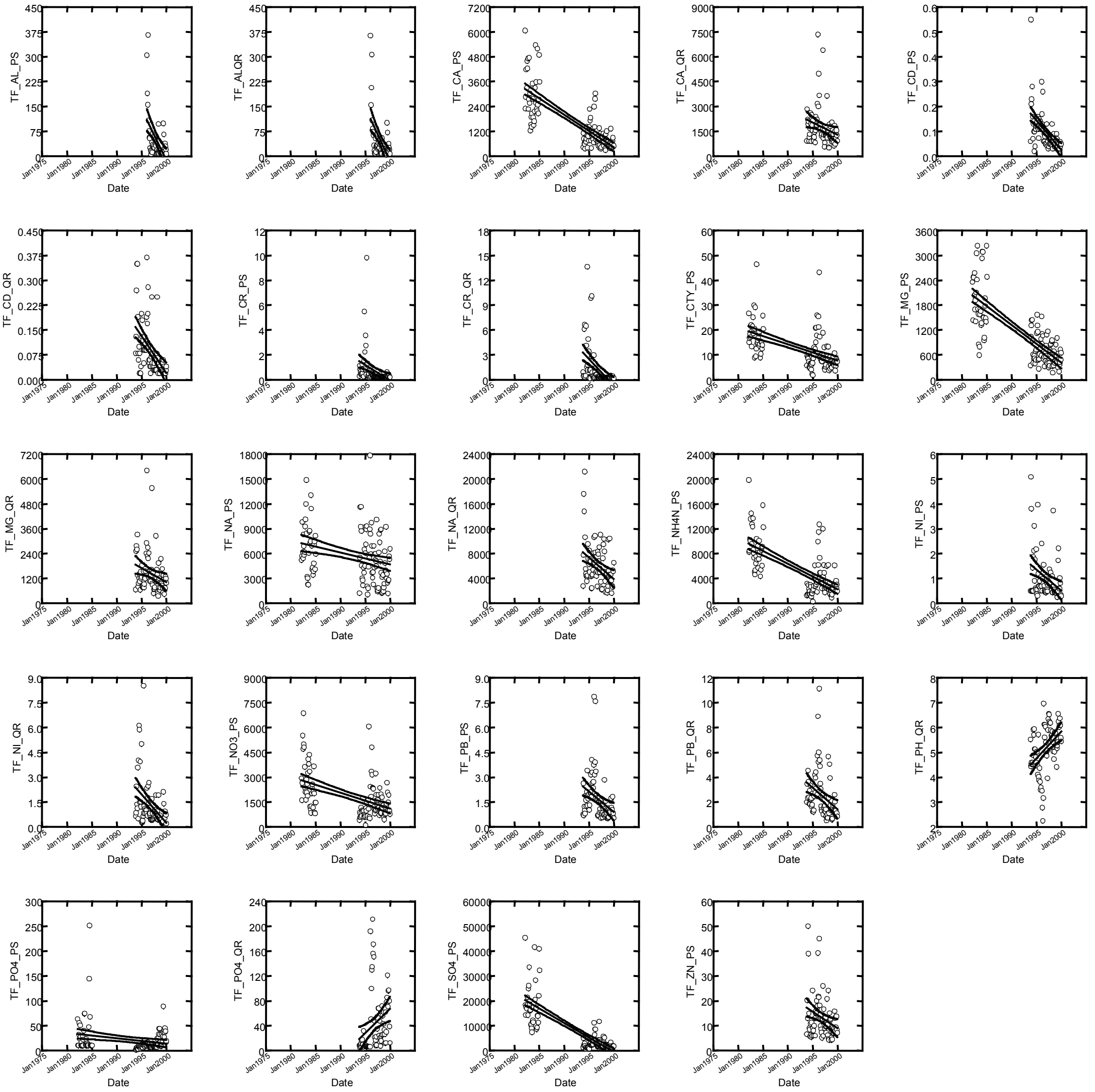


Figure 9: *Trends in chemical variables for throughfall precipitation for Pinus sylvestris (PS) and Quercus robur (QR, subprogramme TF).*

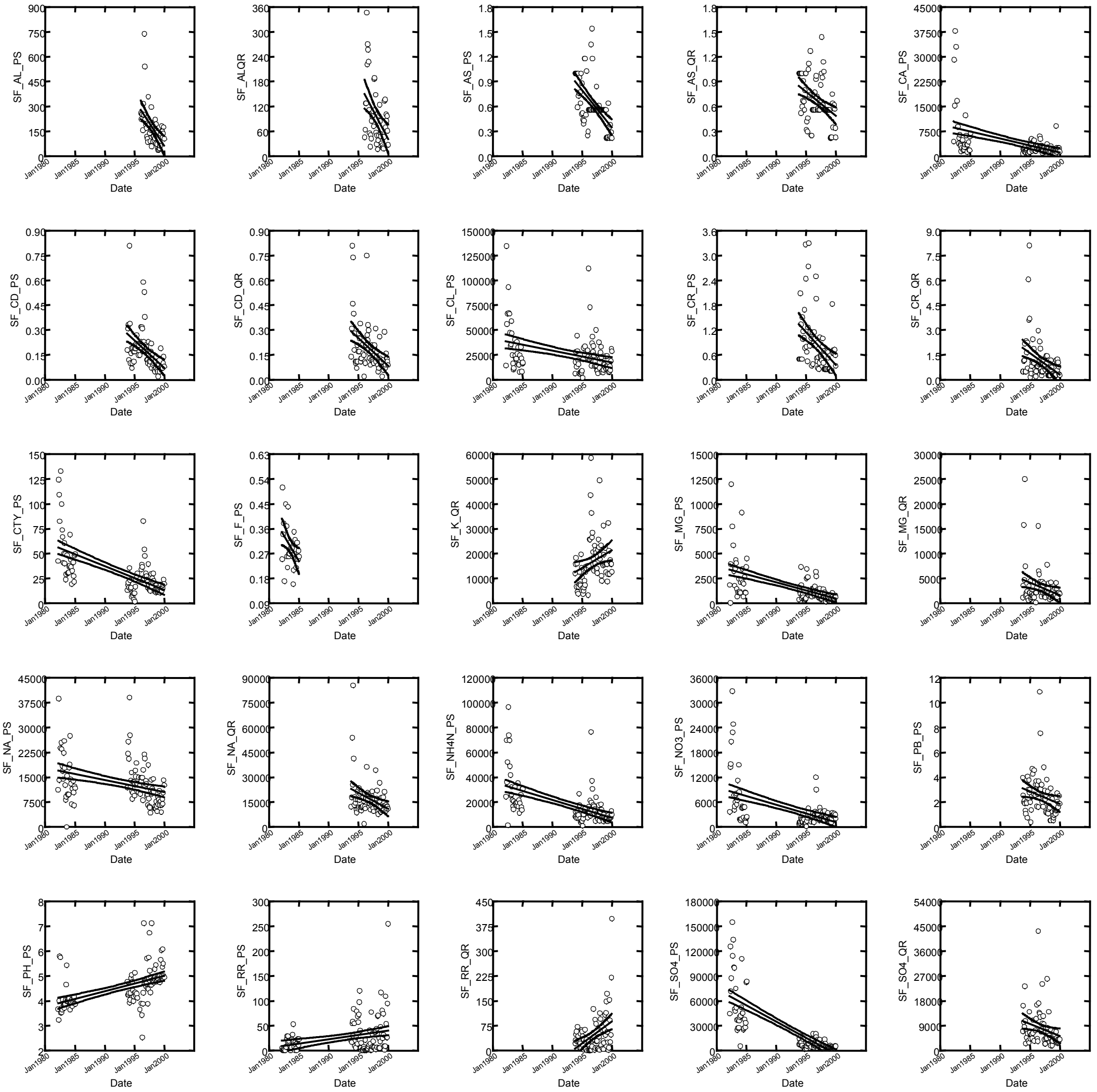


Figure 10: *Trends in chemical variables for stemflow precipitation for Pinus sylvestris (PS) and Quercus robur (QR, subprogramme SF).*

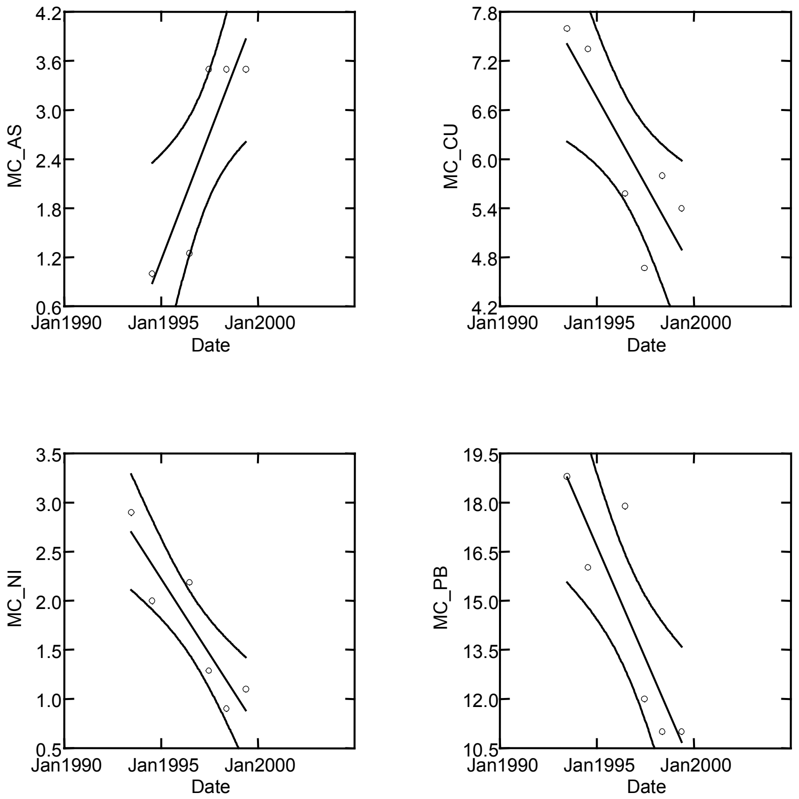


Figure 11: *Trends in chemical variables for mosses (subprogramme MC).*

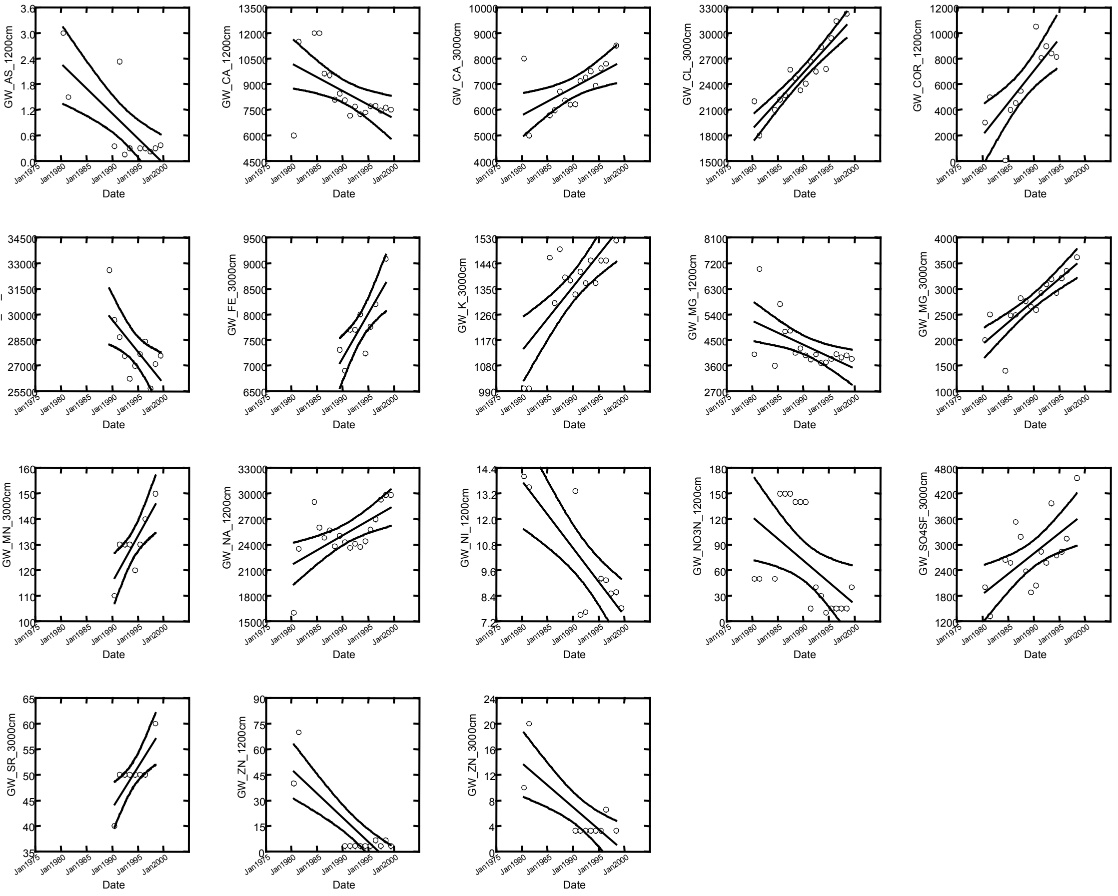


Figure 12: *Trends in chemical variables for groundwater, depth 1200 cm and 3000 cm below ground level (subprogramme GW).*

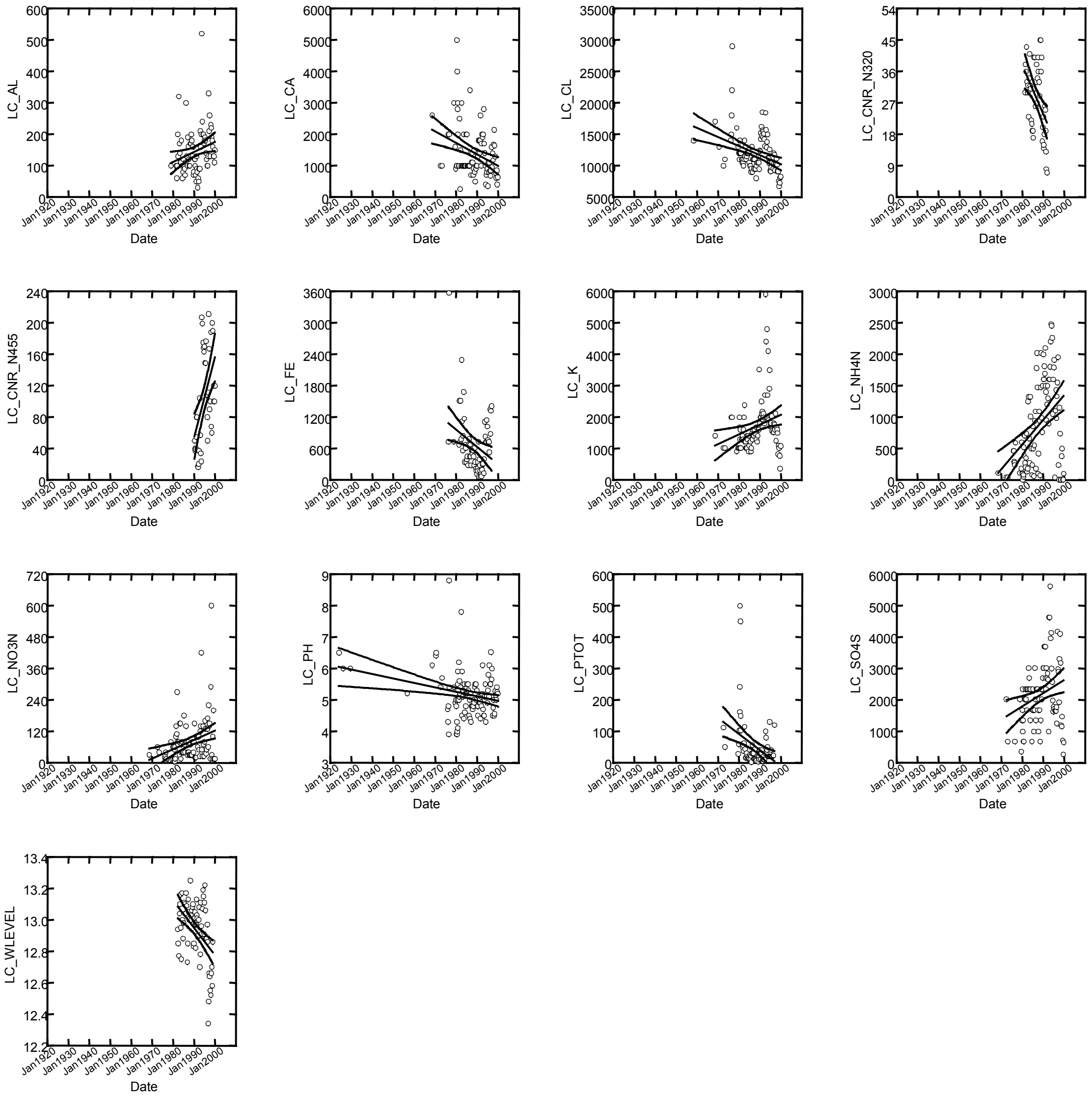


Figure 13: *Trends in chemical variables for lake-water (subprogramme LC).*

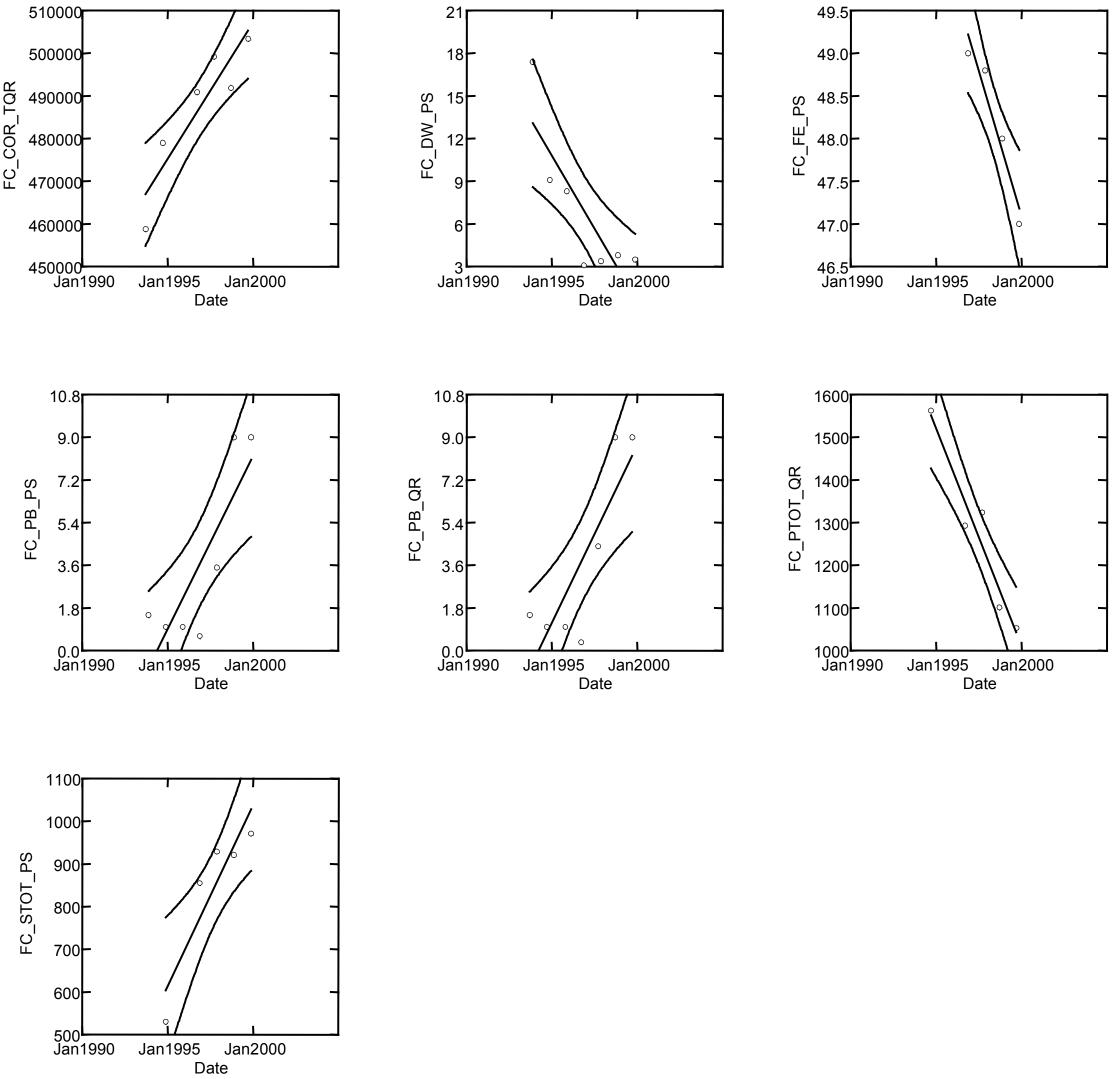


Figure 14: Trends in chemical variables for living needles (PS) and leaves (QR, subprogramme FC).

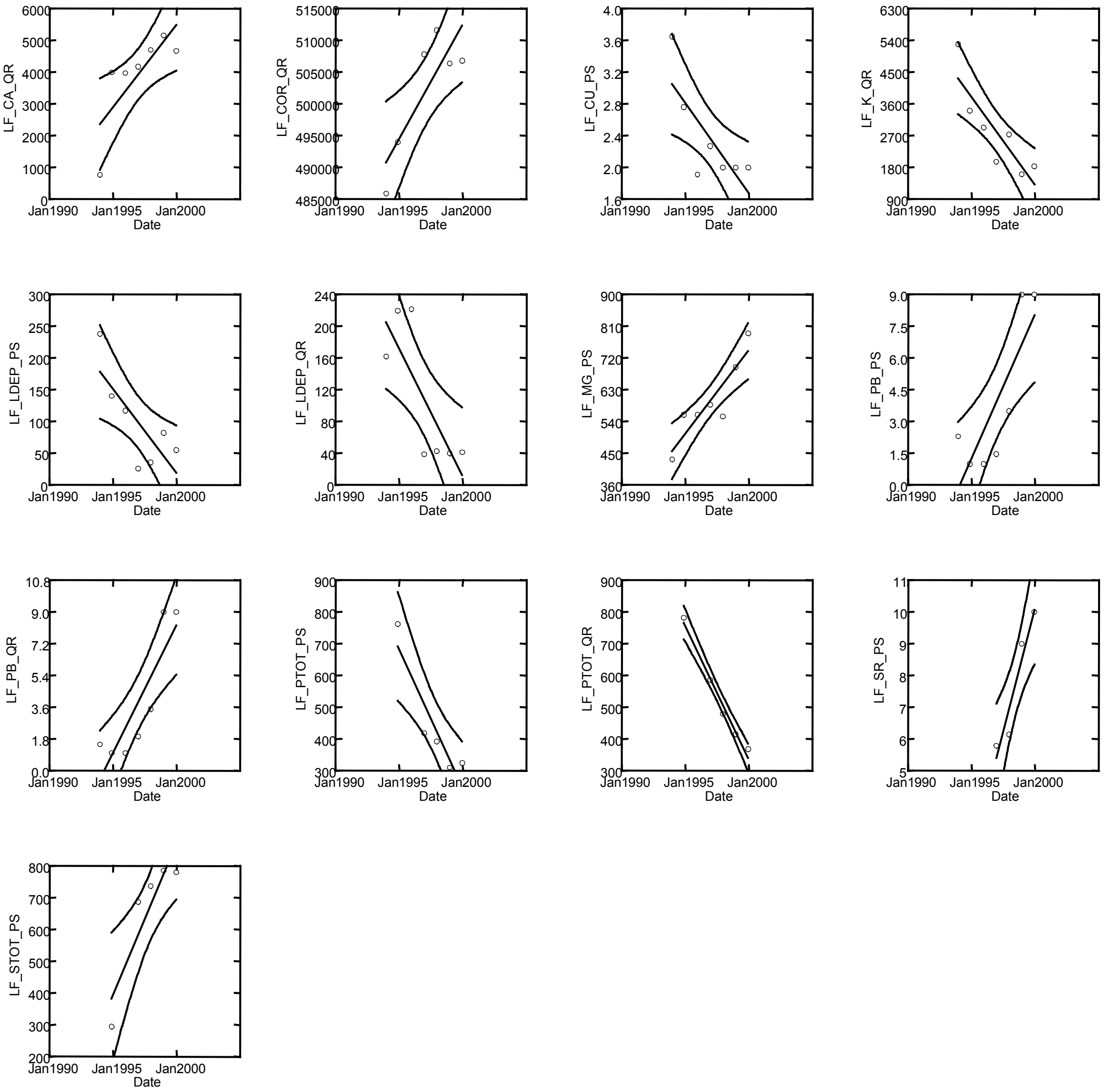


Figure 15: Trends in chemical variables for dead needles (PS) and leaves (QR, subprogramme LF).

APPENDIX V GRAPHS OF CORRELATIONS

Figure 1: Scatterplots between number of birds per species and chemical substances in air.

Figure 2: Scatterplots between number of birds per species and chemical substances in deposition.

Figure 3: Scatterplots between number of birds per species and chemical substances in throughfall of Pinus sylvestris.

Figure 4: Scatterplots between number of birds per species and chemical substances in throughfall of Quercus robur.

Figure 5: Scatterplots between number of birds per species and chemical substances in stemflow water of Pinus sylvestris.

Figure 6: Scatterplots between number of birds per species and chemical substances in stemflow water of Quercus robur.

Figure 7: Scatterplots between number of birds per species and chemical substances in living needles.

Figure 8 Scatterplots between number of birds per species and chemical substances in living leaves.

Figure 9: Scatterplots between number of birds per species and chemical substances in dead needles.

Figure 10: Scatterplots between number of birds per species and chemical substances in dead leaves.

Figure 11: Scatterplots between number of butterflies per species or biological group and chemical substances in air.

Figure 12: Scatterplots between number of butterflies per species or biological group and chemical substances in depositon.

Figure 13: Scatterplots between number of butterflies per species or biological group and chemical substances in throughfall water of Pinus sylvestris.

Figure 14: Scatterplots between number of butterflies per species or biological group and chemical substances in throughfall water of Quercus robur.

Figure 15: Scatterplots between number of butterflies per species or biological group and chemical substances in stemflow water of Pinus sylvestris.

Figure 16: Scatterplots between number of butterflies per species or biological group and chemical substances in stemflow water of Quercus robur.

Figure 17: Scatterplots between number of butterflies per species or biological group and chemical substances in living needles.

Figure 18: Scatterplots between number of butterflies per species or biological group and chemical substances in living leaves.

Figure 19: Scatterplots between number of butterflies per species or biological group and chemical substances in dead needles.

Figure 20: Scatterplots between number of butterflies per species or biological group and chemical substances in dead leaves.

Figure 21: Scatterplots between number of species of leafminers and chemical substances in air.

Figure 22: Scatterplots between number of species of leafminers and chemical substances in

deposition.

Figure 23: Scatterplots between number of species of leafminers and chemical substances in throughfall water of Pinus sylvestris.

Figure 24: Scatterplots between number of species of leafminers and chemical substances in throughfall water of Quercus robur.

Figure 25: Scatterplots between number of species of leafminers and chemical substances in stemflow water of Pinus sylvestris.

Figure 26: Scatterplots between number of species of leafminers and chemical substances in stemflow water of Quercus robur.

Figure 27: Scatterplots between number of species of leafminers and chemical substances in living needles.

Figure 28: Scatterplots between number of species of leafminers and chemical substances in living leaves.

Figure 29: Scatterplots between number of species of leafminers and chemical substances in dead needles.

Figure 30: Scatterplots between number of species of leafminers and chemical substances in dead leaves.

Figure 31: Scatterplots between number of species of epiphytes and chemical substances in air.

Figure 32: Scatterplots between number of species of epiphytes and chemical substances in deposition.

Figure 33: Scatterplots between number of species of epiphytes and chemical substances in throughfall water of Pinus sylvestris.

Figure 34: Scatterplots between number of species of epiphytes and chemical substances in throughfall water of Quercus robur.

Figure 35: Scatterplots between number of species of epiphytes and chemical substances in stemflow water of Pinus sylvestris.

Figure 36: Scatterplots between number of species of epiphytes and chemical substances in stemflow water.

Figure 37: Scatterplots between number of species of epiphytes and chemical substances in living needles.

Figure 38: Scatterplots between number of species of epiphytes and chemical substances in living leaves.

Figure 39: Scatterplots between number of species of epiphytes and chemical substances dead needles.

Figure 40: Scatterplots between number of species of epiphytes and chemical substances in dead leaves.

Figure 41: Scatterplots between number of organisms per species of the macrofauna and chemical substances in lakewater.

Figure 42: Scatterplots between defoliation of Pinus sylvestris and chemical substances in air.

Figure 43: Scatterplots between defoliation of Pinus sylvestris and chemical substances in deposition.

Figure 44: *Scatterplots between defoliation of Pinus sylvestris and chemical substances in throughfall water of Pinus sylvestris*

Figure 45: *Scatterplots between defoliation for trees and chemical substances in stemflow water of Pinus sylvestris.*

Figure 46: *Scatterplots between defoliation for trees and chemical substances living needles.*

Figure 47: *Scatterplots between defoliation for trees and chemical substances in dead needles.*

Figure 48 *Scatterplots between defoliation for trees and chemical substances in groundwater (1200 cm).*

Figure 49: *Scatterplots between defoliation for trees and chemical substances in groundwater (3000 cm).*

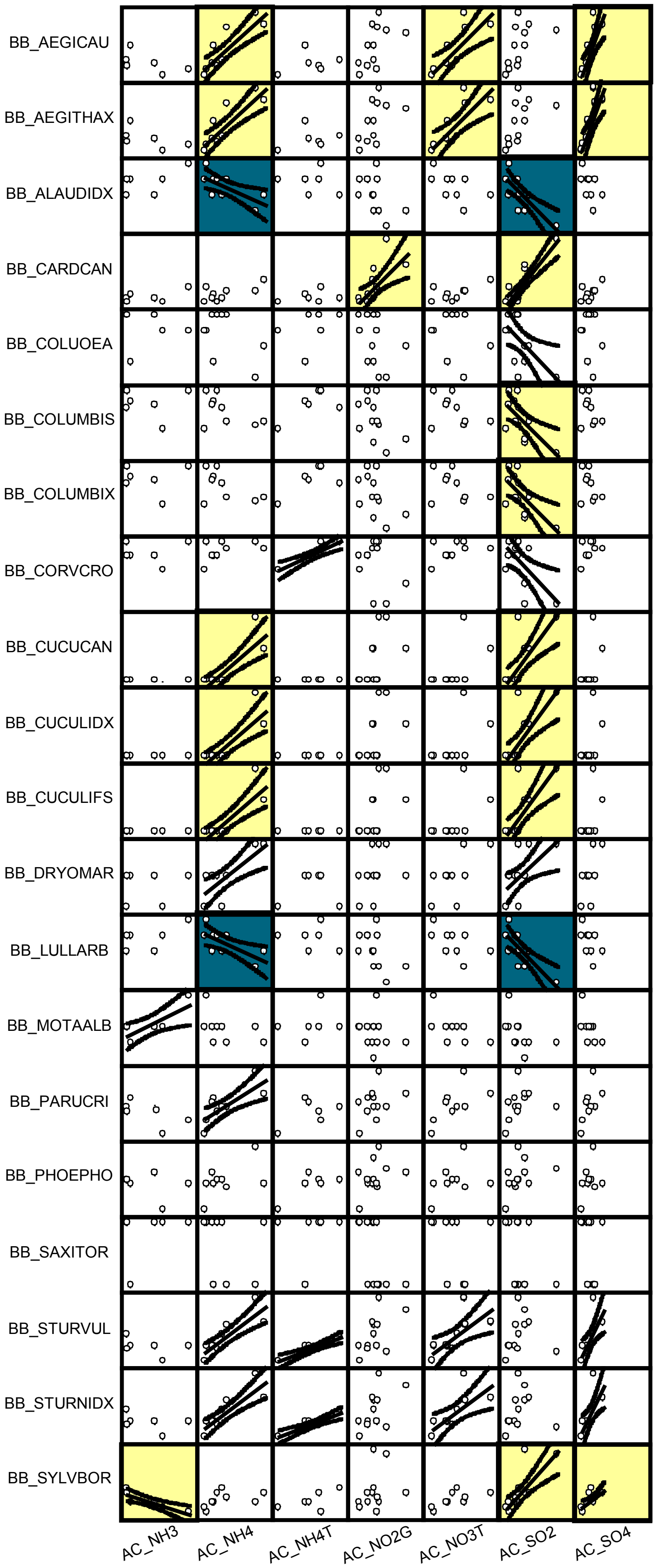


Figure 1: *Scatterplots between number of birds per species and chemical substances in air.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

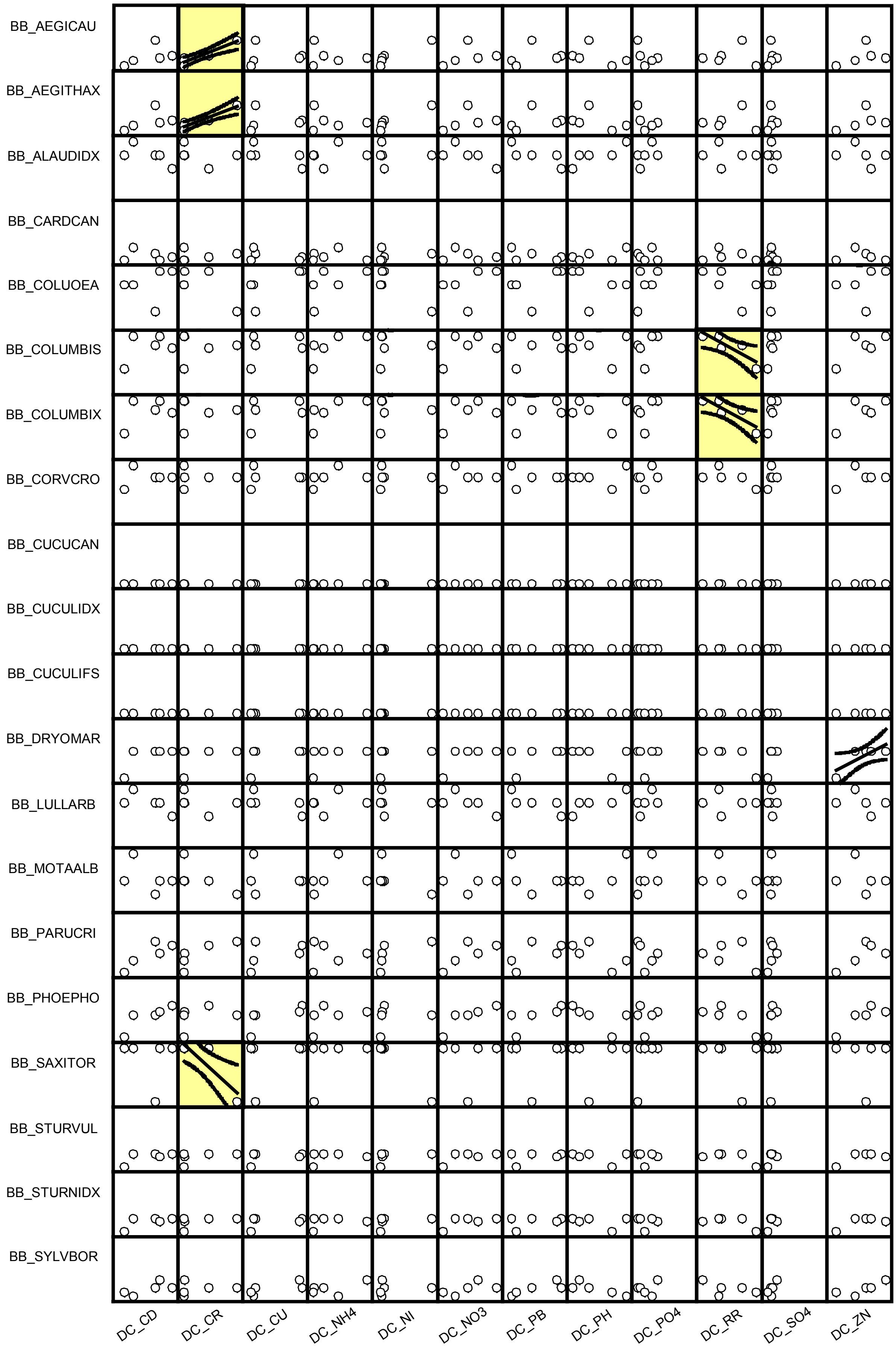


Figure 2: *Scatterplots between number of birds per species and chemical substances in deposition.*

Fitted lines:

P < 0.05	P < 0.01	P< 0.001

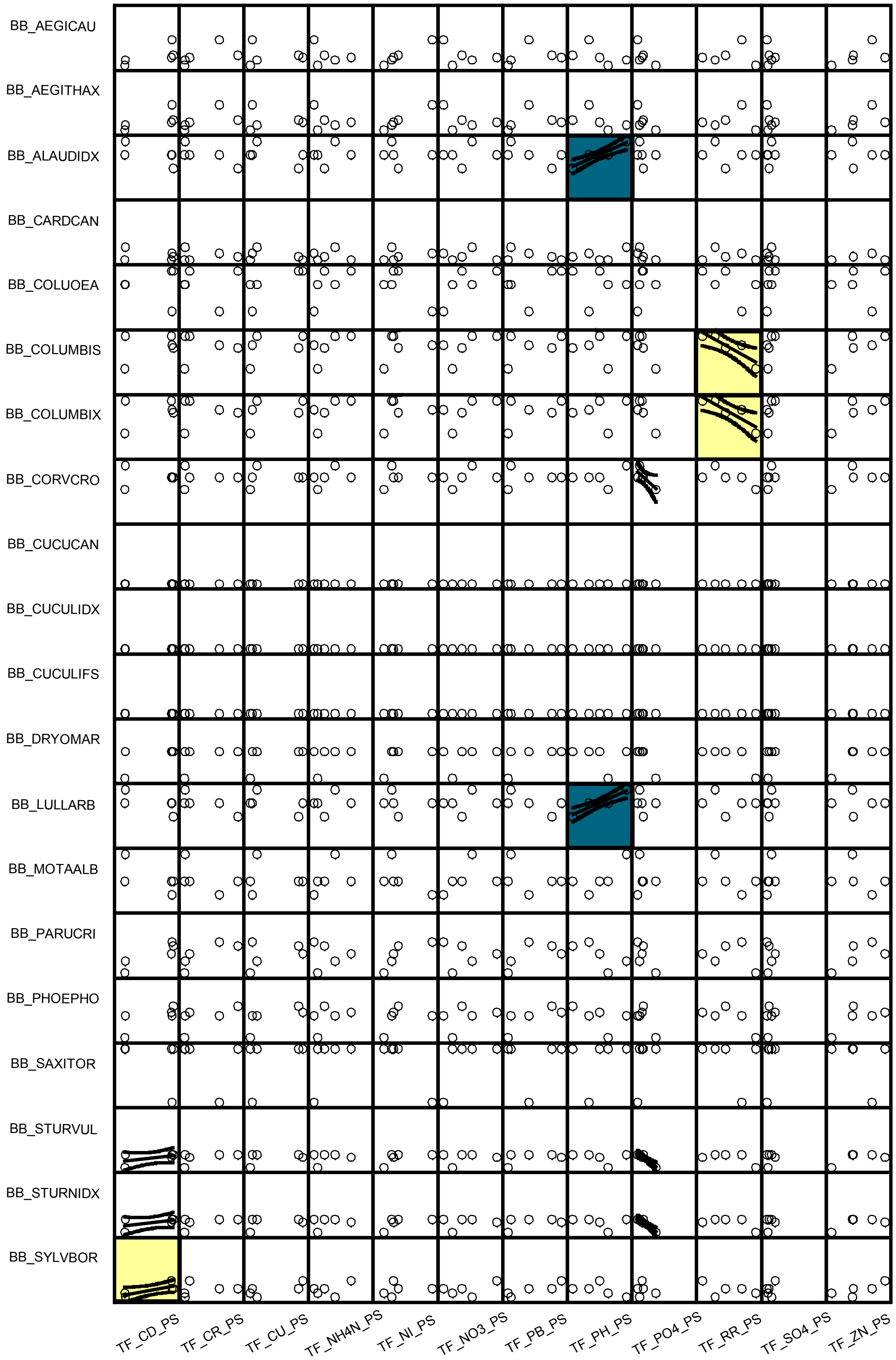


Figure 3: *Scatterplots between number of birds per species and chemical substances in throughfall of Pinus sylvestris.*

Fitted lines:

$P < 0.05$	$P < 0.01$	$P < 0.001$

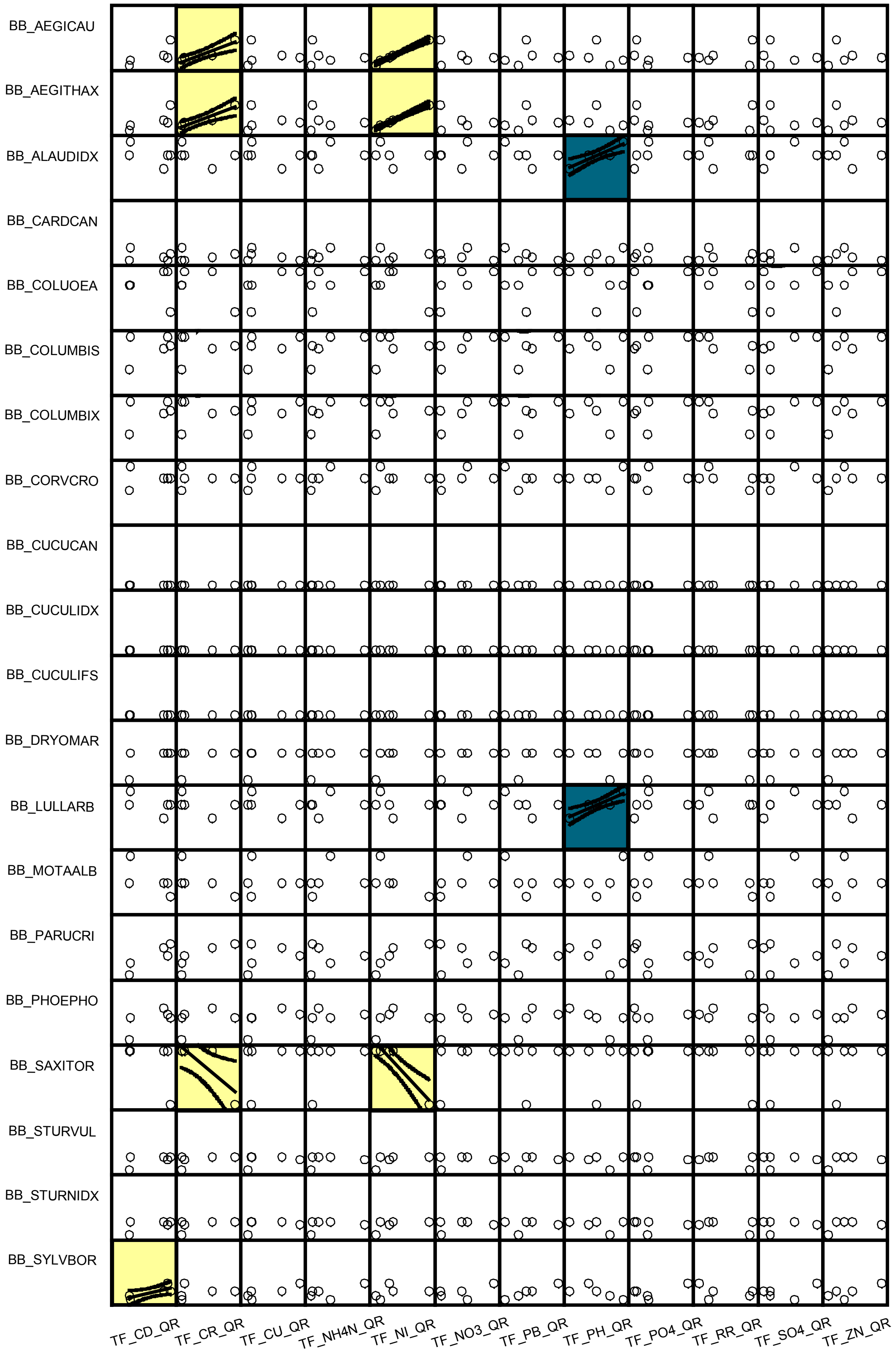


Figure 4: *Scatterplots between number of birds per species and chemical substances in throughfall of Quercus robur.*

Fitted lines:

P < 0.05	P< 0.01	P< 0.001

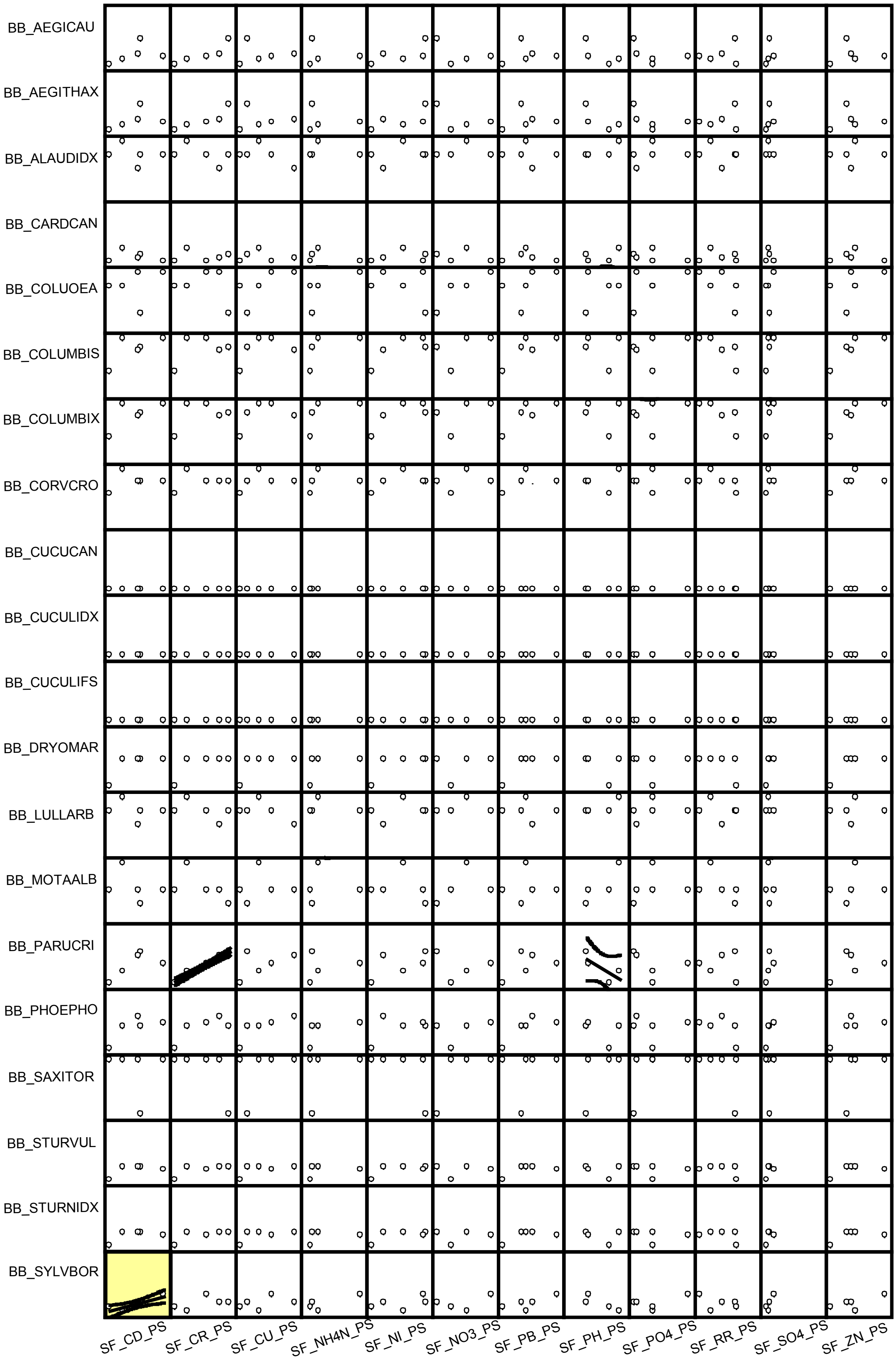


Figure 5: *Scatterplots between number of birds per species and chemical substances in stemflow water of Pinus sylvestris.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

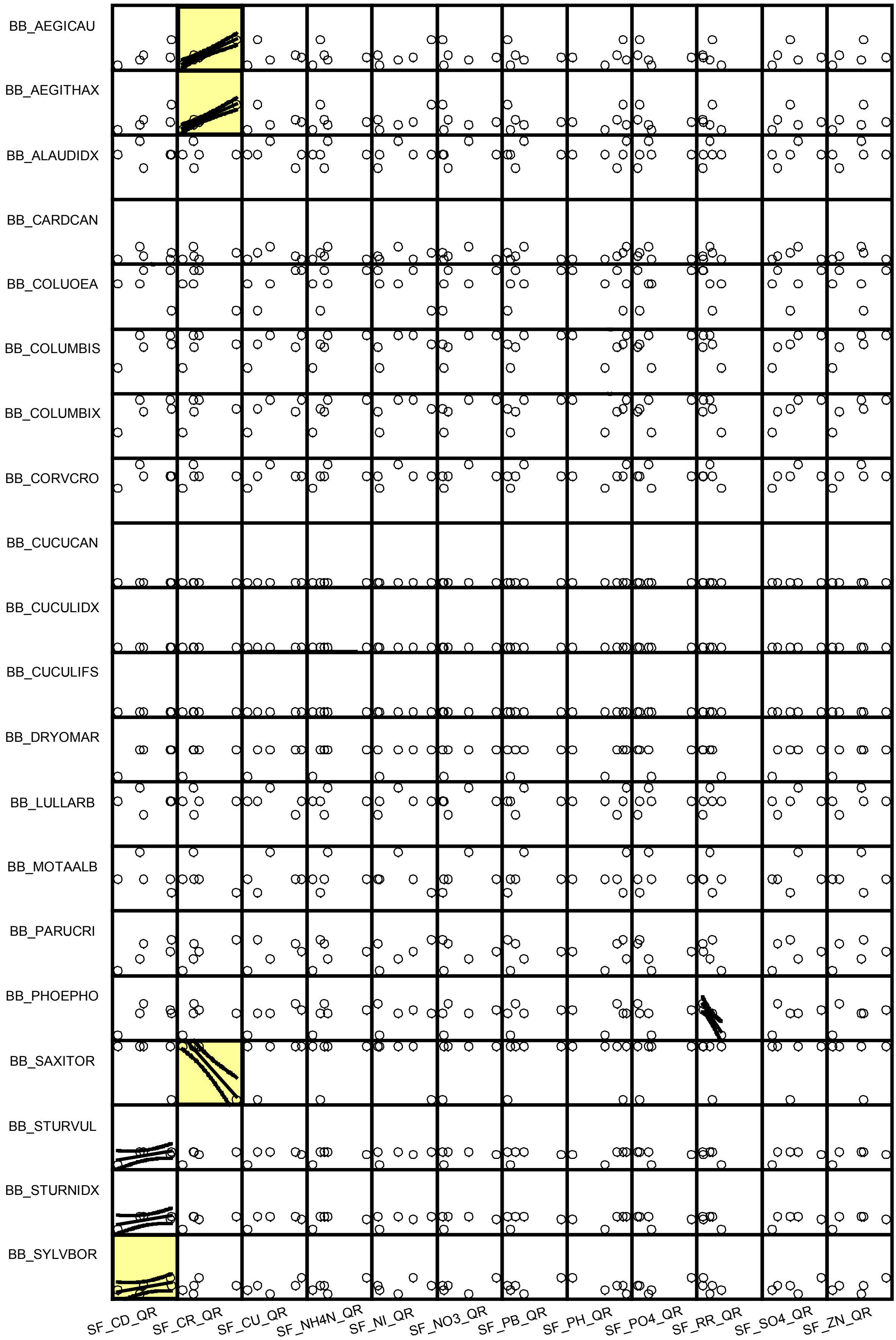


Figure 6: *Scatterplots between number of birds per species and chemical substances in stemflow water of Quercus robur.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

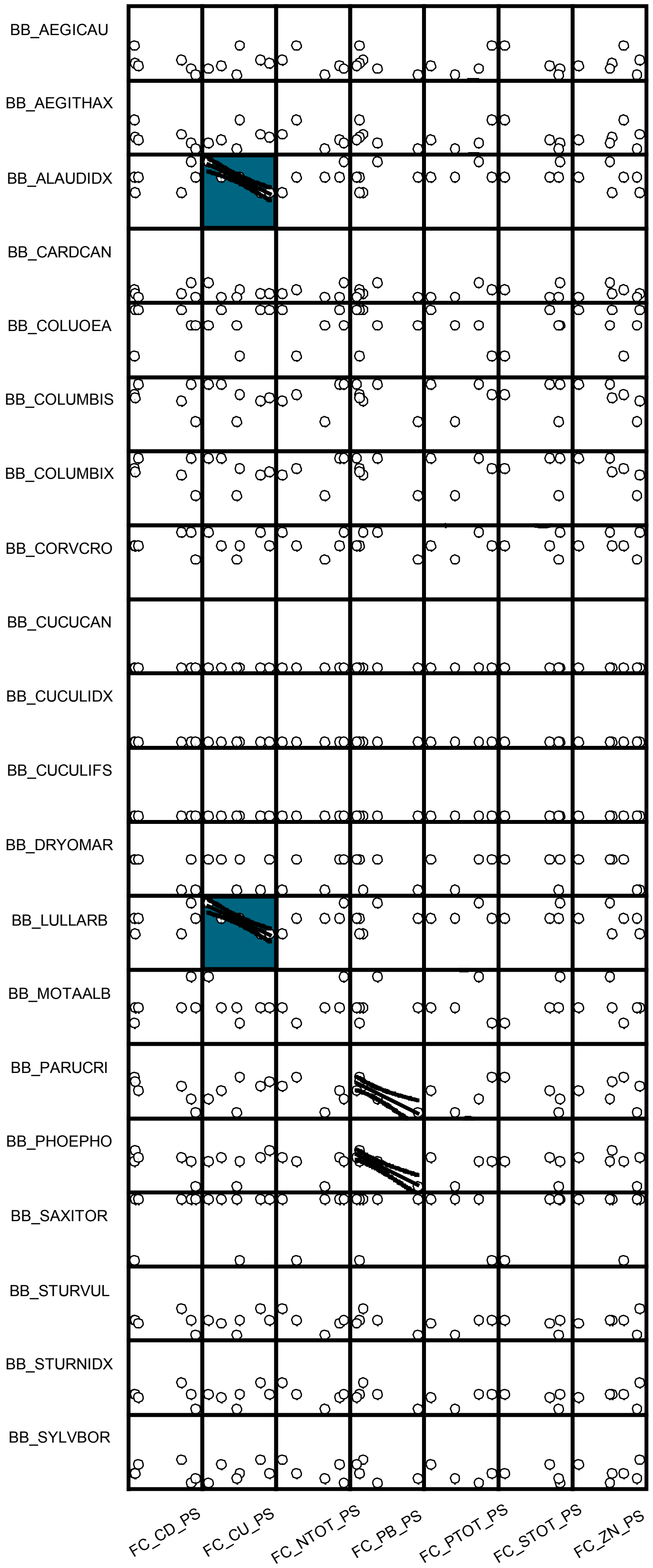


Figure 7: *Scatterplots between number of birds per species and chemical substances in living needles.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

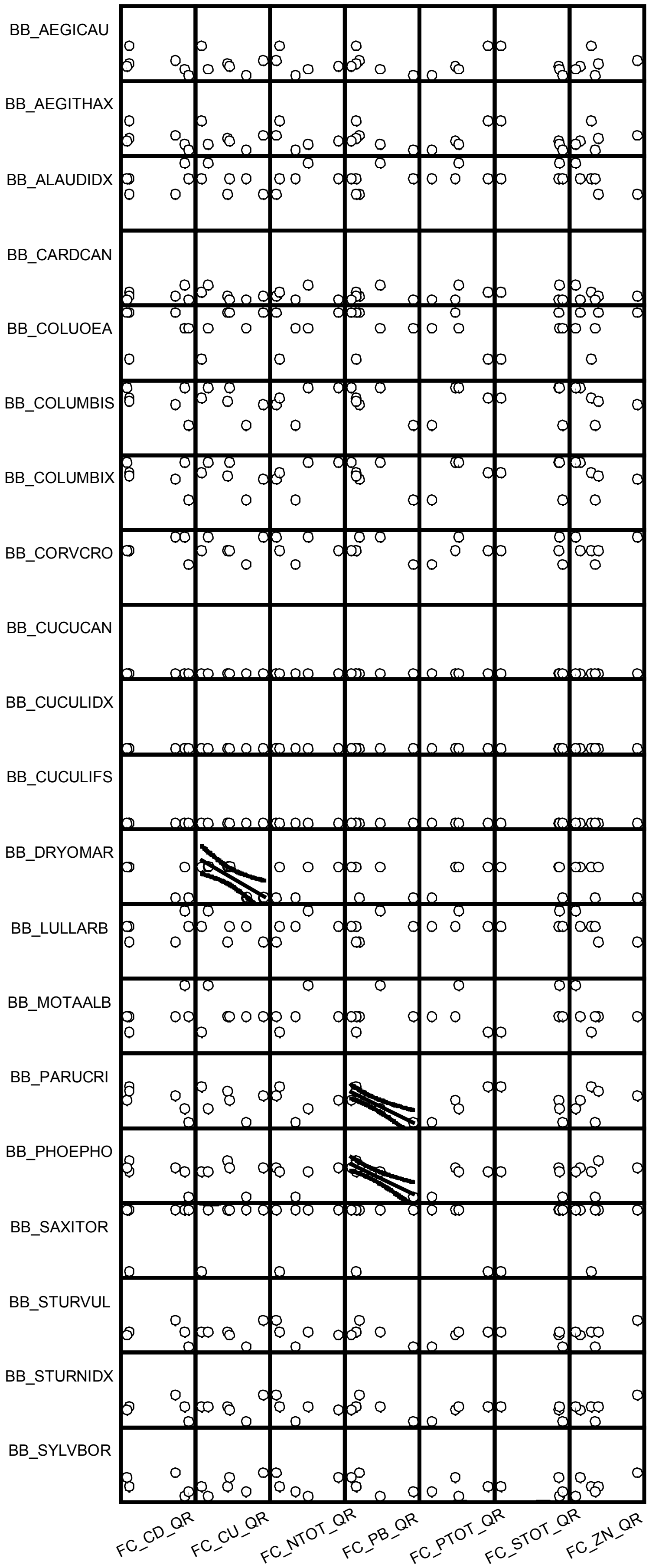


Figure 8 *Scatterplots between number of birds per species and chemical substances in living leaves.*

Fitted lines:

P < 0.05	P < 0.01	P < 0.001

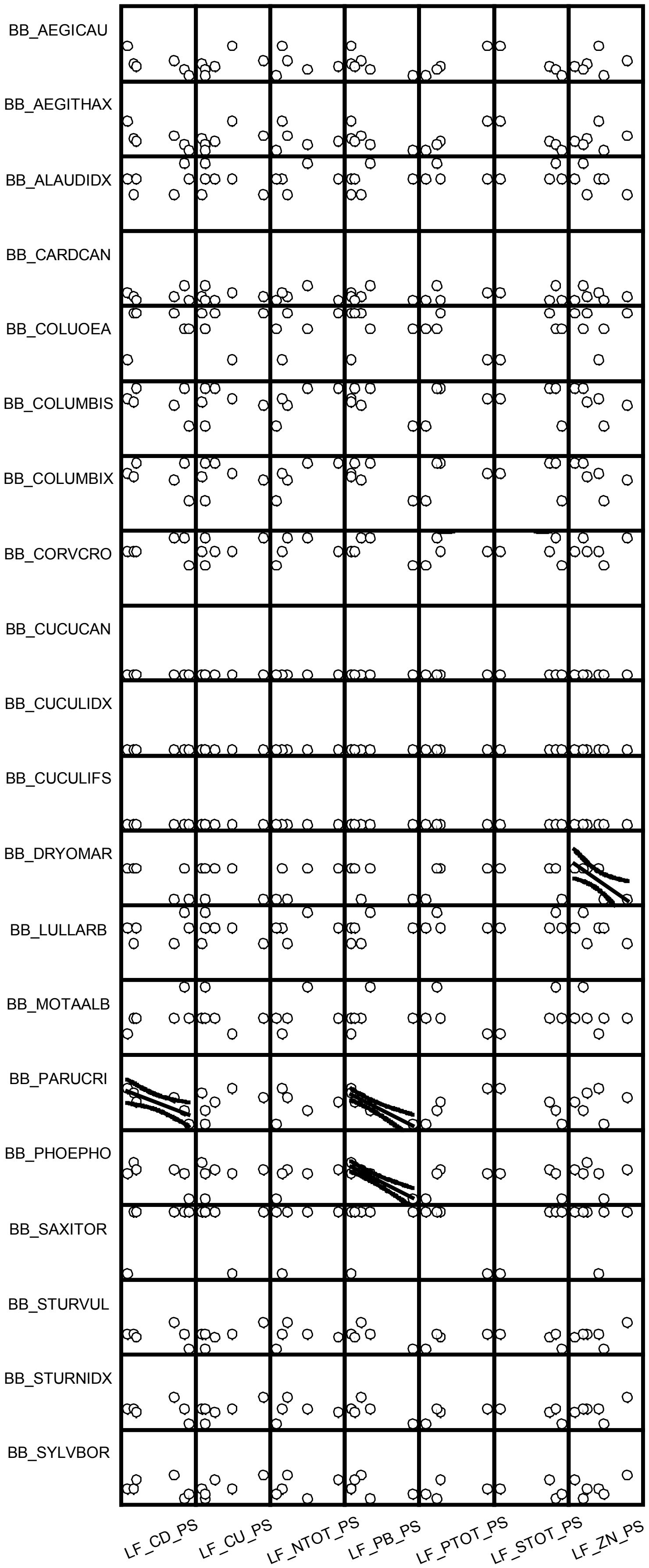


Figure 9: *Scatterplots between number of birds per species and chemical substances in dead needles.*

Fitted lines:

P < 0.05	P < 0.01	P< 0.001

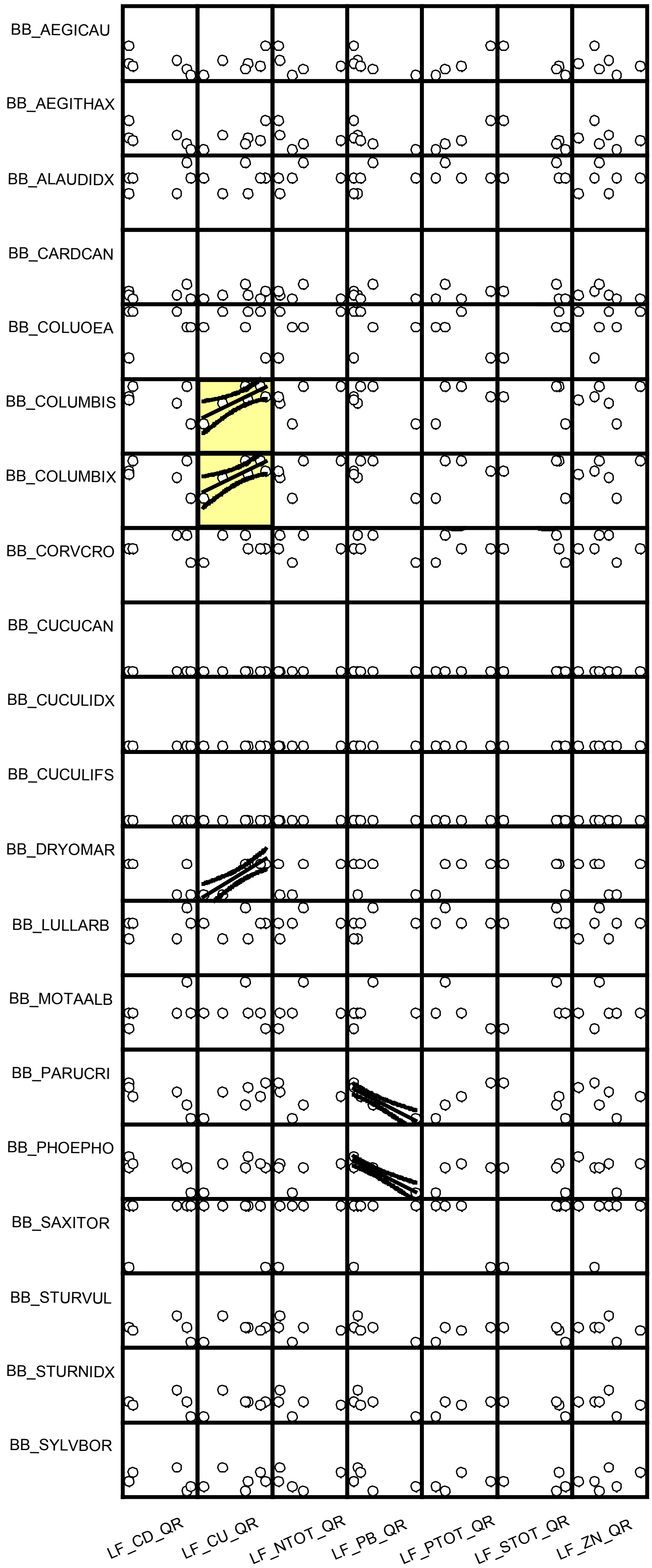


Figure 10: Scatterplots between number of birds per species and chemical substances in dead leaves.

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

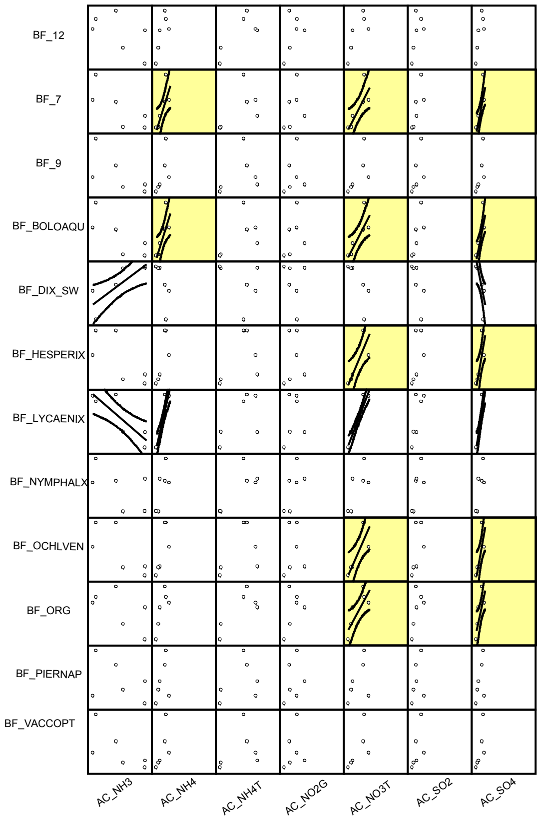


Figure 11: *Scatterplots between number of butterflies per species or biological group and chemical substances in air.*

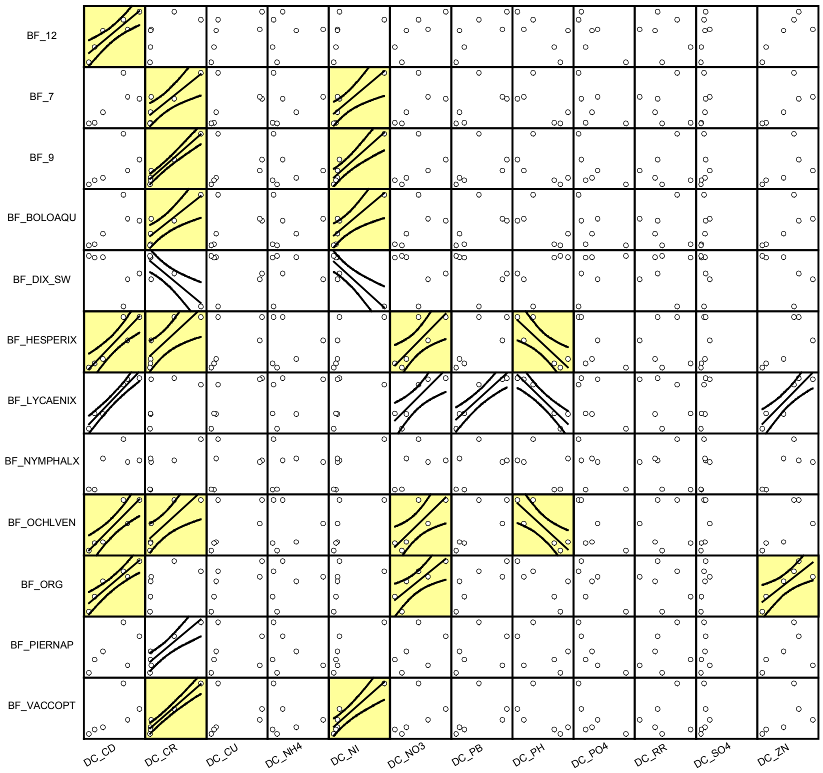


Figure 12: *Scatterplots between number of butterflies per species or biological group and chemical substances in deposition.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

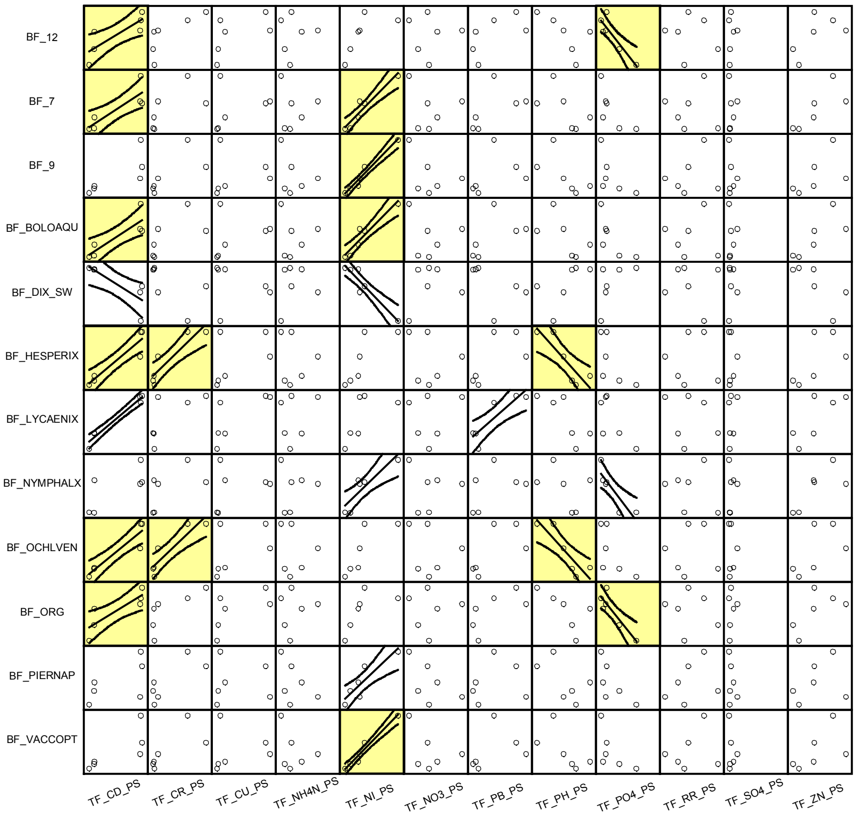


Figure 13: *Scatterplots between number of butterflies per species or biological group and chemical substances in throughfall water of Pinus sylvestris.*

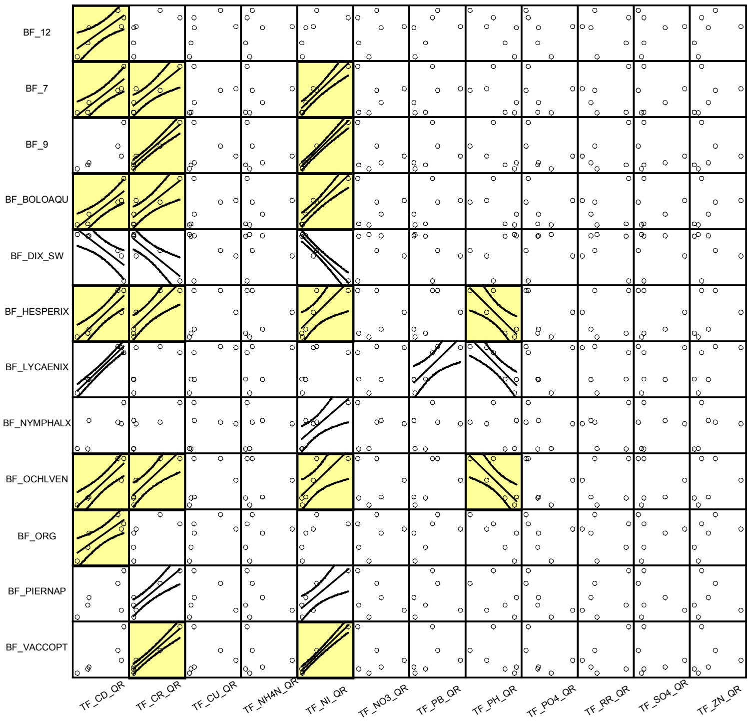


Figure 14: *Scatterplots between number of butterflies per species or biological group and chemical substances in throughfall water of Quercus robur.*

Fitted lines:

$P < 0.05$	$P < 0.01$	$P < 0.001$

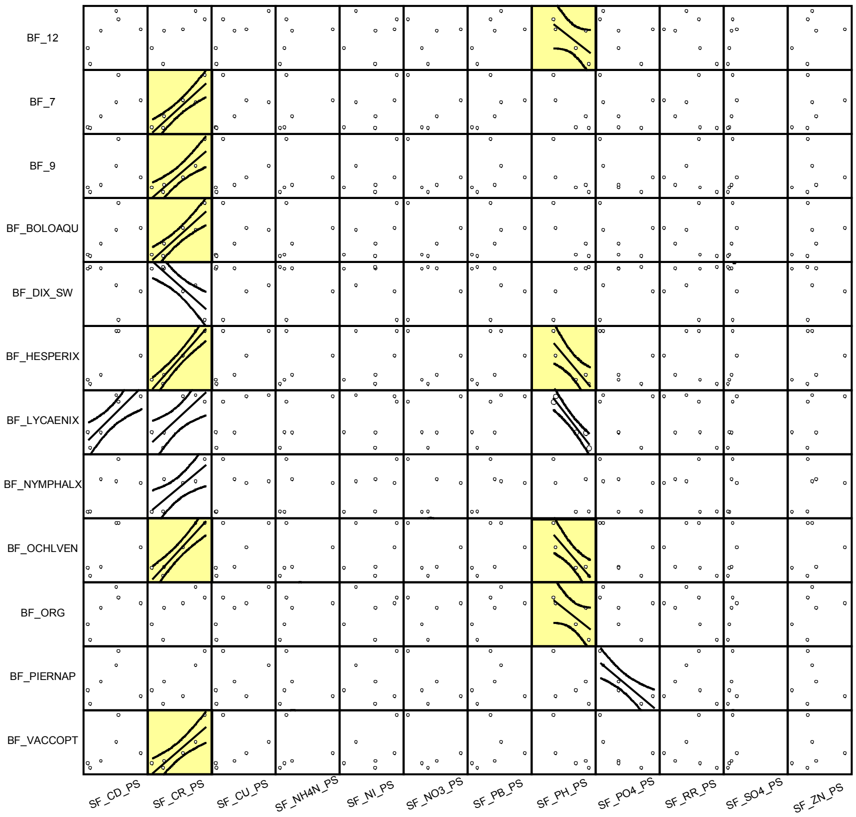


Figure 15: Scatterplots between number of butterflies per species or biological group and chemical substances in stemflow water of Pinus sylvestris.

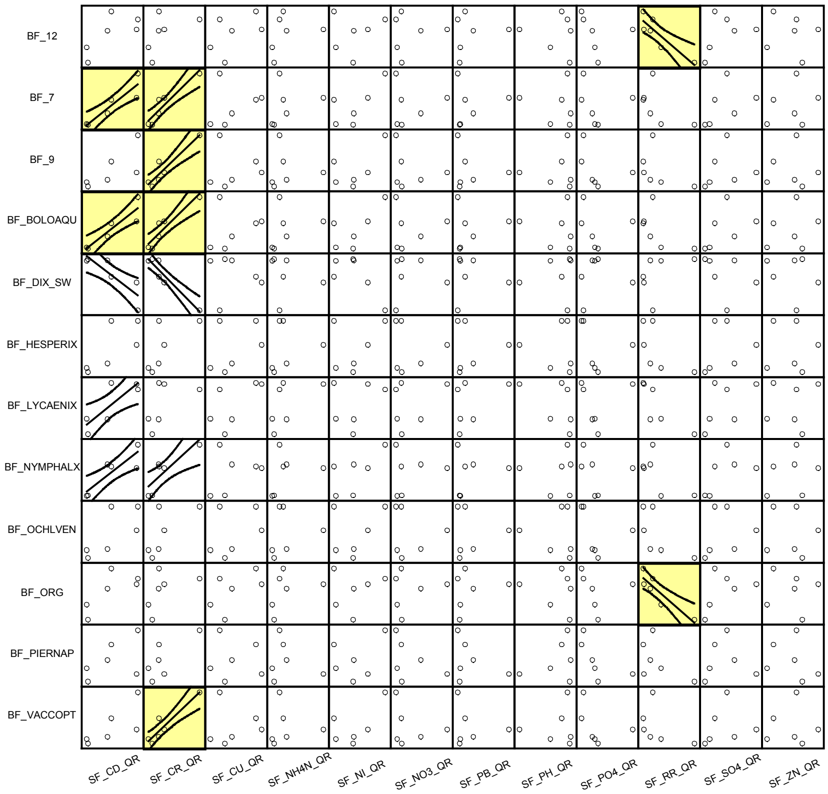


Figure 16: Scatterplots between number of butterflies per species or biological group and chemical substances in stemflow water of Quercus robur.

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

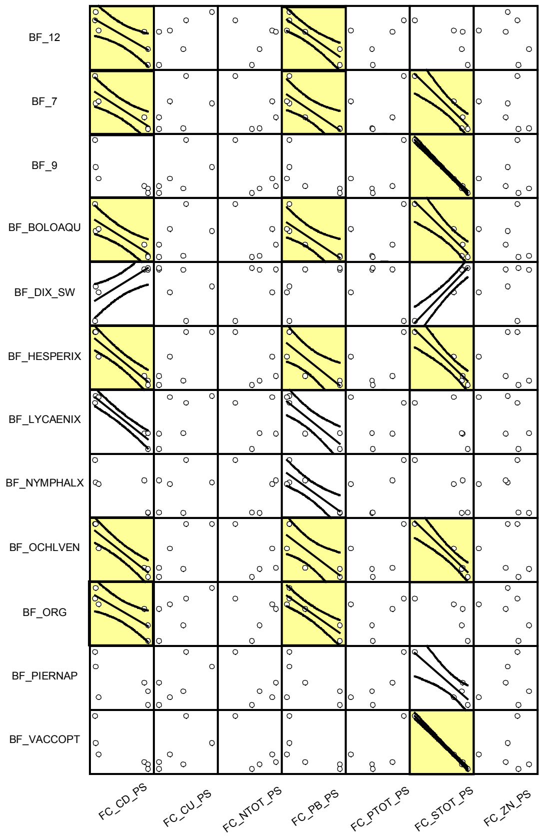


Figure 17: *Scatterplots between number of butterflies per species or biological group and chemical substances in living needles.*

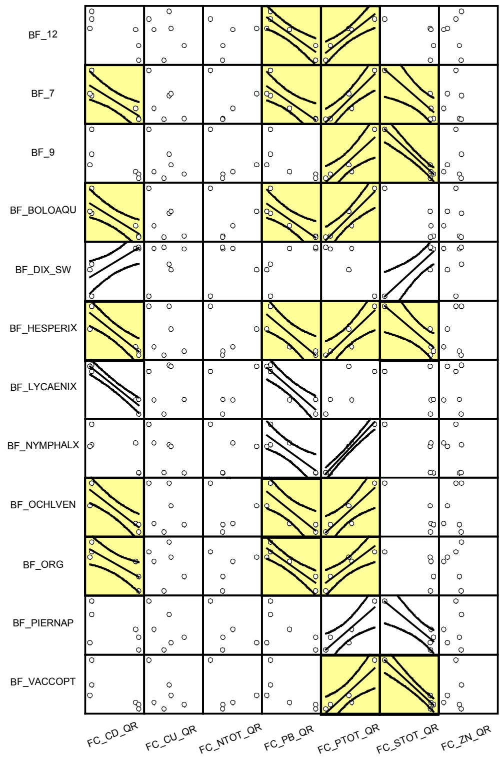


Figure 18: *Scatterplots between number of butterflies per species or biological group and chemical substances in living leaves.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

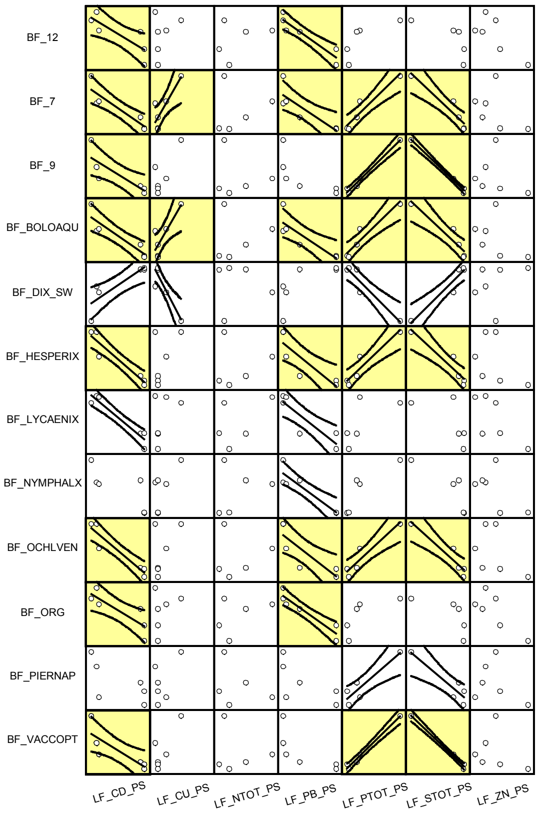


Figure 19: *Scatterplots between number of butterflies per species or biological group and chemical substances in dead needles.*

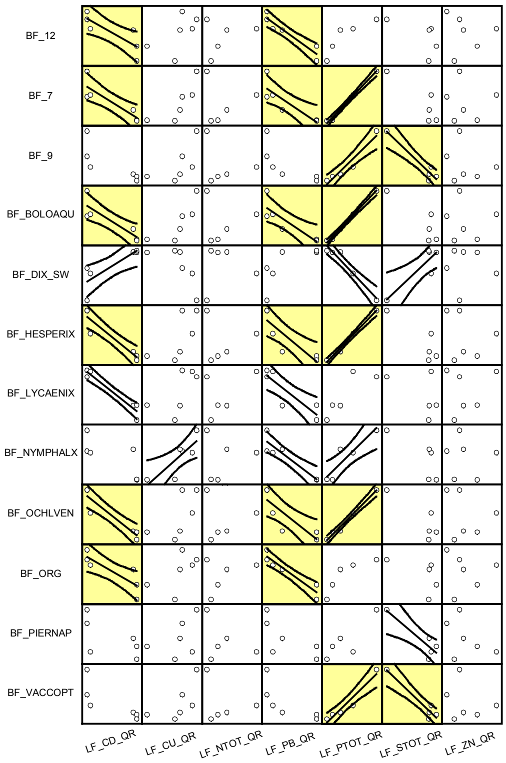


Figure 20: *Scatterplots between number of butterflies per species or biological group and chemical substances in dead leaves.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

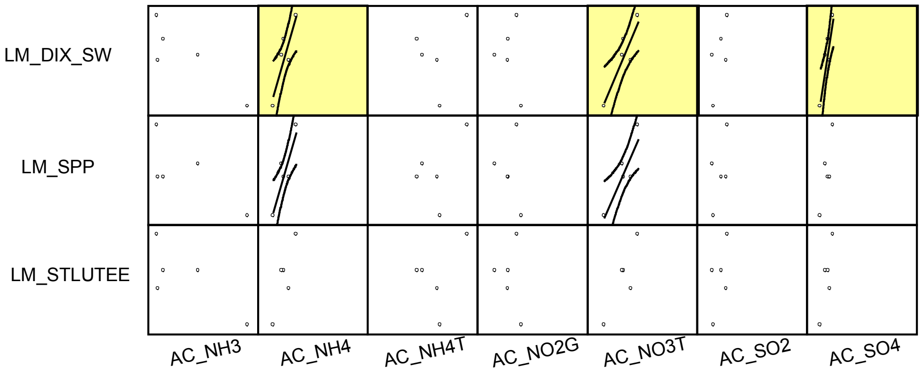


Figure 21: *Scatterplots between number of species of leafminers and chemical substances in air.*

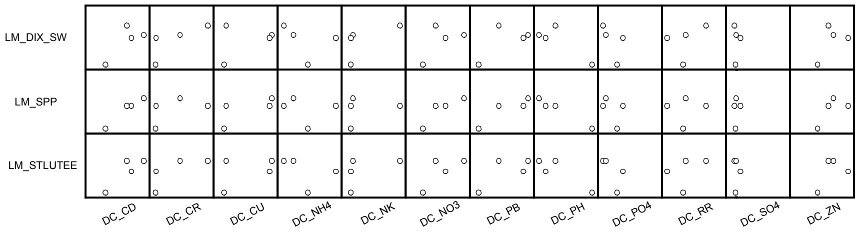


Figure 22: *Scatterplots between number of species of leafminers and chemical substances in deposition.*

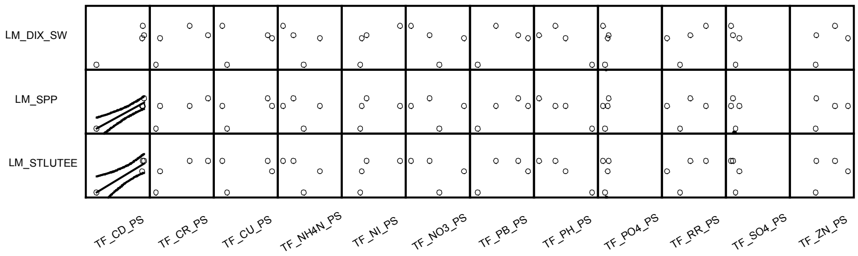


Figure 23: *Scatterplots between number of species of leafminers and chemical substances in throughfall water of Pinus sylvestris.*

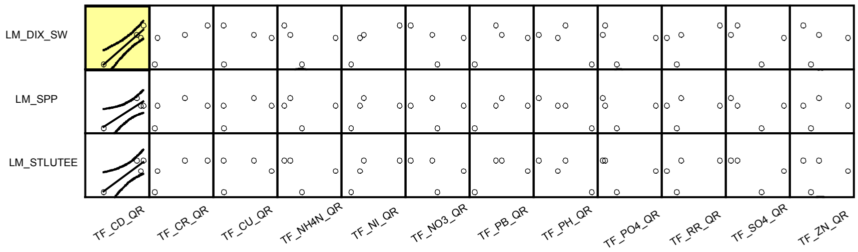


Figure 24: *Scatterplots between number of species of leafminers and chemical substances in throughfall water of Quercus robur.*

Fitted lines:

$P < 0.05$	$P < 0.01$	$P < 0.001$

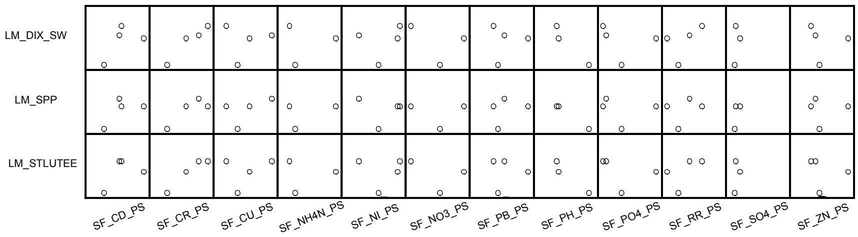


Figure 25: *Scatterplots between number of species of leafminers and chemical substances in stemflow water of Pinus sylvestris.*

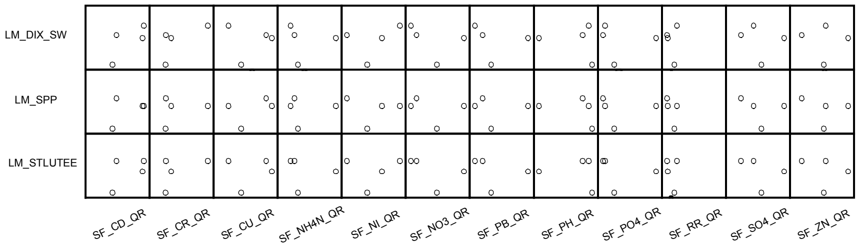


Figure 26: *Scatterplots between number of species of leafminers and chemical substances in stemflow water of Quercus robur.*

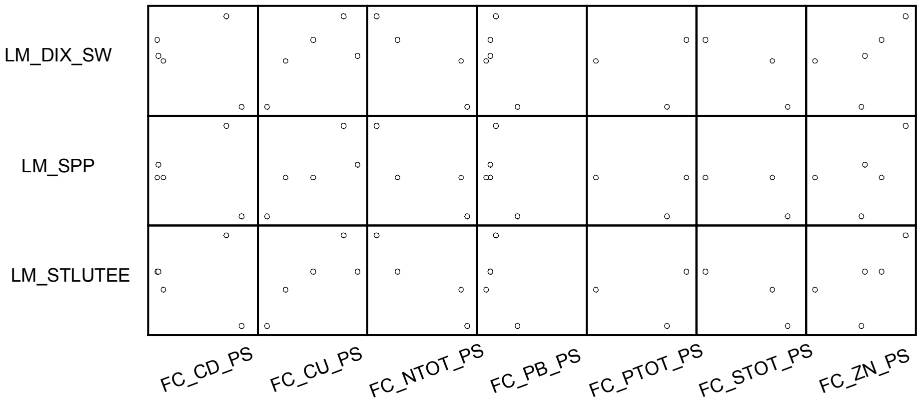


Figure 27: *Scatterplots between number of species of leafminers and chemical substances in living needles.*

Fitted lines:

$P < 0.05$	$P < 0.01$	$P < 0.001$

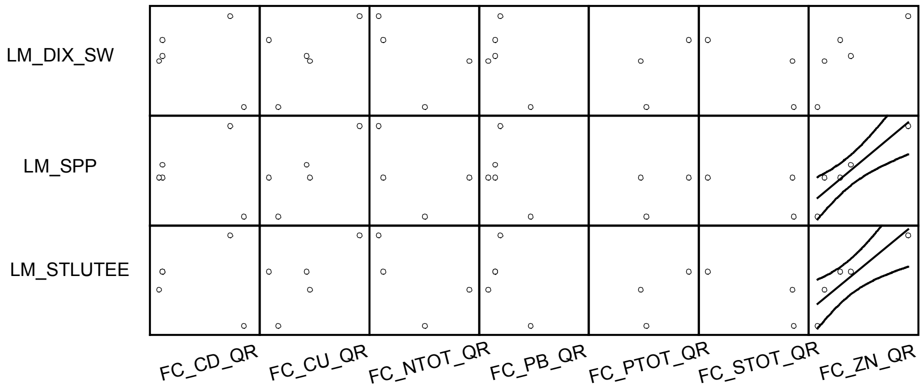


Figure 28: *Scatterplots between number of species of leafminers and chemical substances in living leaves.*

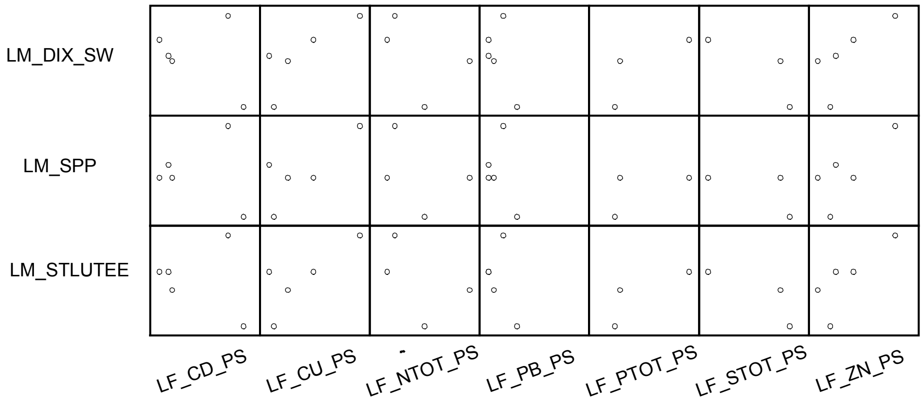


Figure 29: *Scatterplots between number of species of leafminers and chemical substances in dead needles.*

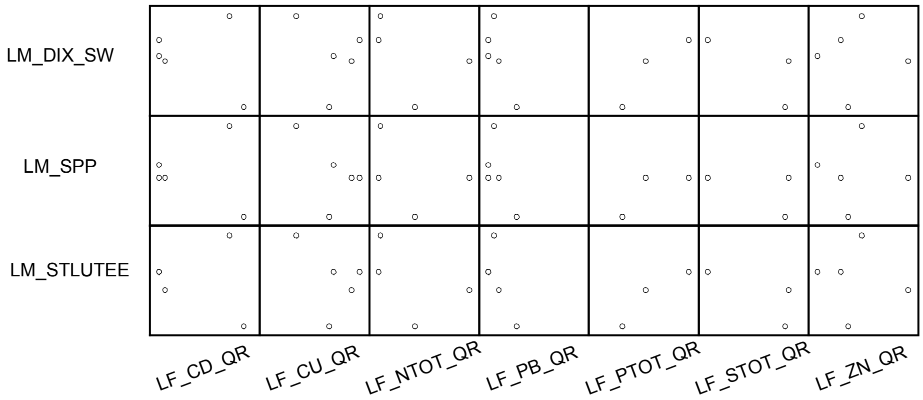


Figure 30: *Scatterplots between number of species of leafminers and chemical substances in dead leaves.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

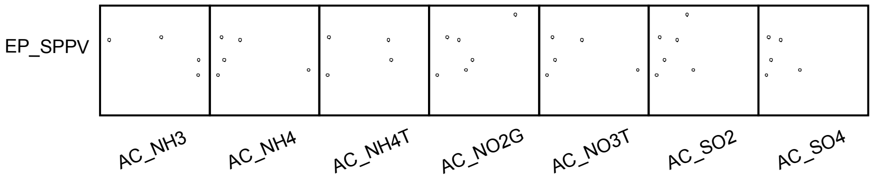


Figure 31: *Scatterplots between number of species of epiphytes and chemical substances in air.*

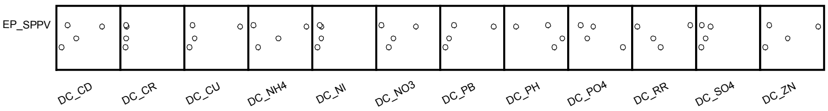


Figure 32: *Scatterplots between number of species of epiphytes and chemical substances in deposition.*

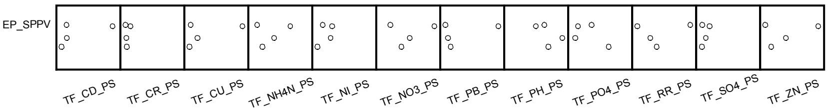


Figure 33: *Scatterplots between number of species of epiphytes and chemical substances in throughfall water of Pinus sylvestris.*

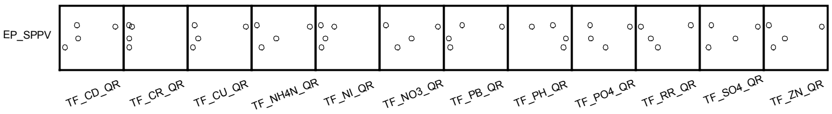


Figure 34: *Scatterplots between number of species of epiphytes and chemical substances in throughfall water of Quercus robur.*

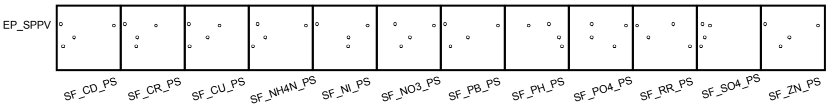


Figure 35: *Scatterplots between number of species of epiphytes and chemical substances in stemflow water of Pinus sylvestris.*

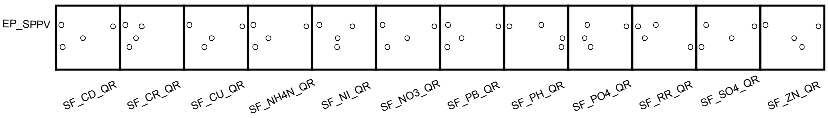


Figure 36: *Scatterplots between number of species of epiphytes and chemical substances in stemflow water.*

Fitted lines:

$P < 0.05$	$P < 0.01$	$P < 0.001$

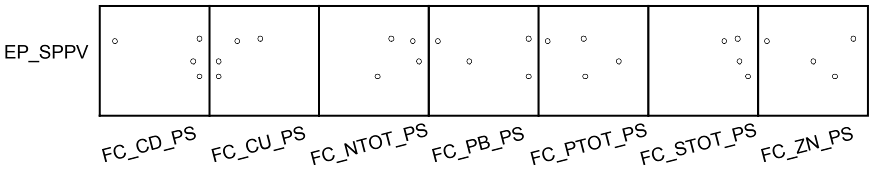


Figure 37: *Scatterplots between number of species of epiphytes and chemical substances in living needles.*

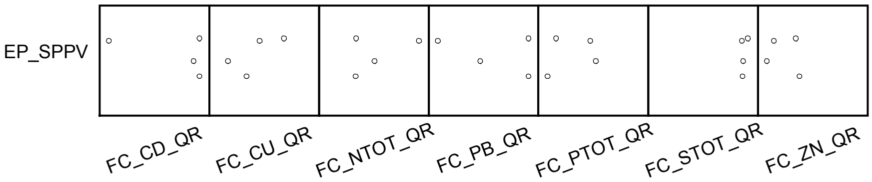


Figure 38: *Scatterplots between number of species of epiphytes and chemical substances in living leaves.*

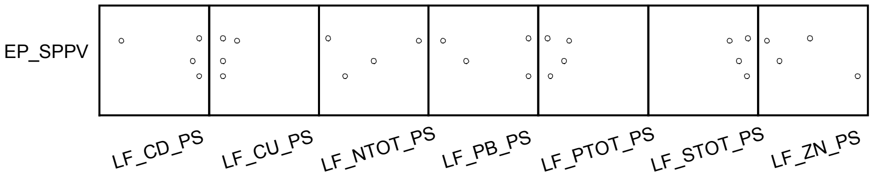


Figure 39: *Scatterplots between number of species of epiphytes and chemical substances dead needles.*

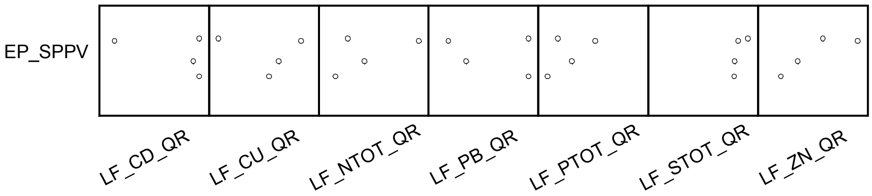


Figure 40: *Scatterplots between number of species of epiphytes and chemical substances in dead leaves.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

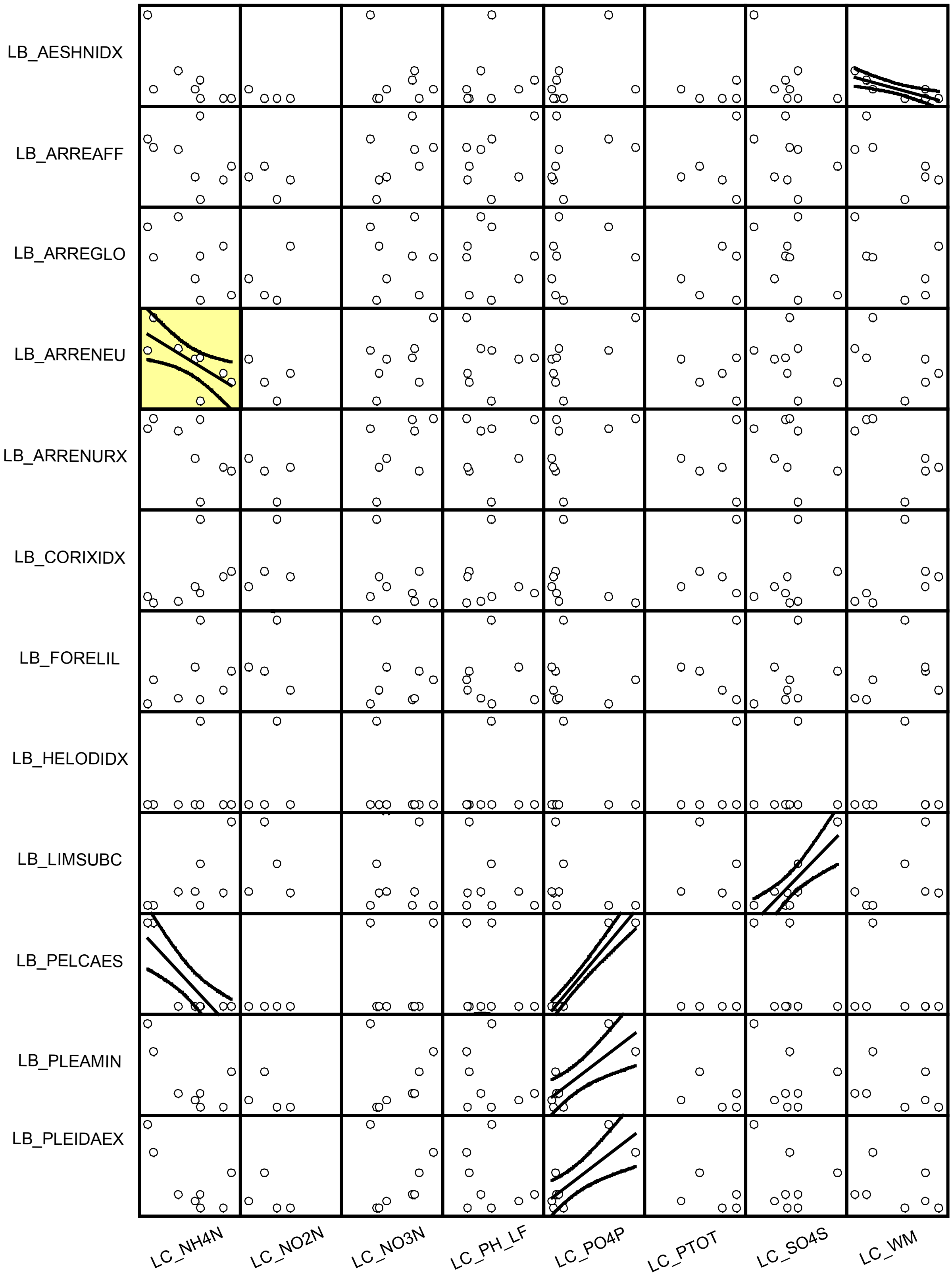


Figure 41: *Scatterplots between number of organisms per species of the macrofauna and chemical substances in lakewater.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

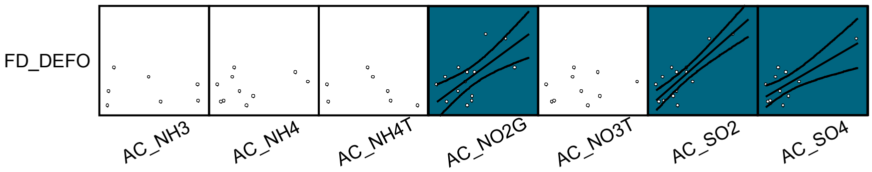


Figure 42: *Scatterplots between defoliation of Pinus sylvestris and chemical substances in air.*

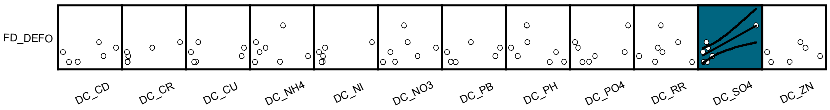


Figure 43: *Scatterplots between defoliation of Pinus sylvestris and chemical substances in deposition.*

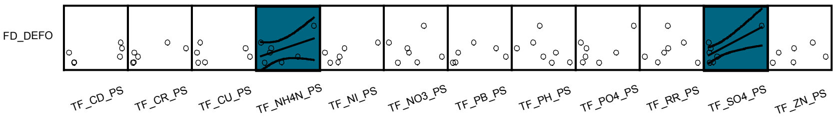


Figure 44: *Scatterplots between defoliation of Pinus Sylvestris and chemical substances in throughfall water of Pinus sylvestris*

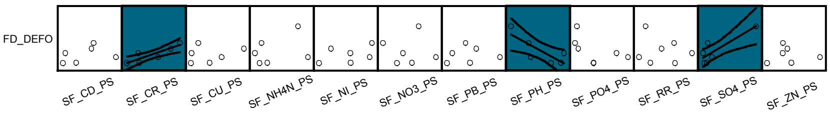


Figure 45: *Scatterplots between defoliation for trees and chemical substances in stemflow water of Pinus sylvestris.*

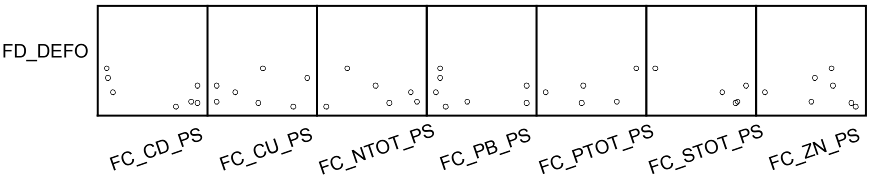


Figure 46: *Scatterplots between defoliation for trees and chemical substances living needles.*

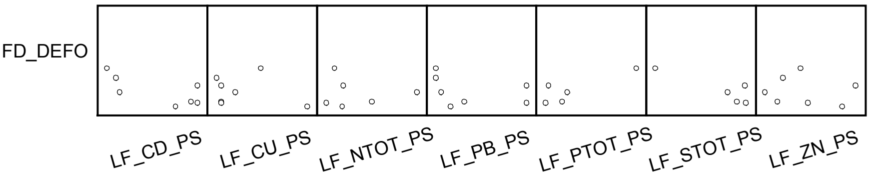


Figure 47: *Scatterplots between defoliation for trees and chemical substances in dead needles.*

Fitted lines:

P< 0.05	P< 0.01	P< 0.001

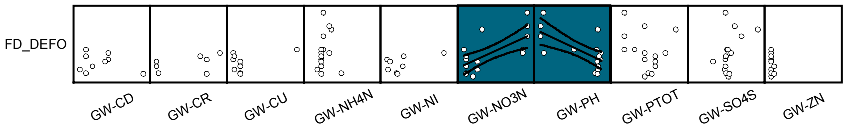


Figure 48 *Scatterplots between defoliation for trees and chemical substances in groundwater (1200 cm).*

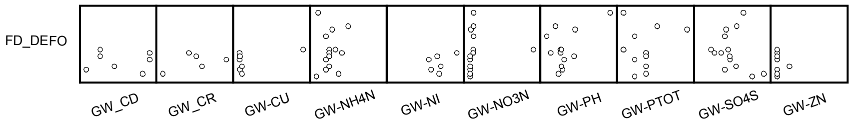


Figure 49: *Scatterplots between defoliation for trees and chemical substances in groundwater (3000 cm).*

Fitted lines:

$P < 0.05$	$P < 0.01$	$P < 0.001$

APPENDIX VI LIST OF ABBREVIATIONS

ALTERRA	Green World Research
CLRTAP	Convention on Long-Range Transboundary Air Pollution
DGM	Directoraat Generaal Milieu
ECN	Energieonderzoek Centrum Nederland
ICP-IM	International Co-operative Programme on Integrated Monitoring (on Air Pollution Effects on Ecosystems)
KNMI	Het Koninklijk Nederlands Meteorologisch Instituut
KvI	Klimaatverandering en Industrie
LON	Lichenologisch Onderzoekbureau Nederland

MONITORING PROGRAMME CODES

AC	Air Chemistry
AM	Climate
BB	Inventory of Birds
BF	Inventory of Butterflies
BV	Inventory of Plants
DC	Precipitation Chemistry
EP	Trunk Epiphytes
FC	Foliage Chemistry
FD	Forest Damage
GW	Groundwater Chemistry
LB	Hydrobiology of Lakes
LC	Lake water Chemistry
LF	Litterfall Chemistry
LM	Inventory of Leafminers
MC	Metal Chemistry of Mosses
SC	Soil Chemistry
SF	Stemflow Chemistry
SW	Soil water Chemistry
TF	Throughfall Chemistry
VG	Vegetation

NFP	National Focal Point

PARAMETER CODES

Biological codes

AEGICAU	*Aegithalos caudatus*
AEGITHAX	*Aegithaidae*
AESHNIDX	*Aeshnidae*
ALAUDIDX	*Alaudidae*
ARREAFF	*Arrenurus affinis*
ARREGLO	*Arrenurus globator*
ARRENEU	*Arrenurus neumani*
ARRENURX	*Arrenuridae*

BF12	Species that hibernate as a juvenile caterpillar and have a long maturation (section group 4)
BF7	Species that hibernate as a small caterpillar (section group 4)
BF9	Species that hibernate as a juvenile caterpillar and have a short maturation (section group 4)
BOLOAQU	*Boloria aquilonaris*
CARDCAN	*Carduelis cannabina*
COLUMBIS	*Columbiformes*
COLUMBIX	*Columbidae*
COLUOEA	*Columba oenas*
CORIXIDX	*Corixidae*
CORVCRO	*Corvus corone*
CUCUCAN	*Cuculus canorus*
CUCULIDX	*Cuculidae*
CUCULIFS	*Cuculiformes*
DAM	Damaged trees
DBH	Diameter
DEFO	Defoliation
DISC	Discoloration
DIX_SW	Shannon-Wiener diversity index
DIX_SW.Bp	Shannon-Wiener diversity index of *Betula pendula*
DRYOMAR	*Dryocopus martius*
FORELIL	*Forelia liliacea*
HCROW	Crown height
HEIG	Height of trees
HESPERIX	*Hesperiidae*
LIMSUBC	*Limnephilus subcentralis*
LULLARB	*Lullula arborea*
LYCAENIX	*Lycaenidae*
MOTAALB	*Motacilla alba*
NRORG	Total number of organisms
NUM	Number of trees
NYMPHALX	*Nymphalidae*
OCHLVEN	*Ochlodes venata*
PARUCRI	*Parus cristatus*
PELCAES	*Peltodytes caesus*
PHOEPHO	*Phoenicurus phoenicurus*
PIERNAP	*Pieris napi*
PLEAMIN	*Plea minutissima*
PLEIDAEX	*Pleidae*
PS	*Pinus sylvestris*
Qr	*Quercus robur*
SAXITOR	*Saxicola torquata*

SECS	Secondary shoots
SPP.Bp	Number of species on *Betula pendula*
SPPV	Number of species
ST LUTEE.Bp	*Stigmella luteella* on *Betula pendula*
STURNIDX	*Sturnidae*
STURVUL	*Sturnus vulgaris*
SYLVBOR	*Sylvia borin*
VACCOPT	*Vacciniina optilete*
WCROW	Crown width

Chemical and physical codes

Al	Aluminium
ALK	Alkalinity
ANF	Annual needle fascicles
As	Arsenic
Ba	Barium
Ca	Calcium
Cd	Cadmium
CEC_E	Cation exchange capacity effective
Cl	Chloride
CNR.N320	Colour number 320 nm
CNR.N455	Colour number 455 nm
Co	Cobalt
COR	Organic carbon
CORT	Total organic carbon
COVE	Coverage
CP	Chlorophyll-A
CR	Chromium
CTY.25	Specific conductivity at 25 °C
Cu	Copper
F	Fluor
Fe	Iron
Hg	Mercury
HH	Humidity
K	Potassium
Ldep	Litterfall amount
Mg	Magnesium
Mn	Manganese
Mo	Molybdenium
Na	Sodium
NH_3	Ammonium
NH_4	Ammonium
NH_{4tot}	Nitrogen ammonium total ($NH_{3\text{ (gaseous)}}$ + $NH_{4\text{ (particulate)}}$)
Ni	Nickel

NO_2	Nitrite
NO_3	Nitrate
N_{tot}	Total nitrogen
O_2	Oxygen
O_3	Ozone
Pb	Lead
pH	pH
PO_4	Phosphate
P_{tot}	Total phosphorous
RET	Residue total (dry weight) 100 needles/leaves
RR	Precipitation
Sb	Antimony
SiO_2	Silica
SO_2	Sulphur
SO_4	Sulphate
SOL_G	Global radiation
SOL_U	Insolation, UV-radiation
Sr	Strontium
S_{tot}	Total sulphur
T	Temperature
Ti	Titanium
V	Vanadium
WL	Water level
WM (NAP)	Watermark
Zn	Zinc

WUR	Wageningen Universiteit en Research Centrum
RIVM	Rijksinstituut voor Volksgezondheid en Milieu (National Institute for Public Health and the Environment)
-ECO	Laboratorium voor Ecotoxicologie (Laboratory for Ecotoxicology)
-LBG	Laboratorium voor Bodem- en Grondwateronderzoek
-LLO	Laboratorium Luchtonderzoek (Laboratory of Air Research)
RIN	Research Institute for Nature Management
SBB	Staats Bosbeheer (State Forestry Service)
SOVON	Samenwerkende Organisaties Vogelonderzoek Nederland (Cooperative Organizations for Bird Research in The Netherlands)
Tinea	Stichting faunistisch onderzoek Microlepidoptera Tinea (Society Faunistic Research Microlepidoptera Tinea)
UN-ECE	United Nations Economic Commitee for Europe