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Atmospheric nitrogen dynamics in Hesse, Germany: Creating the data base : 1. Bulk deposition of acidifying and eutrophying species

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Atmospheric nitrogen dynamics in Hesse, Germany: Creating the data base

1. Bulk deposition of acidifying and eutrophying species

Ulrich Dämmgen¹

Abstract

Between July 2001 and December 2004, a comparatively dense sampling network was operated in Hesse, Germany, to obtain bulk deposition data. The major goal was to collect data to enable closure of the atmospheric nitrogen balance above Hesse. In addition to ammonium- and nitrate-nitrogen, the amount of precipitation and the bulk deposition of sulfate, chloride, sodium, potassium, magnesium and calcium were measured.

The three replicates per sampling site and the short distances between the sites permitted a detailed plausibility control. Although a considerable number of samples had to be rejected, in particular due to fouling by birds, an almost complete dataset could be obtained. Both for ammonium and for nitrate, the mean error of determination was $0.5 \text{ kg ha}^{-1} \text{ a}^{-1} \text{ N}$.

For the entire area of Hesse, nitrate nitrogen concentrations were quite constant. Thus, the respective deposition was closely related to the amount of precipitation. Ammonium concentrations and depositions varied considerably in space and were strongly influenced by local sources. Ammonium was usually co-deposited with nitrate and sulfate. The soil borne elements potassium, magnesium and calcium were often co-deposited as were sodium and chloride. Any relations found between concentrations or depositions in the whole area were most distinctive in the Vogelberg catena.

Key words: nitrogen balance, deposition

Zusammenfassung

Atmosphärische Dynamik von Stickstoff in Hessen: Erstellung der Datenbasis

1. Bulk-Deposition von versauernden und eutrophierenden Spezies

In Hessen wurde von Juli 2001 bis Dezember 2004 ein vergleichsweise dichtes Messnetz zur Bestimmung der Bulk-Deposition betrieben. Hauptziel der Messungen war es, Datensätze zur Schließung der atmosphärischen Stickstoff-Bilanz über Hessen bereit zu stellen. Neben Ammonium- und Nitrat-Stickstoff wurden Niederschlagsmengen sowie Sulfat-, Chlorid-, Natrium-, Kalium-, Magnesium- und Calcium-Bulk-Depositionen bestimmt.

Die drei Parallelen pro Standort und das dichte Messnetz erlaubten eine intensive Plausibilitätskontrolle. Obwohl zahlreiche Einzelproben wegen der Verunreinigung insbesondere durch Vögel verworfen werden mussten, ergab sich ein nahezu vollständiger Datensatz. Sowohl für Nitrat- als für Ammonium Stickstoff werden die jährlichen Depositionen mit einem Fehler von $0,5 \text{ kg ha}^{-1} \text{ a}^{-1} \text{ N}$ bestimmt.

Während die Nitrat-Stickstoff-Konzentrationen über das ganze Land relativ konstant waren und so zu Depositionen führten, die stark von der Niederschlagsmenge abhingen, waren die örtlichen Ammonium-Stickstoff-Konzentrationen und –Depositionen stark von regionalen Quellen geprägt. Ammonium wurde gemeinsam mit Nitrat und Sulfat deponiert. Die bodenbürtigen Elemente Kalium, Magnesium und Calcium wurden oft gemeinsam deponiert, ebenso Natrium und Chlorid. Die gefundenen Zusammenhänge waren vor allem in der Catena über den Vogelsberg ausgeprägt.

Schlüsselworte: Stickstoff-Bilanz, Deposition

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1 The Hessian initiative to balance atmospheric fluxes of reactive nitrogen

Despite the fact that increased attention has been paid to the problem of eutrophication, few attempts have been made to establish regional or national approaches to quantify emissions, atmospheric transport and deposition in such a way that the atmospheric nitrogen balance can be established. Such a balance is crucial in the patchwork of models and measurements needed to describe cause-effect relationships and to induce political and administrative measures to reduce air pollution in general and pollution with reactive nitrogen species in particular (Dämmgen et al., 2006; Gauger et al., 2006). In 2001, the Hessian Agency for the Environment and Geology (HLUG) in cooperation with the Institute of Agroecology of the Federal Agricultural Research Centre (FAL) and the Institute for Plant Ecology of the Justus-Liebig University, Giessen, started an initiative to establish a research programme to quantify emissions, atmospheric concentrations and chemistry as well as deposition fluxes using a matching set of measurements and models, aiming at the closure of the atmospheric nitrogen balance. All reactive species were to be covered: gaseous ammonia (NH_3 , emission and deposition), nitrous oxide (N_2O , emission), nitric oxide (NO , emission), nitrogen dioxide (NO_2 , deposition), nitrous and nitric acids (HNO_2 and HNO_3 , deposition), as well as ammonium (NH_4^+) and nitrate (NO_3^-) deposition in sedimenting and non-sedimenting particles. Due to the restricted availability of agricultural census data, the year

of the detailed census, 2003, was selected the period for which a comprehensive data set was to be generated. Fig. 1 illustrates the related sequels of activities needed to establish the data set and the subsequent modelling activities.

This paper is the first in a series and describes the measurement of the deposition of sedimenting particles (bulk deposition).

2 Sampling network and periods, properties of Rotenkamp samplers

In 2001, a network consisting of 50 sampling sites covering the German federal state of Hesse was established. Six catenae were built with a northwest southeast direction (sites D3 to D9, D10 to D14, D17 to D22, D27 to D33, D38 to D43, D44 to D49) and supplemented with a second catena stretching southeast to northwest across the Vogelsberg (D13 to D26). The Rhön region was of particular interest due to its varying elevations (D32 to D37). Single sites along the borders of Hesse (D1, D2, D50) complete the pattern (Fig. 2, Table 1). The mean distance between sites in one catena is 20 km. The distance between the catenae is approx. 40 km. Site D19, Linden, is the major climate observation site of the Institute for Plant Ecology of the University of Giessen. It has been used in joint activities between this institute and the FAL Institute of Agroecology since 1996, including the determination of bulk deposition.

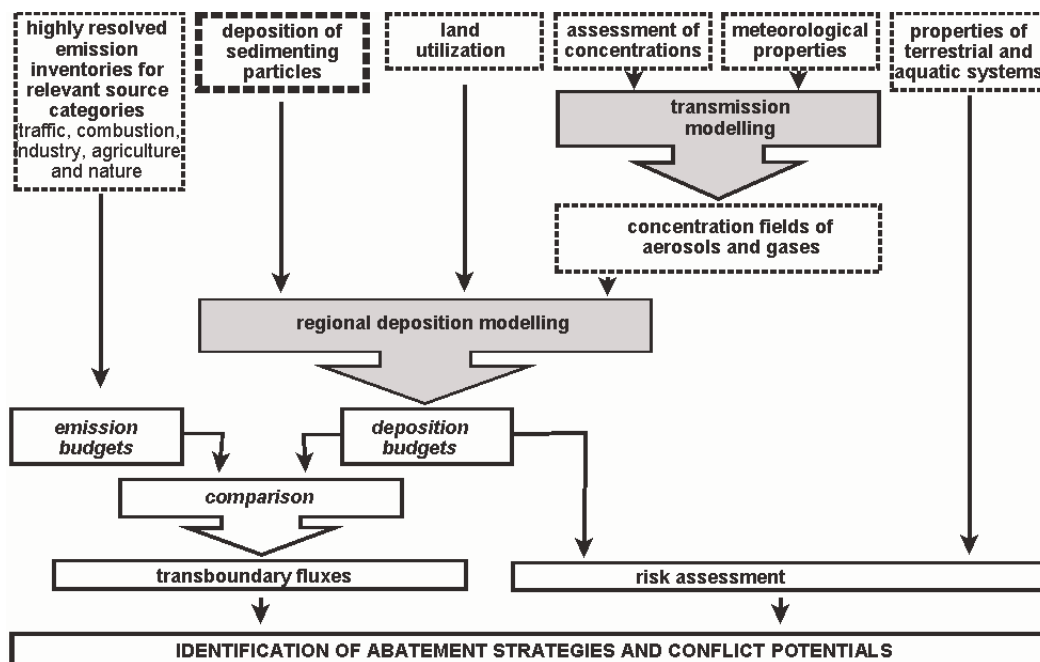


Fig. 1 Data fluxes in the Hessian atmospheric nitrogen dynamics project (Grünhage et al., 2002, modified). Rectangles in the uppermost line illustrate the data sets needed.



Map 1
Locations of the sampling stations

Table 1
Sampling stations for bulk deposition

No	Name	Geographic co-ordinates Gauss-Krüger co-ordinates ("Rechts wert" easting, "Hochwert", northing)		Located at (OT: Ortsteil)	Elevation above sea level	Correction class according to Richter (1995)
D01	Viernheim	8° 34' 48.0" E	49° 32' 49.7" N		98 m	VI c
D02	Neckarsteinach	8° 51' 21.2" E	49° 23' 56.2" N		115 m	VI c
D03	Lorch	7° 50' 34.9" E	50° 06' 22.9" N	OT Wollmerschied	443 m	IV b
D04	Eltville	8° 06' 59.4" E	50° 03' 13.1" N	OT Martinsthal	248 m	IV d
D05	Wiesbaden	8° 14' 38.6" E	50° 02' 08.9" N		88 m	VI d
D06	Groß-Gerau	8° 29' 40.1" E	49° 54' 49.9" N		89 m	VI d
D07	Mühltal	8° 42' 12.7" E	49° 48' 13.1" N	OT Waschenbach	225 m	VI c
D08	Brombachtal	8° 55' 10.6" E	49° 43' 59.2" N	OT Hembach	395 m	VI b
D09	Erbach	9° 03' 25.5" E	49° 39' 54.0" N	OT Ernsbach	364 m	VI d
D10	Limburg	8° 05' 20.7" E	50° 26' 31.8" N	OT Ahlbach	245 m	IV b
D11	Selters	8° 19' 07.5" E	50° 21' 35.9" N	OT Haintchen	362 m	IV d
D12	Kleiner Feldberg	8° 26' 51.2" E	50° 13' 22.9" N		822 m	IV b
D13	Bad Vilbel	8° 44' 48.9" E	50° 12' 26.9" N	OT Dortelweil	124 m	VI d
D14	Hanau	9° 00' 07.3" E	50° 08' 02.5" N		112 m	VI d
D15	Niddatal	8° 49' 47.4" E	50° 16' 27.3" N	OT Ilbenstadt	184 m	VI d



Fig. 2
Three sets of Rotenkamp samplers. Each set comprises two samplers - one sampler stabilized with hydrochloric acid and one with sodium hydroxide solution.

For a short period, sites D51 and 52 were used to obtain additional information on the effect of sampling height on bulk deposition. In order to check the plausibility of the data obtained at site D19, site D53 was operated as a site with comparable climate but different sources of potential artefacts, from birds in particular.

Samples were taken every month at the beginning of the month. Due to the size of Hesse, two days were needed to address all sites. Sampling started in July 2001 and ended in December 2004.

Table 1 (Continued)

No	Name	Geographic co-ordinates Gauss-Krüger co-ordinates ("Rechts wert" easting, "Hochwert", northing)	Located at (OT: Ortsteil)	Elevation above sea level	Correction class according to Richter (1995)
D16	Reichelsheim	8° 52' 40.8" E 50° 20' 05.3" N 3491315 5577525		121 m	VI a
D17	Herborn	8° 21' 36.5" E 50° 42' 36.9" N 3454810 5619470	OT Seelbach	276 m	IV c
D18	Biebertal	8° 36' 59.9" E 50° 36' 46.7" N 3472870 5608525	OT Rodheim-Bieber	203 m	IV d
D19	Linden	8° 41' 12.6" E 50° 32' 01.8" N 3477780 5599695	Steinweg	172 m	VI a
D20	Nidda	9° 00' 08.1" E 50° 25' 04.0" N 3500160 5586745		195 m	VI c
D21	Kefenrod	9° 12' 32.7" E 50° 20' 49.1" N 3514880 5578890		265 m	VI c
D22	Bad Soden- Salmünster	9° 25' 57.7" E 50° 16' 06.7" N 3530845 5570215	OT Alsberg	429 m	IV c
D23	Schotten	9° 05' 47.2" E 50° 28' 54.8" N 3506845 5593880		239 m	IV b
D24	Schotten	9° 12' 34.2" E 50° 30' 29.7" N 3514860 5596830	OT Breungeshain	583 m	VI d
D25	Ulrichstein	9° 14' 07.5" E 50° 34' 22.3" N 3516675 5604020	OT Rebgeshain	588 m	IV c
D26	Lauterbach	9° 21' 41.1" E 50° 36' 37.1" N 3525580 5608220	OT Frischborn	373 m	IV d
D27	Hatzfeld (Eder)	8° 34' 44.5" E 50° 57' 57.0" N 3470430 5647790	OT Eifa	424 m	IV d
D28	Cölbe	8° 48' 56.2" E 50° 52' 41.6" N 3487025 5637975	OT Schönstadt	245 m	VI a
D29	Stadtallendorf	9° 01' 40.4" E 50° 49' 28.5" N 3501965 5631995		263 m	VI c
D30	Alsfeld	9° 14' 57.4" E 50° 43' 33.0" N 3517600 5621040	OT Liederbach	292 m	IV d
D31	Wartenberg	9° 28' 24.8" E 50° 36' 44.5" N 3533515 5608495	OT Landenhausen	288 m	IV d
D32	Fulda	9° 42' 50.2" E 50° 33' 46.1" N 3550580 5603120		380 m	IV d
D33	Gersfeld	9° 50' 54.2" E 50° 27' 11.6" N 3560245 5591030	OT Altenfeld	399 m	IV c
D34	Gersfeld	9° 53' 30.4" E 50° 27' 24.6" N 3563320 5591470		453 m	IV d
D35	Wasserkuppe	9° 56' 16.7" E 50° 29' 57.7" N 3566540 5596240		942 m	IV b
D36	Hilders	9° 55' 52.1" E 50° 31' 44.7" N 3566015 5599540	OT Dietges	582 m	IV d
D37	Ehrenberg (Rhön)	10° 00' 39.9" E 50° 29' 20.2" N 3571745 5595150	OT Wüstensachsen	660 m	IV a
D38	Vöhl	8° 52' 58.0" E 51° 14' 16.1" N 3491815 5677965	OT Dorffitter	348 m	IV d
D39	Edertal	9° 05' 59.8" E 51° 09' 56.8" N 3506990 5669950	OT Mehlen	196 m	IV c
D40	Borken	9° 16' 42.7" E 51° 04' 51.8" N 3519515 5660560		225 m	IV d
D41	Knüllwald	9° 31' 35.5" E 50° 59' 22.7" N 3536965 5650485	OT Nausis	352 m	IV c
D42	Bad Hersfeld	9° 41' 11.3" E 50° 50' 55.1" N 3548340 5634895	OT Eichhof	201 m	IV a
D43	Hohenroda	9° 56' 16.6" E 50° 48' 52.2" N 3566095 5631290	OT Glaam	349 m	IV c
D44	Breuna	9° 10' 40.7" E 51° 27' 14.1" N 3512370 5702015	OT Wettesingen	232 m	IV d
D45	Calden	9° 22' 52.2" E 51° 24' 20.9" N 3526520 5696715		265 m	IV d
D46	Kassel	9° 28' 53.6" E 51° 18' 07.4" N 3533580 5685215		145 m	IV d
D47	Grossalmerode	9° 46' 32.5" E 51° 17' 38.5" N 3554100 5684500		594 m	IV d
D48	Wehretal	9° 59' 42.0" E 51° 09' 30.0" N 3569600 5669590	OT Reichensachsen	175 m	IV d
D49	Herleshausen	10° 10' 56.2" E 51° 02' 03.1" N 3582920 5655975	OT Willershausen	314 m	IV c
D50	Wahlsburg	9° 33' 03.3" E 51° 37' 12.2" N 3538150 5720625	OT Lippoldsberg	107 m	IV d
D51	Königstein	8° 26' 16.6" E 50° 12' 01.6" N 3459875 5562725	Naturfreundehaus	520 m	IV d
D52	Königstein	8° 25' 45.3" E 50° 12' 02.6" N 3459255 5562760	Turm 28 m	548 m	IV b
D53	Gießen	8° 42' 58.8" E 50° 33' 26.4" N 3479900 5602310	Schiffenberg	198 m	VI d

Table 2
Analytical details

Analyte	Method	Details and standards	Range of calibration		standard deviation of the detection	
Ca K Mg Na	AAS	AAS (flame) in accordance with German standard DIN 38 406, parts 3, 13 and 14	Ca: Na: K: Mg:	0.2 to 2 mg l ⁻¹ 0.1 to 1 mg l ⁻¹ 0.05 to 0.5 mg l ⁻¹ 0.1 to 1 mg l ⁻¹	Ca: K: Mg: Na:	< 2.5 % < 1 % < 2 % < 1 %
NH ₄ -N	SFA	Skalar SANplus system (modified Berthelot reaction)	NH ₄ -N:	0.3 to 3 mg l ⁻¹	NH ₄ -N:	< 1%
Cl NO ₃ -N SO ₄ -S	IC	Dionex DX 100 IC, column combination AG 12A, AS 12A, eluent Na ₂ CO ₃ /NaHCO ₃ , in accordance with German/European standards DIN 38 405 part 20, EN ISO 10304-1	Cl: NO ₃ -N: SO ₄ -S:	0.1 to 1 mg l ⁻¹ 0.1 to 1 mg l ⁻¹ 0.2 to 2 mg l ⁻¹	Cl: NO ₃ -N SO ₄ -S	< 1 % < 1.5 % < 2 %
<i>o</i> -PO ₄ -P	SFA	Skalar SANplus system in accordance with German standard DIN 38 405 D11-1	<i>o</i> -PO ₄ -P:	0.1 to 1 mg l ⁻¹	<i>o</i> -PO ₄ -P:	< 1%

Each site was equipped with three sets of Rotenkamp samplers. Rotenkamp samplers are funnel-bottle type samplers which are typically stabilized with hydrochloric acid or sodium hydroxide solution in order to avoid losses of any potentially fugitive gases (NH₃, SO₂, respectively) and minimize microbial activity. One pair of these samplers forms a set. The funnels are made from borosilicate glass with rounded edges to minimize deposition of aerosol constituents due to eddy formation at the edges. The three sets are mounted at each site in order to allow the detection of fouled samples or artefact formation and to minimize data losses. The apertures of the samplers are 1.5 m above ground level. The samplers were described in detail in Dämmgen et al., 2000; they proved to be effective in an international comparison, as illustrated in Dämmgen et al. (2005).

3 Sample analyses, correction and plausibility control

3.1 Analyses

All samples were weighed to determine the amount of precipitation. Acid samples were samples were micro-filtered and analysed straightaway by atomic absorption spectrometry (AAS: Ca, K, Mg, Na)¹, segmented flow analysis (SFA: NH₄-N, PO₄-P) and ion chromatography (IC: SO₄-S (s)). Alkaline samples were treated with H₂O₂ (10 ml, *c* = 30 g l⁻¹) to oxidize all sulfite. After about four weeks samples were micro-filtered and analyzed using ion chromatography (IC: Cl, NO₃-N, SO₄-S (b)). For details see Table 2.

In addition samples originating from Linden (D19) were analyzed for heavy metals permanently, others during part

of the experiment. The results of these measurements will be published separately (Dämmgen, 2007).

3.2 Plausibility checks

Dämmgen et al. (2000) have shown that for the major deposition constituents, standard deviations between parallels of 10 % are to be considered normal. As three parallel samples can be obtained, standard deviations > 10 % are checked with respect to their ion balance and fouling indicators. In contrast to German Guideline LAWA (1996), which takes into account NO₃-N, SO₄-S, Cl, PO₄-P, NH₄-N, Na, K, Mg, Ca and Al, this check includes NO₃-N, SO₄-S (s), Cl, NH₄-N, Na and Mg, as these ions are deposited together. Ca and K are to a larger extent deposited dry and are typically combined with ions not accounted for in the analysis (silicates, carbonates). Q_{eq} according to:

$$Q_{eq} = \frac{\frac{D_{NO_3-N}}{14} + 2 \cdot \frac{D_{SO_4-S(s)}}{32} + \frac{D_{Cl}}{35.4}}{\frac{D_{NH_4-N}}{14} + \frac{D_{Na}}{23} + 2 \cdot \frac{D_{Mg}}{24.3}}$$

serves as an indicator. High PO₄-P depositions denote fouling by birds, Cl contamination by humans.

In addition, concentrations are checked using the concentration frequency distributions (see Figs. 3 to 11 in chapter 4.1, the bar on the far right denoting concentrations that are considered implausible). Also, the comparatively short distances between sampling sites often allow for a check on plausibility. Furthermore, ions, which are often deposited or related to each other, such as Na with Cl, SO₄ (s) and SO₄ (b), Ca and K can be compared to check plausibility.

¹ Species are named in the alphabetical order of their chemical symbols.

Table 3

Mean corrections $\Delta D_{\text{H}_2\text{O}}$ for annual precipitation reflecting regional and sheltering situations (Richter, 1995), in % to be added to the measured volume

a: unsheltered; b: slightly sheltered; c: moderately sheltered; d: well sheltered

region	degree of shelter	$\Delta D_{\text{H}_2\text{O}}$
II, IV	a	16.6
	b	13.5
	c	11.1
	d	8.6
III	a	18.2
	b	14.6
	c	12.0
	d	9.3
VI	a	14.9
	b	12.3
	c	10.4
	d	8.2

The fact that three parallels exist allows the rejection of the one sample that is not in line with the remaining two. If all three parallels fail to be plausible, data may be replaced by those of a neighbouring site, if the amount of precipitation and the chemical composition are similar. If data are missing and cannot be replaced, annual totals are obtained by linear extrapolation as suggested by Dämmgen et al. (2000). If more than three months per year are missing the annual total of the site is not reported at all.

3.3 Flow distortion correction

At a sampling height of 1.5 m, prevailing wind speeds and rain drop size frequency distributions cause bulk samplers as used to collect less than at ground level. As this bias cannot be overcome by changes in the sampling techniques, corrections have to be applied. As shown by Dämmgen et al. (2000), Rotenkamp samples were shown to sample the same amount of rain as standard Hellmann gauges (German standard DIN 58 666). The German Weather Service corrects precipitation measured with

Hellmann samplers using a correction procedure developed by Richter (1995), which considers regional differences as well as sheltering effects (Table 3). As the species considered are mainly (60 to 100 %) deposited with precipitation (Dämmgen et al., 1996, Dämmgen and Zimmerling, 2002), Richter's correction is also applied to bulk deposition of species considered in this report. For each site, the correction class is listed in Table 1. The correction factors are listed in Table A1 in the annex.

3.4 Sea salt correction

It is common practice in deposition mapping to correct deposition for sea salt inputs, as these are considered to be natural. This presupposes that the air masses responsible for the respective deposition did not pass sources changing the Na and Cl composition significantly. This cannot be assumed for Hesse (cf Armbruster et al., 2002). In addition, this survey aimed at quantifying total bulk deposition without any discrimination of particular sources. Thus, no sea salt correction was applied.

3.5 Data availability

Table 4 illustrates how many data sets were implausible, incomplete, needed replacement or could not even be obtained by replacement. A total loss has to be stated for site D40, which had to be closed as early as October 2001. In 2004, no valid total could be obtained for site D50. The remaining data losses are to a large extent due to extreme weather situations, such as in January and February 2002, when almost all samplers overflowed. During spring, fouling by (young) birds is the major reason for losses. In order to get comparable annual totals, the decision was made to consider the period between March 2002 and February 2003 as surrogate for "real" 2002 annual totals. Thus, the overall three years' mean (2002 to 2004) is the mean of "surrogate 2002", 2003 and 2004.

Table 4

Data availability

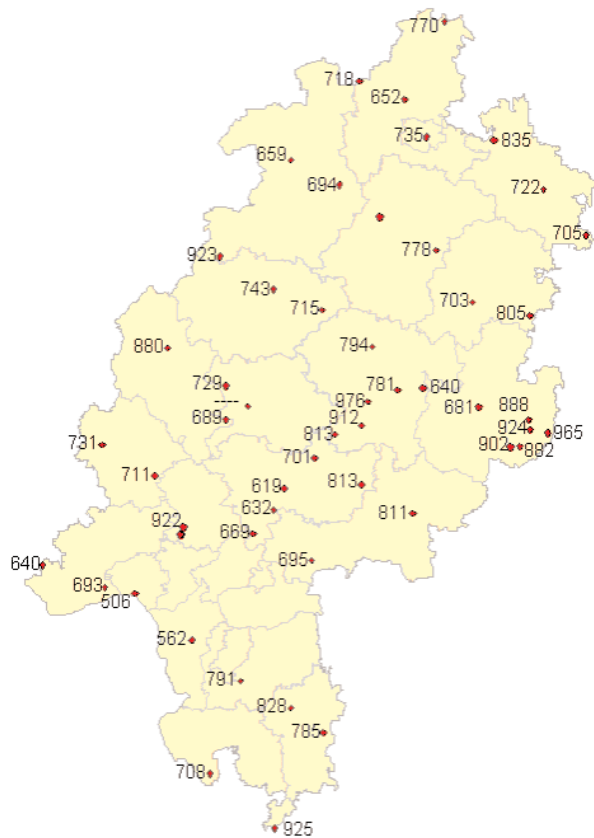
	Maximum number of samples	Number of samples not rejected	Monthly means obtained % of means possible	2002	Valid annual totals obtained Number of sites		
					12 months 2002/2003	2003	2004
Precipitation	6048	5226	91.4	49	49	49	48
Ca	6048	4188	87.3	47	49	49	48
Cl	6048	4702	90.5	49	49	49	47
K	6048	3835	84.8	48	49	47	47
Mg	6048	4288	87.6	47	49	49	47
NH ₄ -N	6048	3907	87.2	48	49	48	47
NO ₃ -N	6048	5052	90.7	49	49	49	48
Na	6048	4413	87.9	48	49	49	47
SO ₄ -S(b)	6048	4850	90.5	48	49	49	48
SO ₄ -S(s)	6048	4548	88.7	48	49	49	47

4 Results

4.1 Bulk deposition means for 2002 to 2004 – an overview

The data are condensed in maps for each of the ten species considered. These are complemented with frequency distributions and a short summary containing maximum and minimum values. Details are provided in Tables A1 to A5 in the appendix.

4.1.1 Precipitation



Map 2
 Mean annual precipitation in mm a^{-1} , 2002 to 2004.
 overall mean: 764 mm a^{-1}
 standard deviation of the 3 annual means: 122 mm a^{-1}
 highest overall total: 976 mm a^{-1} , Ulrichstein (D25)
 lowest overall total: 507 mm a^{-1} , Wiesbaden (D05)
 highest annual amount observed: 1095 mm a^{-1} , Neckarsteinach (D02), 2002
 lowest annual amount observed: 429 mm a^{-1} , Wiesbaden (D05), 2004

4.1.2 Calcium (Ca)

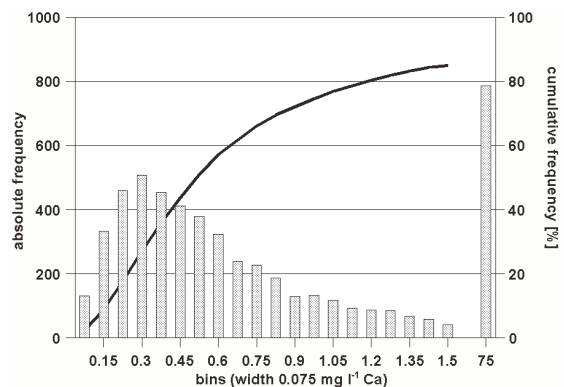
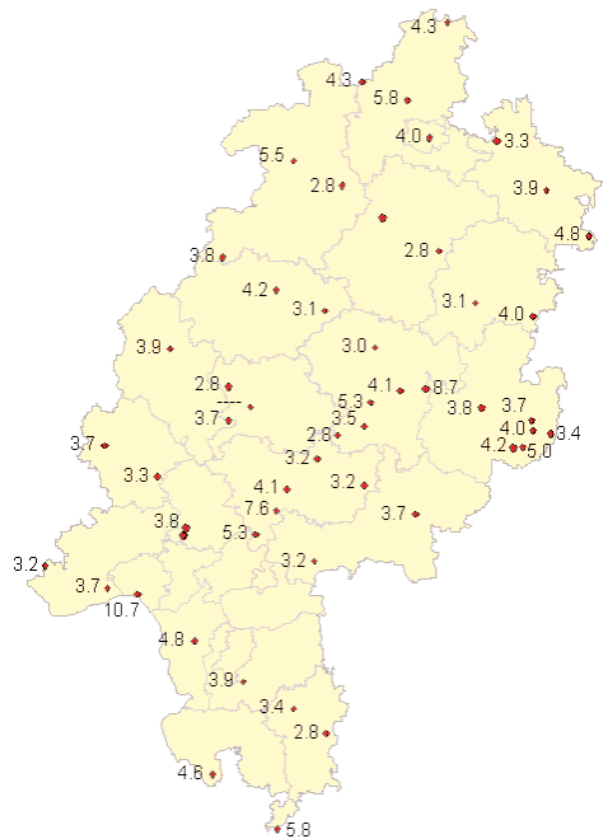


Fig. 3
 Frequency distribution of Ca concentrations (single samples). Bin width 0.075 mg l^{-1} .



Map 3
 Mean Ca deposition in $\text{kg ha}^{-1} \text{ a}^{-1}$ Ca, 2002 to 2004.
 overall mean: $4.2 \text{ kg ha}^{-1} \text{ a}^{-1}$ Ca
 standard deviation of the 3 annual means: $1.7 \text{ kg ha}^{-1} \text{ a}^{-1}$ Ca
 highest overall total: $10.7 \text{ kg ha}^{-1} \text{ a}^{-1}$, Wiesbaden (D05)
 lowest overall total: $2.8 \text{ kg ha}^{-1} \text{ a}^{-1}$, Erbach (D08), Biebertal (D18), Schotten (D23), Edertal (D39), Knüllwald (D41)
 highest annual total observed: $12.7 \text{ kg ha}^{-1} \text{ a}^{-1}$, Wiesbaden (D05), 2003
 lowest annual total observed: $2.1 \text{ kg ha}^{-1} \text{ a}^{-1}$, Biebertal (D18), 2002

4.1.3 Chloride (Cl)

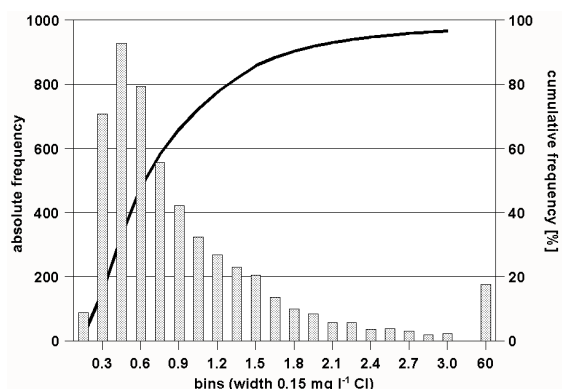


Fig. 4
Frequency distribution of Cl concentrations (single samples). Bin width 0.15 mg l⁻¹

4.1.4 Potassium (K)

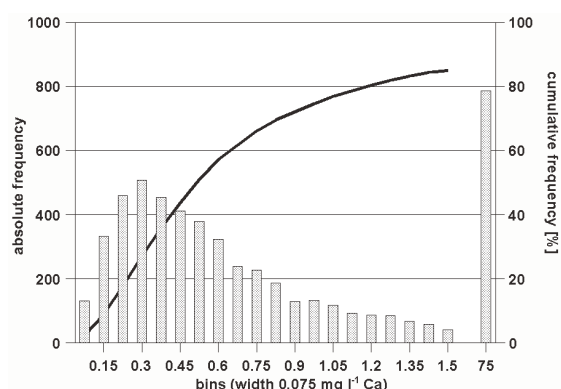
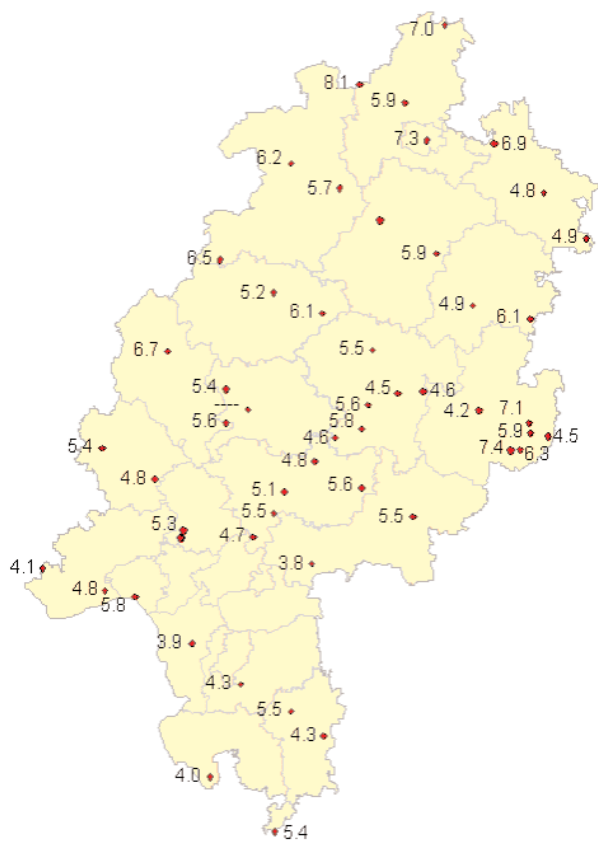
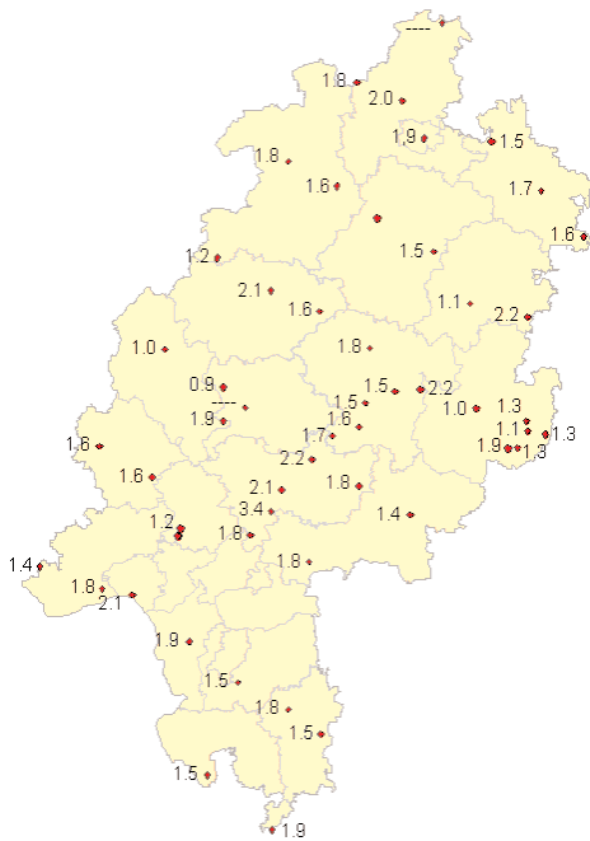


Fig. 5
Frequency distribution of K concentrations (single samples). Bin width 0.03 mg l⁻¹



Map 4
Mean Cl deposition in kg ha⁻¹ a⁻¹ Cl. 2002 to 2004.
overall mean: 5.5 kg ha⁻¹ a⁻¹ Cl
standard deviation of the 3 annual means: 1.3 kg ha⁻¹ a⁻¹ Cl
highest overall total: 8.1 kg ha⁻¹ a⁻¹, Breuna (D44)
lowest overall total: 3.8 kg ha⁻¹ a⁻¹, Hanau (D22)
highest annual total observed: 11.3 kg ha⁻¹ a⁻¹, Kassel (D46), 2004
lowest annual total observed: 3.1 kg ha⁻¹ a⁻¹, Groß-Gerau (D06), 2003



Map 5
Mean K deposition in kg ha⁻¹ a⁻¹ K. 2002 to 2004.
overall mean: 1.7 kg ha⁻¹ a⁻¹ K
standard deviation of the 3 annual means: 0.6 kg ha⁻¹ a⁻¹ K
highest overall total: 3.4 kg ha⁻¹ a⁻¹, Niddatal (D15)
lowest overall total: 0.9 kg ha⁻¹ a⁻¹, Biebental (D18)
highest annual total observed: 3.9 kg ha⁻¹ a⁻¹, Niddatal (D15), 2004
lowest annual total observed: 0.8 kg ha⁻¹ a⁻¹, Biebental (D18),
Wasserkuppe (D35), Bad Hersfeld (D42), 2002

4.1.5 Magnesium (Mg)

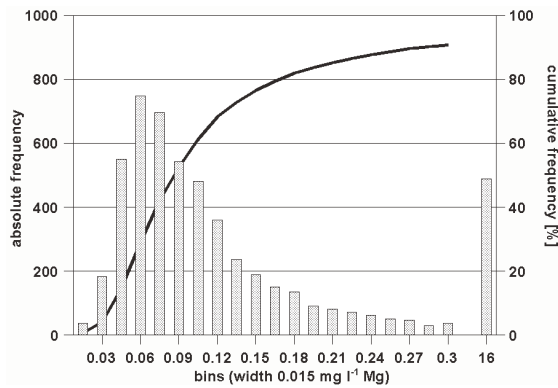


Fig. 6
Frequency distribution of Mg concentrations (single samples). Bin width 0.015 mg l⁻¹

4.1.6 Ammonium-nitrogen (NH₄-N)

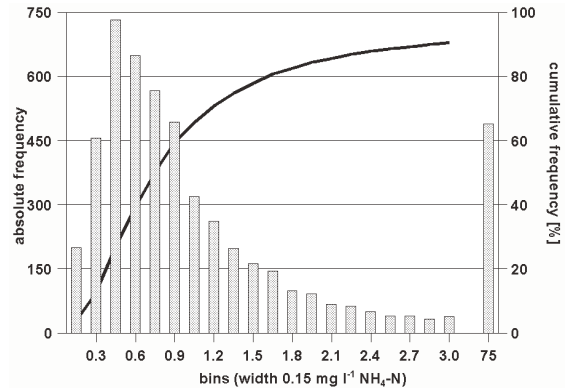
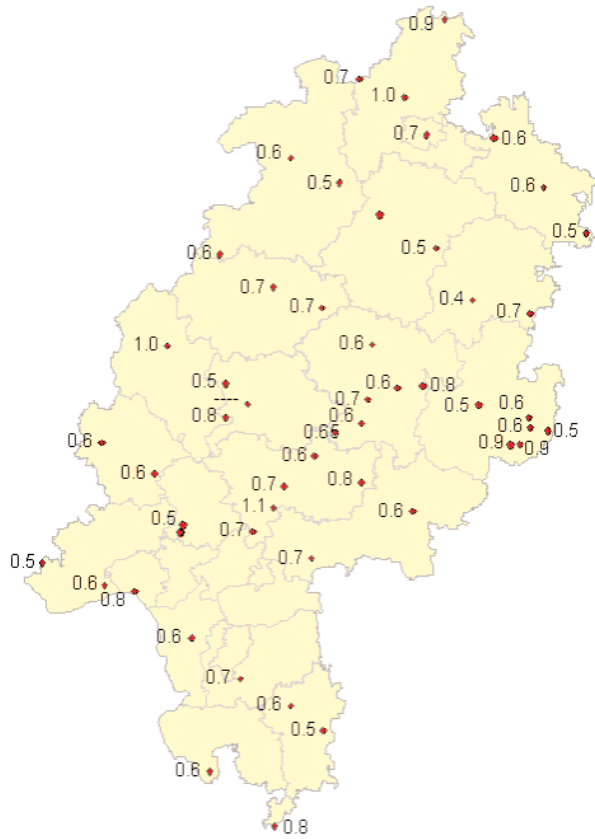
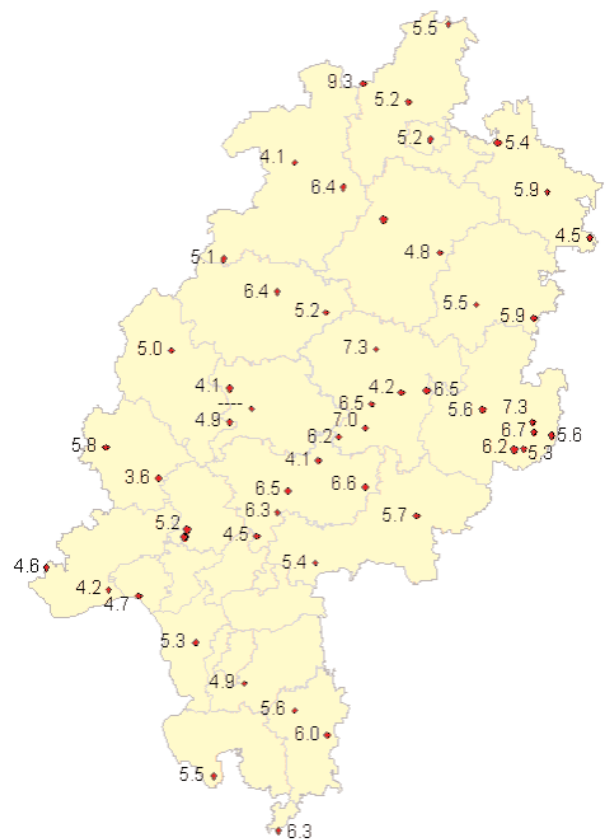


Fig. 7
Frequency distribution of NH₄-N concentrations (single samples). Bin width 0.15 mg l⁻¹



Map 6
Mean Mg deposition in kg ha⁻¹ a⁻¹ Mg. 2002 to 2004.

overall mean: 0.66 kg ha⁻¹ a⁻¹ Mg
 standard deviation of the 3 annual means: 0.20 kg ha⁻¹ a⁻¹ Mg
 highest overall total: 1.1 kg ha⁻¹ a⁻¹, Niddatal (D15)
 lowest overall total: 0.45 kg ha⁻¹ a⁻¹, Bad Hersfeld (D42)
 highest annual total observed: 1.7 kg ha⁻¹ a⁻¹, Gersfeld (D34), 2003
 lowest annual total observed: 0.39 kg ha⁻¹ a⁻¹, Fulda (D32), 2004



Map 7
Mean NH₄-N deposition in kg ha⁻¹ a⁻¹ NH₄-N. 2002 to 2004.

overall mean: 5.6 kg ha⁻¹ a⁻¹ NH₄-N
 standard deviation of the 3 annual means: 1.3 kg ha⁻¹ a⁻¹ NH₄-N
 highest overall total: 9.3 kg ha⁻¹ a⁻¹, Breuna (D44)
 lowest overall total: 3.6 kg ha⁻¹ a⁻¹, Selters (D11)
 highest annual total observed: 10.1 kg ha⁻¹ a⁻¹, Breuna (D44), 2002
 lowest annual total observed: 3.2 kg ha⁻¹ a⁻¹, Selters (D11), 2004

4.1.7 Nitrate-nitrogen (NO₃-N)

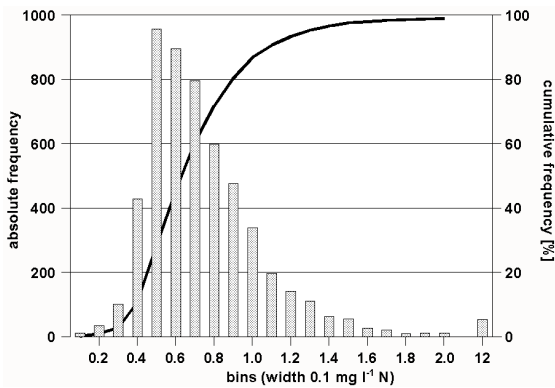


Fig. 8
Frequency distribution of NO₃-N concentrations (single samples). Bin width 0.10 mg l⁻¹

4.1.8 Sodium (Na)

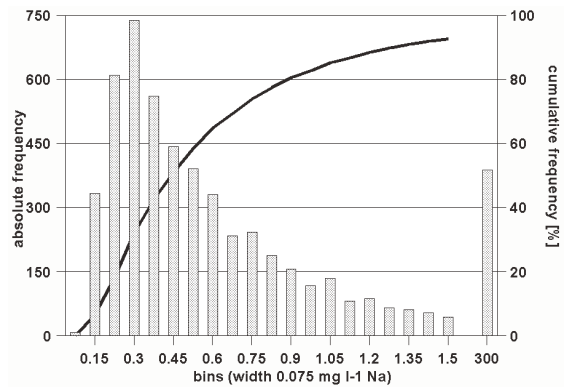
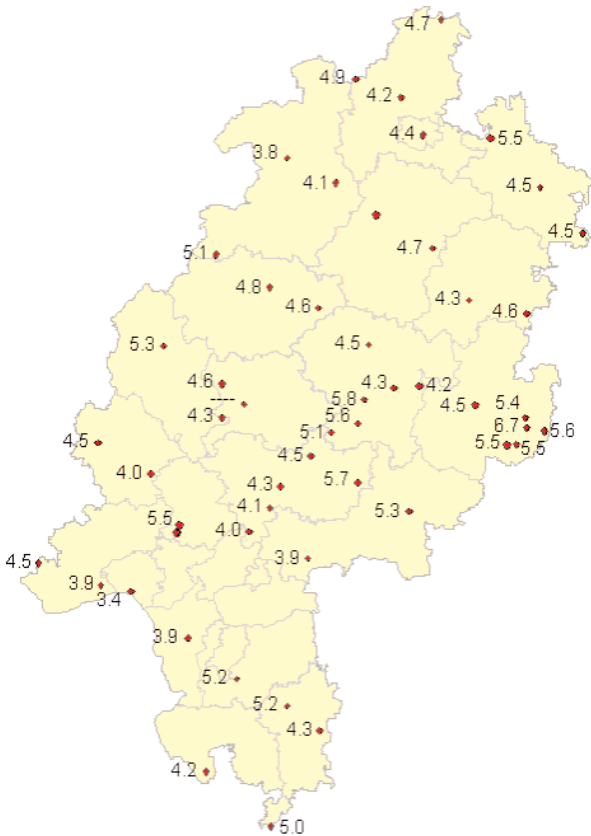
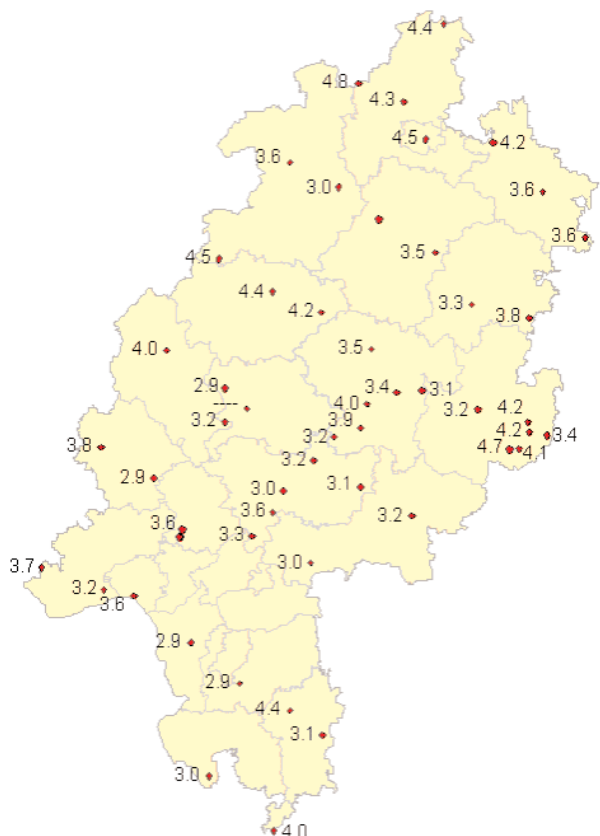


Fig. 9
Frequency distribution of Na concentrations (single samples). Bin width 0.075 mg l⁻¹



Map 8
Mean NO₃-N deposition in kg ha⁻¹ a⁻¹ NO₃-N. 2002 to 2004.

overall mean: 4.7 kg ha⁻¹ a⁻¹ NO₃-N
 standard deviation of the 3 annual means: 0.8 kg ha⁻¹ a⁻¹ NH₄-N
 highest overall total: 6.7 kg ha⁻¹ a⁻¹, Wasserkuppe (D35)
 lowest overall total: 3.4 kg ha⁻¹ a⁻¹, Wiesbaden (D05)
 highest annual total observed: 7.2 kg ha⁻¹ a⁻¹, Wasserkuppe (D35), 2002
 lowest annual total observed: 3.1 kg ha⁻¹ a⁻¹, Wiesbaden (D05), 2004



Map 9
Mean Na deposition in kg ha⁻¹ a⁻¹ Na. 2002 to 2004.

overall mean: 3.7 kg ha⁻¹ a⁻¹ Na
 standard deviation of the 3 annual means: 0.8 kg ha⁻¹ a⁻¹ Na
 highest overall total: 4.8 kg ha⁻¹ a⁻¹, Breuna (D44)
 lowest overall total: 2.9 kg ha⁻¹ a⁻¹, Biebertal (D18) und Selters (D11)
 highest annual total observed: 7.8 kg ha⁻¹ a⁻¹, Hatzfeld (D27), 2004
 lowest annual total observed: 2.1 kg ha⁻¹ a⁻¹, Kleiner Feldberg (D12), 2002

4.1.9 Sulfate-sulfur (basic) (SO₄-S (b))

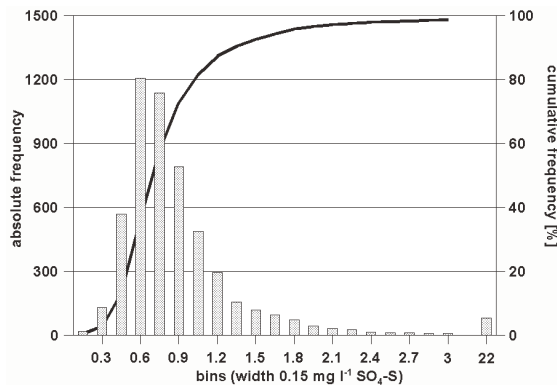


Fig. 10
Frequency distribution of SO₄-S (b) concentrations (single samples).
Bin width 0.15 mg l⁻¹

4.1.10 Sulfate-sulfur (acid) (SO₄-S (s))

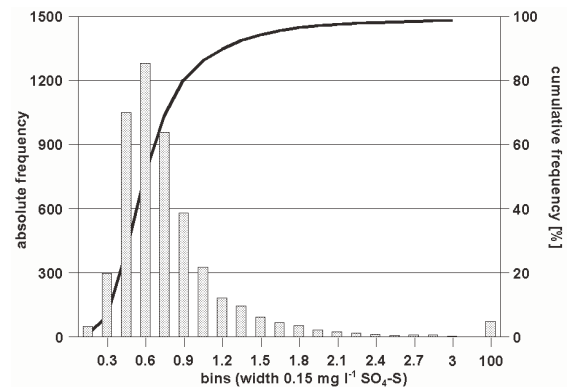
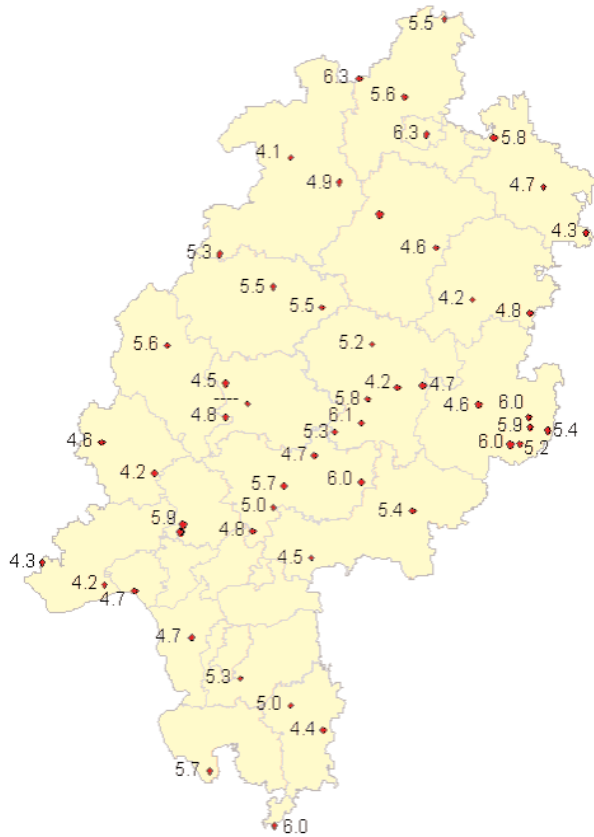
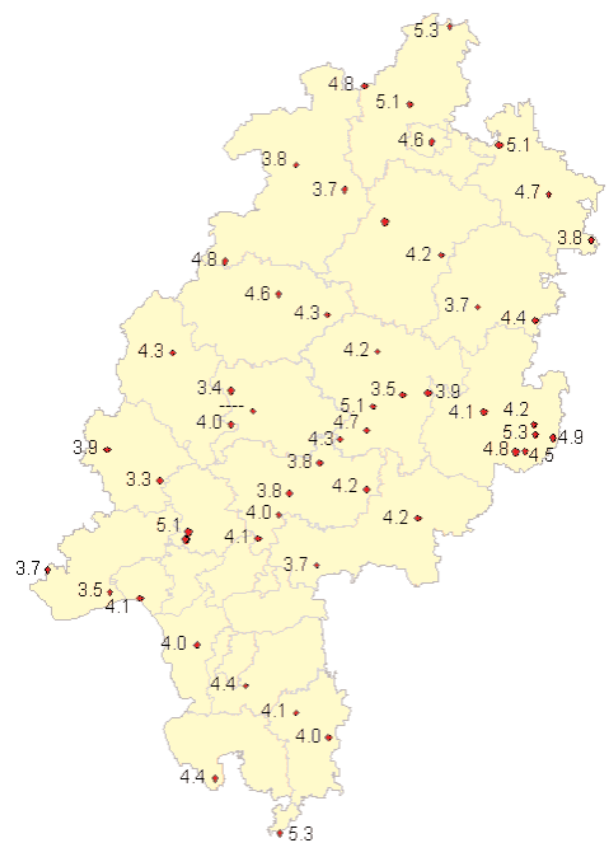


Fig. 11
Frequency distribution of SO₄-S (s) concentrations (single samples).
Bin width 0.15 mg l⁻¹



Map 10
Mean SO₄-S (b) deposition in kg ha⁻¹ a⁻¹ SO₄-S. 2002 to 2004.

overall mean: 5.1 kg ha⁻¹ a⁻¹ SO₄-S
standard deviation of the 3 annual means: 0.9 kg ha⁻¹ a⁻¹ SO₄-S
highest overall total: 6.3 kg ha⁻¹ a⁻¹, Kassel (D46)
lowest overall total: 4.1 kg ha⁻¹ a⁻¹, Vöhl (D38)
highest annual total observed: 8.4 kg ha⁻¹ a⁻¹, Breuna (D44), 2002
lowest annual total observed: 3.6 kg ha⁻¹ a⁻¹, Lauterbach (D26), 2003



Map 11
Mean SO₄-S (s) deposition in kg ha⁻¹ a⁻¹ SO₄-S. 2002 to 2004.

overall mean: 4.3 kg ha⁻¹ a⁻¹ SO₄-S
standard deviation of the 3 annual means: 0.7 kg ha⁻¹ a⁻¹ SO₄-S
highest overall total: 5.3 kg ha⁻¹ a⁻¹, Wasserkuppe (D35)
lowest overall total: 3.3 kg ha⁻¹ a⁻¹, Selters (D11)
highest annual total observed: 7.3 kg ha⁻¹ a⁻¹, Caldén (D45), 2002
lowest annual total observed: 2.7 kg ha⁻¹ a⁻¹, Stadtlendorf (D29), 2003

4.2 Correlations

4.2.1 Precipitation and elevation

As rule, precipitation increases with height in the central German mountains (Mittelgebirge). This also applies to Hesse where the highest precipitation was observed at Kleiner Feldberg (D12) in the northern part of Vogelsberg (Ulrichstein, D25) and the Rhön (Ehrenberg, D37). However, no clear linear correlation can be observed for either the whole of Hesse or for the Vogelsberg catena (Fig. 12).

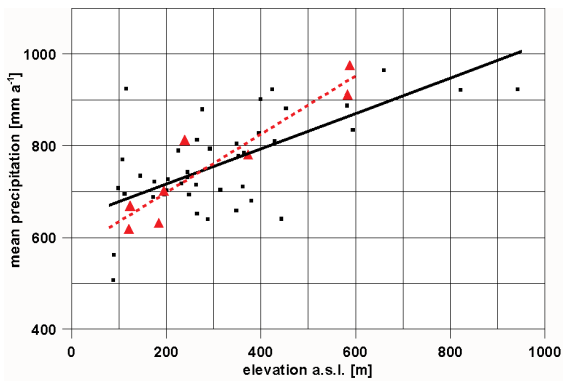


Fig. 12
Relation between overall mean precipitation and elevation a.s.l. for all Hesse (squares) and the Vogelsberg catena (triangles). The respective linear regression is shown as solid (Hesse) or dotted line (Vogelsberg)

4.2.2 Selected species and precipitation

Species vary considerably with respect to their behaviour as compared with precipitation. $\text{NO}_3\text{-N}$ deposition increases with precipitation, whereas concentrations decrease slightly. This is observed both for Hesse as a whole and the Vogelsberg catena in particular (Figs. 13 and 14).

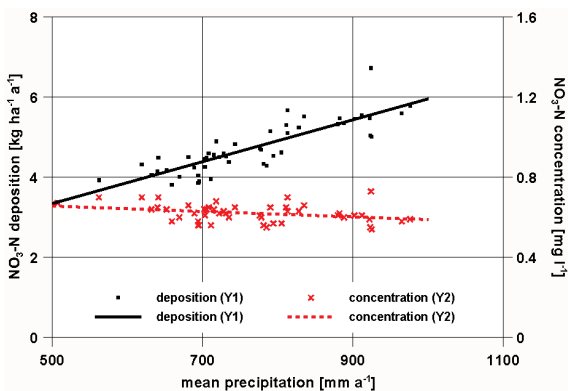


Fig. 13
Mean bulk deposition (Y1, squares) and concentrations (Y2, crosses) of $\text{NO}_3\text{-N}$ in Hesse as a function of precipitation (2002 to 2004) and the respective linear regressions (depositions solid, $R^2 = 0.74$; concentrations dotted, $R^2 = 0.11$).

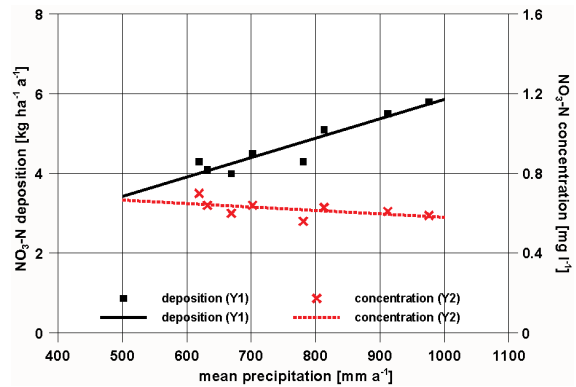


Fig. 14
Mean bulk deposition (Y1, squares) and concentrations (Y2, crosses) of $\text{NO}_3\text{-N}$ in the Vogelsberg catena as a function of precipitation (2002 to 2004) and the respective linear regressions (depositions solid, $R^2 = 0.88$; concentrations dotted, $R^2 = 0.31$).

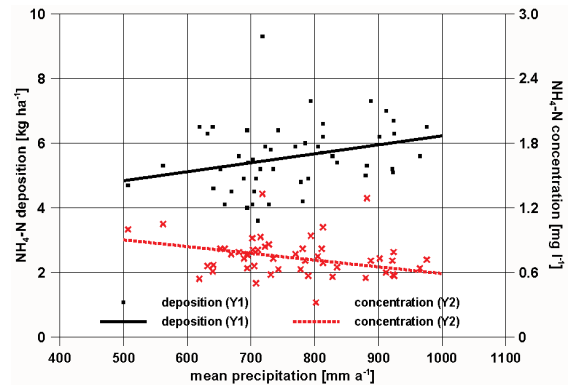


Fig. 15
Mean bulk deposition (Y1, squares) and concentrations (Y2, crosses) of $\text{NH}_4\text{-N}$ in Hesse as a function of precipitation (period 2002 to 2004) and the respective linear regressions (depositions solid, $R^2 = 0.08$; concentrations dotted, $R^2 = 0.20$).

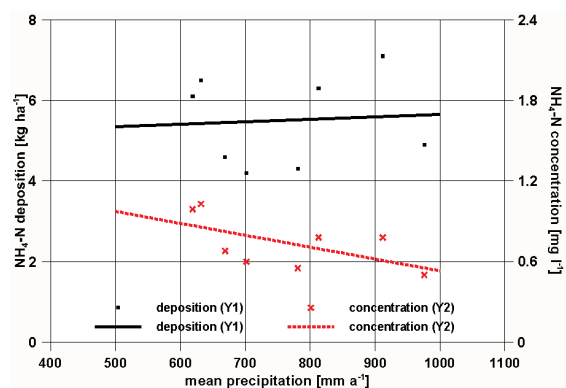


Fig. 16
Mean bulk deposition (Y1, squares) and concentrations (Y2, crosses) of $\text{NH}_4\text{-N}$ in the Vogelsberg catena as a function of precipitation (period 2002 to 2004) and the respective linear regressions (depositions solid, $R^2 = 0.08$; concentrations dotted, $R^2 = 0.20$).

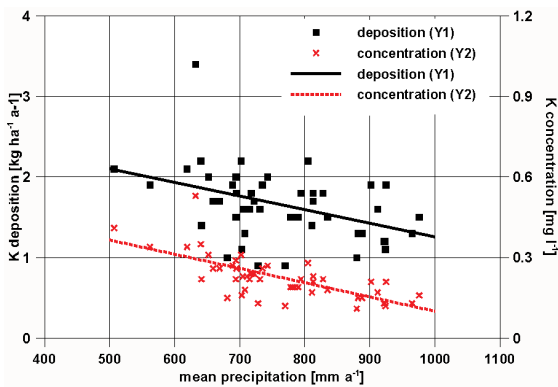


Fig. 17 Mean bulk deposition (Y1, squares) and concentrations (Y2, crosses) of K in Hesse as a function of precipitation (period 2002 to 2004) and the respective linear regressions (depositions solid, $R^2 = 0.19$; concentrations dotted, $R^2 = 0.52$).

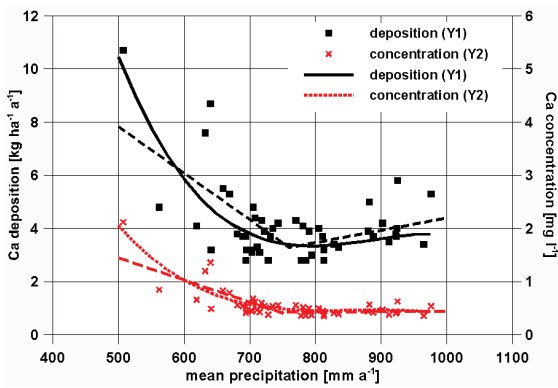


Fig. 18 Mean bulk deposition (Y1, squares) and concentrations (Y2, crosses) of Ca in Hesse as a function of precipitation (period 2002 to 2004) and the respective polynomial regressions (depositions solid, $R^2 = 0.51$; concentrations dotted, $R^2 = 0.72$).

For $\text{NO}_3\text{-N}$, this relation can be used for data gap filling. Similar behaviour can be observed for deposition and precipitation for $\text{NH}_4\text{-N}$, $\text{SO}_4\text{-S}$, Cl and Na, though the slope of the regression line decreases in this order. However, for $\text{NH}_4\text{-N}$ the regression becomes so weak that the gap-filling procedure recommended for $\text{NO}_3\text{-N}$ cannot be used for $\text{NH}_4\text{-N}$ (Figs 15 and 16)!

For K, both deposition and concentration decrease with increasing precipitation, indicating that emission and atmospheric transport of K differ from those of the species mentioned previously (Fig. 17).

Finally, the relation between Ca deposition and concentration and precipitation appears to be best described as non-linear.

From this I conclude that the ratio of precipitation amounts $D_{\text{H}_2\text{O},1}$ and $D_{\text{H}_2\text{O},2}$ at two locations 1 and 2 and a concentration of a species c_A at location 1

$$D_{A,2} \neq \frac{D_{\text{H}_2\text{O},2}}{D_{\text{H}_2\text{O},1}} \cdot c_{A,1}$$

cannot be used to extrapolate a deposition $D_{A,2}$ apart from a very limited number of exceptions (i.e. similar amounts of precipitation and similar structure of nearby ammonia sources).

4.2.3 Relations between depositions and concentrations of species in bulk deposition

Na and Cl in rain and atmospheric aerosol and hence in bulk deposition predominantly originate in sea spray. This should be and is reflected by the observations of aerosol constituents in Northern Germany (Dämmgen, 2002, 2005). However, the few measurements performed at Linden indicate that this is unlikely to be true for Hesse (Dämmgen, unpublished). The molar ratio of Na to Cl, $x_{\text{NaCl, sea}}$ is $0.85 \text{ mol mol}^{-1}$. This is also true for the regions with large depositions of Cl. It increases with decreasing Cl depositions and is about 1.0 for a deposition of $0.15 \text{ kmol ha}^{-1} \text{ a}^{-1}$ Cl, indicating that the air mass resulting in bulk deposition is slightly depleted with Cl. One reason may be the reaction of NaCl with strong and less volatile acids during atmospheric transport (Fig. 19).

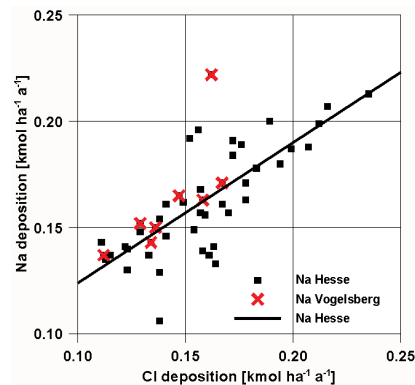


Fig. 19 Mean bulk deposition of Na throughout Hesse (squares) and in the Vogelsberg catena (crosses) as a function of Cl bulk deposition (means for 2002 to 2004) and the respective linear regression (Hesse solid, $R^2 = 0.62$; Vogelsberg dotted, $R^2 = 0.60$).

A regression analysis for relations between nitrogen and other depositions shows that these are generally closer along the Vogelsberg catena than in total Hesse. Table 5 illustrates that NH_4 , SO_4 , NO_3 and to some extent Cl are transported and deposited together. Interestingly, Ca and Cl are deposited together along the Vogelsberg catena. The correlations between (partly soil-borne) Ca, Mg and K are significantly high.

4.3 Annual variation

Mean precipitation, Na and Cl depositions peak in January. However, the influence of the summer monsoon can also be traced in the period under investigation. In contrast

Table 5

Correlation coefficients R^2 for linear regressions between depositions (upper right halves) and concentrations (lower left halves) for all Hessian sites (upper table) and the sites of the Vogelsberg catena (lower table). H_2O : precipitation; cells with $R^2 > 0.6$ with frame

	H_2O	Ca	Cl	K	Mg	NH_4-N	NO_3-N	Na	$SO_4-S(b)$	$SO_4-S(s)$	SO_2-S
H_2O	1.00	0.12	0.10	0.19	0.00	0.08	0.74	0.18	0.28	0.38	0.00
Ca	0.34	1.00	0.00	0.24	0.25	0.00	0.09	0.00	0.00	0.00	0.02
Cl	0.20	0.22	1.00	0.00	0.11	0.14	0.12	0.62	0.32	0.18	0.13
K	0.52	0.49	0.18	1.00	0.26	0.02	0.18	0.00	0.00	0.01	0.01
Mg	0.32	0.57	0.50	0.64	1.00	0.01	0.00	0.09	0.08	0.04	0.04
NH_4-N	0.20	0.16	0.21	0.28	0.17	1.00	0.16	0.13	0.38	0.20	0.17
NO_3-N	0.11	0.06	0.08	0.05	0.06	0.20	1.00	0.18	0.43	0.42	0.04
Na	0.25	0.30	0.67	0.29	0.29	0.21	0.10	1.00	0.31	0.43	0.00
$SO_4-S(b)$	0.32	0.30	0.40	0.38	0.37	0.47	0.33	0.39	1.00	0.57	0.34
$SO_4-S(s)$	0.31	0.49	0.29	0.38	0.33	0.30	0.20	0.53	0.63	1.00	0.01

	H_2O	Ca	Cl	K	Mg	NH_4-N	NO_3-N	Na	$SO_4-S(b)$	$SO_4-S(s)$	SO_2-S
H_2O	1.00	0.09	0.16	0.40	0.22	0.11	0.88	0.53	0.22	0.59	0.30
Ca	0.37	1.00	0.19	0.45	0.85	0.02	0.13	0.16	0.01	0.01	0.23
Cl	0.45	0.67	1.00	0.01	0.16	0.72	0.23	0.60	0.58	0.44	0.00
K	0.61	0.71	0.63	1.00	0.76	0.02	0.27	0.02	0.01	0.07	0.12
Mg	0.55	0.90	0.50	0.91	1.00	0.06	0.19	0.04	0.00	0.01	0.03
NH_4-N	0.21	0.28	0.76	0.44	0.46	1.00	0.32	0.22	0.80	0.37	0.03
NO_3-N	0.31	0.05	0.38	0.34	0.22	0.66	1.00	0.40	0.50	0.69	0.02
Na	0.56	0.92	0.80	0.77	0.93	0.36	0.14	1.00	0.21	0.60	0.01
$SO_4-S(b)$	0.49	0.25	0.74	0.47	0.45	0.78	0.83	0.43	1.00	0.54	0.04
$SO_4-S(s)$	0.49	0.58	0.68	0.59	0.61	0.49	0.44	0.65	0.69	1.00	0.05

to the past, maximum SO_4 depositions can be observed to coincide with maximum NH_4 depositions during the periods where NH_3 emissions from the application of slurry, farmyard manure and mineral fertilizer are highest.

4.4 Representativeness in time of Hessian annual deposition means

As the deposition data are to be used to establish a representative atmospheric nitrogen balance, it has to be checked whether or not they fit into the order of magnitude of the available time series. Although January 2002 was very wet and the summer of 2003 was very dry, the

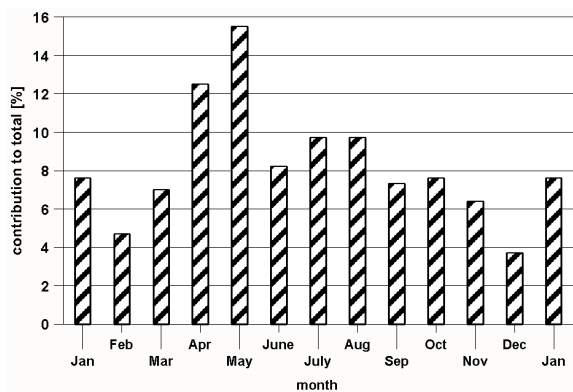


Fig. 20 Mean annual variation of the NH_4-N deposition in Hesse, showing the mean monthly contribution to the annual deposition.

data used for comparison indicate that the three years considered were not extraordinary. The time series of Rotenkamp (near Königslutter) and FAL near Braunschweig as well as the 10 years of measurements at Linden show the same pattern and similar depositions as the Hessian data (Figs 22 to 25).

Whereas the depositions of NO_3-N and SO_4-S in overall Hesse are quite similar to those measured in the Braunschweig region, Linden is obviously a site with less air pollution. The NH_4-N time series exhibits a peak value for 1994 and Rotenkamp, which cannot be explained. In general, the pattern follows that of the emissions modelled for this period: the only major kink coincides with the Ger-

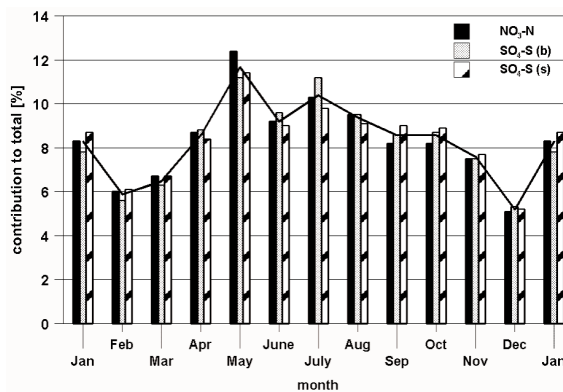


Fig. 21 Mean annual variation of the NO_3-N and SO_4-S depositions in Hesse, showing the mean monthly contribution to the annual deposition.

man unification and the subsequent decrease in agricultural emissions due to the decrease in animal populations (Lüttich and Dämmgen, unpublished). For both $\text{NO}_3\text{-N}$

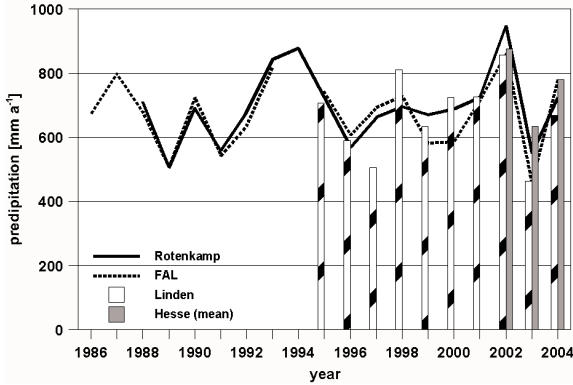


Fig. 22 Annual overall precipitation measured at FAL (west of Braunschweig), Rotenkamp (east of Braunschweig), Linden (D19) and in Hesse (mean)

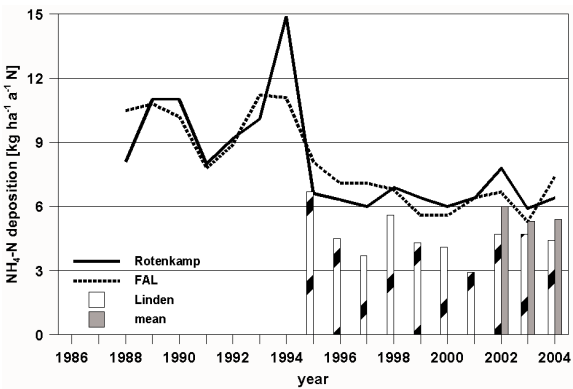


Fig. 23 Annual overall $\text{NH}_4\text{-N}$ deposition measured at FAL (west of Braunschweig), Rotenkamp (east of Braunschweig), Linden (D19) and in Hesse (mean)

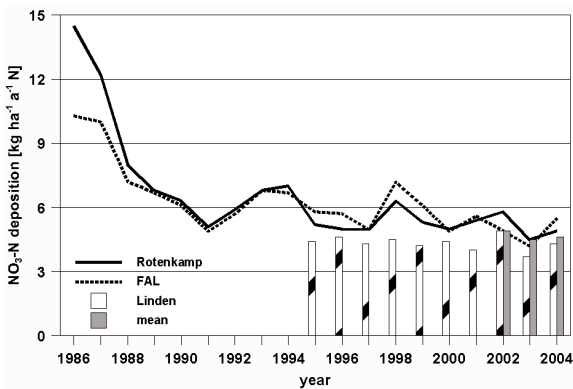


Fig. 24 Annual overall $\text{NO}_3\text{-N}$ deposition measured at FAL (west of Braunschweig), Rotenkamp (east of Braunschweig), Linden (D19) and in Hesse (mean)

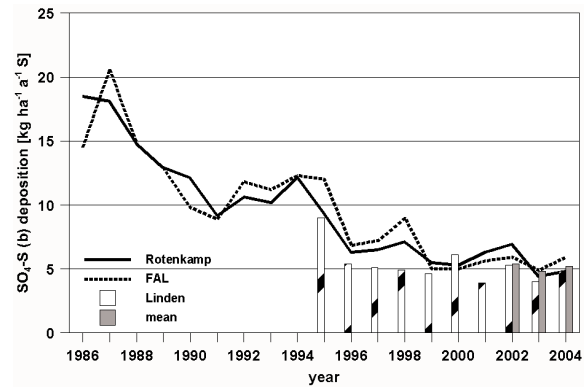


Fig. 25 Annual overall $\text{SO}_4\text{-S}$ (b) deposition measured at FAL (west of Braunschweig), Rotenkamp (east of Braunschweig), Linden (D19) and in Hesse (mean)

and $\text{SO}_4\text{-S}$ there was a distinctive reduction in depositions within the two decades considered. For $\text{SO}_4\text{-S}$, depositions levelled out from around year 2000, for $\text{NO}_3\text{-N}$ from 1990. The rapid decrease before 1990 (both for $\text{SO}_4\text{-S}$ and $\text{NO}_3\text{-N}$) can be attributed to the effect of the legislation concerning large combustion sources (“Großfeuerungsanlagen-Verordnung”) in former West Germany. The effect of the breakdown of the economy (energy generation in particular) in the new federal states after unification is partly responsible for the reduction of SO_2 emissions after 1990 and thus of $\text{SO}_4\text{-S}$ depositions.

4.5 Representativeness in space and accuracy of nitrogen bulk deposition measurements

In order to derive quality criteria, the overall N depositions are compared with the critical loads for eutrophying N. These critical loads are reported with an accuracy or resolution of 3 to 5 $\text{kg ha}^{-1} \text{a}^{-1} \text{N}$ (Skeffington, 2006; Skeffington et al., 2006). From this it can be deduced that the resolution of the individual constituents of the overall N deposition should be in the order of magnitude of 0.5 $\text{kg ha}^{-1} \text{a}^{-1} \text{N}$, if the contributions are in the same order of magnitude.

For bulk deposition of $\text{NO}_3\text{-N}$ this accuracy can be achieved without problems. If one compares sites with similar amounts of precipitation (such as D1, D4, D11, D19, D20, D29, D32, D39, D42, D43, D48 and D49, all with $700 \pm 20 \text{ mm a}^{-1}$ precipitation), the mean $\text{NO}_3\text{-N}$ bulk deposition amounts to $4.32 \pm 0.26 \text{ kg ha}^{-1} \text{a}^{-1} \text{N}$, with 0.26 $\text{kg ha}^{-1} \text{a}^{-1}$ being the standard deviation $\sigma_{\text{NO}_3\text{-N}}$. If one assumes the standard error $G_{\text{NO}_3\text{-N}}$ to be twice the standard deviation $\sigma_{\text{NO}_3\text{-N}}$, an accuracy of 0.5 $\text{kg ha}^{-1} \text{a}^{-1} \text{N}$ can be achieved for this species. It can also be stated for the group of sites mentioned above as well as from other similar combinations (D13 and D14; D15 and D16; D21, D22, D23) that $\text{NO}_3\text{-N}$ depositions can be applied satisfactorily to other sites with similar precipitation.

Previous investigations had shown that these figures can also be obtained for $\text{NH}_4\text{-N}$ if the number of replicates made allows for a rejection of fouled samples in particular. $\text{NH}_4\text{-N}$ deposition does to some extent correlate with precipitation. However the influence of local emissions is crucial. Therefore, considerations similar to those made for $\text{NO}_3\text{-N}$ have to be made for neighbouring sites with similar precipitation such as D15, D16 or D21, D22, D23. Here, the standard error is also $0.5 \text{ kg ha}^{-1} \text{ a}^{-1} \text{ N}$ but, application of results is restricted to very small areas (with similar amounts of precipitation and similar emission situations).

5 Future activities

The network used for this programme was closed in December 2004. However, sampling continues at a reduced scale: sites D6 (Groß-Gerau), D10 (Limburg), D21 (Kefenrod), D35 (Wasserkuppe), D44 (Breuna) and D49 (Herleshausen) will be operated at least until the end of 2007.

In the UK, measurements of total nitrogen in bulk samples showed that the amount of N analyzed exceeded that of the sum of $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ (Cape et al., 2001, 2004). The time pattern of the additional inputs appears to suggest a relation to the application of slurry and farmyard manures. It would thus be composed of organic nitrogen which must have been emitted as spray during the application procedure. The measurements in 2007 will have to include total N in order to find out whether this effect can be observed in Hesse as well.

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Appendix

Table A1

Annual total depositions and Richter's correction factor (precipitation in mm a⁻¹, depositions in kg ha⁻¹ a⁻¹).
12 months July 2001 to June 2002, January und February 2003.

		elevation (m a.s.l.)	corr. factor	precipi- tation	Ca	Cl	K	Mg	NH ₄ -N	NO ₃ -N	Na	SO ₄ -S (b)	SO ₄ -S (s)
D01	Viernheim	98	1.104	831	4.4	5.1	1.5	0.65	5.8	4.7	2.6	6.8	5.4
D02	Neckarsteinach	115	1.104	1223	6.0	7.2	1.8	0.80	6.5	5.3	3.6	6.7	6.4
D03	Lorch	443	1.135	747	3.4	3.8	1.5	0.53	5.9	4.0	2.7	3.9	3.7
D04	Eltville	248	1.086	714	3.6	4.0	1.4	0.60	4.6	3.6	2.2	4.0	3.2
D05	Wiesbaden	88	1.082	604	8.4	5.5	1.5	0.81	4.1	3.3	2.9	4.3	3.9
D06	Groß-Gerau	89	1.082	673	4.8	4.2	2.8	0.72	8.3	3.9	2.4	5.1	4.4
D07	Mühlthal	225	1.104	875	4.3	5.7	1.0	0.96	5.4	5.1	3.0	5.8	5.3
D08	Brombachtal	395	1.104	1069	3.0	7.1	1.2	0.67	6.6	6.0	3.2	5.6	5.1
D09	Erbach	364	1.082	1020	3.5	5.3	2.5	0.69	7.2	5.0	2.7	5.1	5.1
D10	Limburg	245	1.135	784	3.9	4.9	0.9	0.62	4.5	4.3	2.6	5.9	3.7
D11	Selters	362	1.086	882	4.3	5.3	1.6	0.73	4.4	4.5	3.1	5.1	4.1
D12	Kleiner Feldberg	822	1.135	1113	3.3	6.6	1.1	0.54	4.8	5.6	3.4	6.6	5.6
D13	Bad Vilbel	124	1.082	752	5.6	4.1	1.5	0.76	4.6	4.5	2.6	4.9	4.8
D14	Hanau	112	1.082	960	3.6	5.6	1.8	0.94	6.2	4.8	3.1	6.4	5.3
D15	Niddatal	184	1.082	744	9.6	6.1	3.5	1.22	7.6	4.3	3.7	5.9	5.2
D16	Reichelsheim	121	1.149	672	2.6	5.4	2.6	0.66	8.9	4.3	2.5	5.6	4.3
D17	Herborn	276	1.111	757	4.9	5.3	1.5	1.09	4.7	4.3	3.1	5.0	3.9
D18	Biebertal	203	1.086	690	2.7	3.8	0.9	0.52	3.7	4.2	2.3	3.9	3.4
D19	Linden	172	1.149	617	2.2	4.6	0.8	0.5	2.9	3.9	2.1	3.5	3.1
D20	Nidda	195	1.104	801	3.2	4.9	2.2	0.70	5.4	4.6	2.8	5.0	4.9
D21	Kefenrod	265	1.104	974	2.4	7.8	1.5	1.08	7.9	6.3	3.4	7.2	5.7
D22	Bad Soden-Salmünster	429	1.111	936	3.6	5.8	1.3	0.95	6.8	5.5	3.3	6.0	6.0
D23	Schotten	239	1.135	988	2.6	6.9	2.2	0.69	7.0	6.2	3.5	6.1	5.0
D24	Schotten	583	1.082	1151	3.5	7.7	1.5	0.67	7.5	7.1	3.5	6.5	5.8
D25	Ulrichstein	588	1.111	1155	3.6	7.6	2.0	0.77	8.0	6.5	3.6	6.5	5.7
D26	Lauterbach	373	1.086	891	3.4	5.3	1.2	0.50	4.4	4.8	3.0	4.8	4.0
D27	Hatzfeld (Eder)	424	1.086	940	2.1	6.6	1.7	0.60	4.6	4.8	3.7	5.1	4.7
D28	Cölbe	245	1.149	856	2.4	5.5	1.5	0.70	6.9	5.1	3.2	5.0	4.9
D29	Stadtallendorf	263	1.104	835	2.9	6.1	2.3	0.99	6.1	4.8	4.2	5.7	5.5
D30	Alsfield	292	1.086	861	2.9	5.0	1.6	0.55	5.8	5.2	2.6	5.4	4.0
D31	Wartenberg	288	1.086	713	7.7	4.9	1.8	0.87	7.1	4.4	2.7	4.6	4.1
D32	Fulda	380	1.086	774	3.1	4.2	0.9	0.45	5.3	5.1	2.3	5.4	4.5
D33	Gersfeld	399	1.111	965	2.6	6.7	1.4	0.50	6.7	5.3	3.4	5.9	5.5
D34	Gersfeld	453	1.086	1023	2.5	6.5	1.2	0.50	6.4	5.6	3.3	5.5	5.5
D35	Wasserkuppe	942	1.135	1104	3.0	6.8	0.9	0.52	6.4	7.1	3.5	6.7	5.8
D36	Hilders	582	1.086	1062	5.0	7.7	1.2	0.89	7.8	6.0	4.0	5.9	5.2
D37	Ehrenberg (Rhön)	660	1.166	1118	2.5	5.8	1.3	0.52	5.3	6.1	2.9	5.6	5.0
D38	Vöhl	348	1.086	658	5.4	5.7	1.5	0.81	4.2	3.2	3.0	3.2	3.6
D39	Edertal	196	1.111	794	3.0	5.6	1.9	0.70	8.1	4.5	3.1	4.6	4.0
D41	Knüllwald	352	1.111	1078	3.5	7.8	1.7	0.79	6.6	6.2	4.0	6.0	5.0
D42	Bad Hersfeld	201	1.111	787	2.2	5.7	0.8	0.38	5.5	4.7	2.6	4.3	3.4
D43	Hohenroda	349	1.166	899	2.8	7.1	2.3	0.47	6.5	5.5	3.6	5.5	4.9
D44	Breuna	232	1.111	902	4.4	10.8	2.2	1.10	11.0	5.7	4.7	7.5	6.0
D45	Calden	265	1.086	783	5.0	7.9	4.5	0.76	8.4	4.8	4.3	6.4	5.3
D46	Kassel	145	1.086	847	3.2	5.9	2.7	0.67	6.0	4.7	3.7	5.4	5.2
D47	Grossalmerode	594	1.086	986	2.6	7.5	1.4	0.58	5.3	5.9	3.4	6.2	5.5
D48	Wehretal	175	1.086	866	2.6	6.0	1.8	0.52	7.1	5.3	3.3	5.6	5.0
D49	Herleshausen	314	1.086	887	4.0	6.6	1.9	0.38	4.4	5.6	3.4	5.5	4.8
D50	Wahlsburg	107	1.111	776	2.0	8.3	1.7	0.63	5.5	4.4	4.3	5.0	4.9

Table A2

Annual total depositions (precipitation in mm a⁻¹, depositions in kg ha⁻¹ a⁻¹).
12 months, March 2002 to February 2003. Richter's correction factor as in Table A1

		elevation (m a.s.l.)	precipitation	Ca	Cl	K	Mg	NH ₄ -N	NO ₃ -N	Na	SO ₄ -S (b)	SO ₄ -S (s)
D01	Viernheim	98	891	5.2	3.5	2.0	0.70	6.8	5.2	2.3	6.2	5.4
D02	Neckarsteinach	115	1095	4.5	3.8	1.9	0.65	6.0	5.1	2.5	6.1	5.7
D03	Lorch	443	795	3.0	3.5	1.6	0.45	5.3	4.5	2.6	4.2	3.8
D04	Eltville	248	714	3.6	4.0	1.4	0.60	4.6	3.6	2.2	4.0	3.2
D05	Wiesbaden	88	640	9.0	5.1	1.7	0.80	5.0	3.8	3.1	4.7	4.4
D06	Groß-Gerau	89	698	4.5	3.3	1.9	0.61	6.2	4.3	2.2	4.8	4.0
D07	Mühlthal	225	999	3.3	4.1	1.3	0.85	4.9	5.5	2.6	6.1	5.0
D08	Brombachtal	395	859	2.6	4.5	1.2	0.51	5.6	5.2	3.0	4.8	4.0
D09	Erbach	364	865	2.3	3.4	1.5	0.48	5.7	4.2	2.2	4.2	4.0
D10	Limburg	245	800	3.2	4.5	2.0	0.70	5.5	4.7	2.9	4.5	4.0
D11	Selters	362	851	3.0	4.5	1.5	0.60	3.7	4.8	2.6	4.7	3.9
D12	Kleiner Feldberg	822	983	3.4	4.6	1.2	0.49	5.6	5.4	2.1	6.0	5.8
D13	Bad Vilbel	124	847	4.8	4.0	1.6	0.70	4.7	4.9	2.7	5.4	4.7
D14	Hanau	112	841	2.9	3.7	1.2	0.60	4.3	4.4	2.3	4.9	3.8
D15	Niddatal	184	785	7.4	5.3	3.2	1.04	6.1	4.2	3.3	5.0	4.0
D16	Reichelsheim	121	699	2.7	5.2	2.0	0.63	9.3	4.2	2.6	5.5	4.3
D17	Herborn	276	898	3.7	5.4	0.9	0.93	4.4	4.9	3.4	5.1	4.4
D18	Biebertal	203	736	2.1	4.5	0.8	0.50	4.6	4.4	2.6	3.9	3.4
D19	Linden	172	891	2.6	5.7	1.1	0.58	4.7	4.9	2.7	5.2	3.9
D20	Nidda	195	815	2.6	3.7	2.4	0.62	4.9	4.6	2.6	4.7	4.1
D21	Kefenrod	265	791	2.3	4.1	1.5	0.98	5.3	4.7	2.4	4.5	3.8
D22	Bad Soden-Salmünster	429	959	4.0	4.5	1.9	0.69	6.4	5.5	2.6	5.7	5.1
D23	Schotten	239	872	2.6	3.9	2.6	0.69	8.1	5.1	2.8	5.0	4.5
D24	Schotten	583	1048	3.7	5.7	2.1	0.67	8.3	5.8	2.9	6.6	4.4
D25	Ulrichstein	588	1049	4.5	5.1	1.6	0.72	7.3	5.9	3.0	5.6	5.2
D26	Lauterbach	373	932	3.9	4.3	1.5	0.61	5.2	4.8	2.7	4.9	4.1
D27	Hatzfeld (Eder)	424	922	2.5	3.9	1.0	0.51	4.4	4.6	2.6	4.6	3.8
D28	Cölbe	245	864	3.0	4.6	1.7	0.68	6.5	5.1	2.8	5.4	4.6
D29	Stadtallendorf	263	864	3.1	7.0	2.4	1.09	6.4	5.2	4.7	6.1	6.0
D30	Alsfeld	292	962	2.6	5.4	2.1	0.55	7.2	5.1	2.8	6.1	4.3
D31	Wartenberg	288	838	7.6	4.1	2.6	0.98	8.2	4.6	2.6	4.9	4.6
D32	Fulda	380	834	3.0	3.6	0.9	0.49	5.7	4.9	2.2	5.2	4.4
D33	Gersfeld	399	897	2.5	4.8	1.2	0.45	7.6	5.2	2.5	5.8	4.6
D34	Gersfeld	453	908	2.8	4.1	1.1	0.45	5.5	5.3	2.8	5.2	4.4
D35	Wasserkuppe	942	993	3.3	4.1	0.8	0.48	5.2	6.2	2.7	5.6	4.8
D36	Hilders	582	918	4.9	4.7	1.1	0.80	6.9	5.5	2.9	5.2	4.4
D37	Ehrenberg (Rhön)	660	987	2.7	3.7	1.3	0.46	5.2	5.9	2.3	5.6	5.0
D38	Vöhl	348	757	5.0	6.0	1.1	0.69	4.6	3.6	2.9	4.1	4.1
D39	Edertal	196	861	2.8	4.7	1.9	0.57	7.8	4.6	2.6	5.1	4.7
D41	Knüllwald	352	990	2.6	6.0	1.5	0.51	6.0	5.7	2.9	5.8	5.0
D42	Bad Hersfeld	201	814	2.3	4.0	0.8	0.42	5.8	4.4	2.6	4.3	3.9
D43	Hohenroda	349	1007	3.7	5.6	2.5	0.58	6.3	5.5	3.1	6.2	5.6
D44	Breuna	232	885	3.4	8.9	2.1	0.68	10.1	5.6	3.8	8.4	5.4
D45	Calden	265	860		5.9	2.6		6.1	5.3	4.3	8.2	7.3
D46	Kassel	145	869	3.6	5.4	2.4	0.65	6.7	4.9	3.4	5.9	5.3
D47	Grossalmerode	594	1029	3.2	5.1	1.5	0.56	6.4	6.9	2.9	7.1	6.2
D48	Wehretal	175	852	3.4	4.6	1.7	0.59	7.4	5.0	2.8	5.7	5.5
D49	Herleshausen	314	846	5.1	3.9	1.8	0.51	5.3	5.1	2.7	5.3	4.7
D50	Wahlsburg	107	962	2.6	6.6	1.1	0.58	5.7	5.1	4.2	5.7	5.8

Table A3

Annual total depositions (precipitation in mm a⁻¹, depositions in kg ha⁻¹ a⁻¹).
12 months January to December 2003. Richter's correction factor as in Table A1

		elevation (m a.s.l.)	precipitation	Ca	Cl	K	Mg	NH ₄ -N	NO ₃ -N	Na	SO ₄ -S (b)	SO ₄ -S (s)
D01	Viernheim	98	561	4.6	3.3	1.4	0.63	5.0	3.9	2.6	5.1	3.8
D02	Neckarsteinach	115	823	5.3	3.9	1.6	0.81	5.5	4.3	3.2	5.2	4.6
D03	Lorch	443	526	3.5	4.5	1.1	0.53	4.4	4.3	3.1	4.1	3.5
D04	Eltville	248	714	4.2	4.0	2.0	0.57	4.3	3.6	2.5	3.8	3.4
D05	Wiesbaden	88	451	12.7	5.2		0.87	5.5	3.2	3.1	4.7	3.9
D06	Groß-Gerau	89	463	5.2	3.1	2.5	0.60	4.8	3.6	2.2	4.4	3.7
D07	Mühltal	225	637	5.3	3.5	2.3	0.75	5.4	5.0	2.4	5.0	3.9
D08	Brombachtal	395	776	4.2	4.8	2.6	0.71	5.9	5.2	3.2	5.1	3.9
D09	Erbach	364	638	2.8	3.6	1.6	0.51	5.4	3.9	2.3	4.1	3.3
D10	Limburg	245	636	4.0	4.6	1.0	0.55	5.5	4.6	2.4	4.5	3.3
D11	Selters	362	584	3.3	4.0	1.9	0.54	3.8	3.7	2.3	3.9	3.0
D12	Kleiner Feldberg	822	905	5.1	5.6	1.2	0.68	5.1	6.0	3.0	6.4	4.9
D13	Bad Vilbel	124	532	7.2	3.7	2.1	0.76	5.0	3.9	2.3	4.6	4.1
D14	Hanau	112	557	3.1	3.3	1.6	0.71	4.7	3.9	2.5	4.7	3.5
D15	Niddatal	184	490	7.9	4.8	3.0	0.98	7.0	4.1	2.6	5.1	3.8
D16	Reichelsheim	121	514	2.9	5.4	1.5	0.45	3.7	4.5	2.3	6.6	2.8
D17	Herborn	276	750	4.7	5.0	1.1	1.22	5.1	5.5	2.8	5.9	4.0
D18	Biebertal	203	600	3.4	3.5	1.0	0.55	3.5	4.5	3.1	4.3	3.2
D19	Linden	172	482	3.9	4.1	1.6	0.8	4.7	3.7	2.6	4.0	4.1
D20	Nidda	195	523	3.4	3.3	2.7	0.71	3.8	4.5	2.1	4.8	3.3
D21	Kefenrod	265	649	2.9	5.9	1.6	0.93	6.6	6.0	2.8	6.2	4.1
D22	Bad Soden-Salmünster	429	676	4.1	5.4	1.1	0.57	5.0	5.3	2.9	5.0	3.5
D23	Schotten	239	641	3.0	4.3	1.1	0.45	5.0	4.8	2.6	5.6	3.6
D24	Schotten	583	770	3.4	4.2	1.1	0.51	6.8	5.6	2.9	6.0	4.6
D25	Ulrichstein	588	795	4.6	4.7	1.4	0.72	6.3	5.7	3.1	5.5	4.8
D26	Lauterbach	373	575	4.0	3.5	1.0	0.46	3.8	3.9	2.3	3.6	3.0
D27	Hatzfeld (Eder)	424	758	4.3	4.8	1.1	0.62	5.4	4.8	3.1	4.7	4.7
D28	Cölbe	245	583	4.6	3.2	2.3	0.77	6.0	4.2	2.6	4.2	4.5
D29	Stadtallendorf	263	588	2.2	4.0	1.1	0.42	3.2	4.3	2.6	5.3	2.7
D30	Alsfeld	292	625	3.2	3.6	1.4	0.53	6.1	4.1	2.7	4.2	4.1
D31	Wartenberg	288	483	8.5	4.0	2.0	0.75	5.7	4.0	2.3	4.6	3.2
D32	Fulda	380	518	4.2	4.4	1.1	0.60	6.3	4.2	3.1	4.2	3.9
D33	Gersfeld	399	766	5.1	8.1	3.0	1.36	5.8	5.3	4.8	5.6	4.7
D34	Gersfeld	453	770	8.9	8.3	1.6	1.71	6.2	5.5	4.7	5.2	4.8
D35	Wasserkuppe	942	787	4.3	4.0	1.0	0.61	8.2	6.8	3.0	5.6	5.1
D36	Hilders	582	752	3.0	7.0	1.6	0.57	8.3	5.2	3.7	5.9	3.8
D37	Ehrenberg (Rhön)	660	893	4.1	3.6	1.3	0.58	6.5	5.8	2.9	5.2	5.1
D38	Vöhl	348	559	5.0	6.0	2.4	0.67	3.7	3.9	3.7	4.0	3.7
D39	Edertal	196	574	3.2	4.9	1.6	0.56	7.3	4.1	2.6	4.6	3.5
D41	Knüllwald	352	618	3.5	5.1	1.3	0.50	4.9	4.2	3.1	3.9	3.4
D42	Bad Hersfeld	201	636	2.9	5.1	1.2	0.47	4.6	4.2	3.1	4.0	3.6
D43	Hohenroda	349	606	4.1	5.1	2.0	0.64	5.4	4.1	3.0	3.9	3.6
D44	Breuna	232	596	4.4	5.1	1.3	0.55	9.0	4.7	3.0	5.4	4.2
D45	Calden	265	490	6.0	4.1	1.6	0.96	4.3	3.6	2.9	3.7	3.6
D46	Kassel	145	587	3.4	5.2	1.1	0.54	3.8	4.0	3.0	4.9	3.7
D47	Grossalmerode	594	651	4.1	6.2	1.2	0.81	5.2	5.2	4.3	5.3	4.8
D48	Wehretal	175	533	3.8	3.9	1.6	0.58	5.0	3.7	2.9	3.8	3.9
D49	Herleshausen	314	607	4.3	4.6	1.1	0.59	4.0	4.2	3.2	3.7	3.2
D50	Wahlsburg	107	763	2.7	7.5		1.25		4.8	4.6	5.3	4.7

Table A5

Mean annual total deposition (precipitation in mm a^{-1} , depositions in $\text{kg ha}^{-1} \text{ a}^{-1}$).
2002 to 2004. Richter's correction factor as in Table A1

		Höhe (m)	Nds (mm)	Ca	Cl	K	Mg	NH ₄ -N	NO ₃ -N	Na	SO ₄ -S (b)	SO ₄ -S (s)
D01	Viernheim	98	708	4.6	4.0	1.5	0.60	5.5	4.6	3.0	5.7	4.4
D02	Neckarsteinach	115	925	5.8	5.4	1.9	0.76	6.3	5.0	4.0	6.0	5.3
D03	Lorch	443	641	3.2	5.1	1.4	0.52	4.6	4.5	3.7	4.3	3.7
D04	Eltville	248	694	3.7	4.8	1.8	0.57	4.2	3.9	3.2	4.1	3.5
D05	Wiesbaden	88	507	10.7	5.8	2.1	0.82	4.7	3.4	3.6	4.7	4.1
D06	Groß-Gerau	89	562	4.8	3.9	1.9	0.58	5.3	3.9	2.9	4.6	3.9
D07	Mühlthal	225	790	3.9	4.3	1.5	0.69	4.9	5.1	2.9	5.3	4.4
D08	Brombachtal	395	828	3.4	5.5	1.8	0.56	5.6	5.2	4.4	5.0	4.1
D09	Erbach	364	785	2.8	4.3	1.5	0.52	6.0	4.3	3.1	4.4	4.0
D10	Limburg	245	731	3.7	5.4	1.6	0.64	5.8	4.5	3.8	4.6	3.9
D11	Selters	362	711	3.3	4.8	1.6	0.64	3.6	4.0	2.9	4.2	3.3
D12	Kleiner Feldberg	822	922	3.8	5.3	1.2	0.55	5.2	5.5	3.6	5.9	5.1
D13	Bad Vilbel	124	669	5.3	4.7	1.7	0.67	4.5	4.0	3.3	4.8	4.1
D14	Hanau	112	695	3.2	3.8	1.8	0.70	5.4	3.9	3.0	4.5	3.7
D15	Niddatal	184	632	7.6	5.5	3.4	1.07	6.3	4.1	3.6	5.0	4.0
D16	Reichelsheim	121	619	4.1	5.1	2.1	0.72	6.5	4.3	3.0	5.7	3.8
D17	Herborn	276	880	3.9	6.7	1.0	0.99	5.0	5.3	4.0	5.6	4.3
D18	Biebertal	203	728	2.8	5.4	0.9	0.53	4.1	4.6	2.9	4.5	3.3
D19	Linden	172	689	3.7	5.5	1.9	0.81	4.9	4.3	3.2	4.8	4.0
D20	Nidda	195	702	3.2	3.9	2.2	0.59	4.1	4.5	3.1	4.7	3.8
D21	Kefenrod	265	813	3.2	5.6	1.8	0.84	6.6	5.7	3.1	6.0	4.2
D22	Bad Soden-Salmünster	429	811	3.7	5.5	1.4	0.57	5.7	5.3	3.1	5.4	4.2
D23	Schotten	239	813	2.8	4.6	1.7	0.55	6.2	5.1	3.2	5.3	4.3
D24	Schotten	583	912	3.5	5.8	1.6	0.58	7.0	5.5	3.9	6.1	4.6
D25	Ulrichstein	588	976	5.3	5.6	1.5	0.69	6.5	5.8	4.0	5.8	5.1
D26	Lauterbach	373	781	4.1	4.5	1.5	0.64	4.2	4.3	3.4	4.2	3.5
D27	Hatzfeld (Eder)	424	923	3.7	6.5	1.2	0.63	5.1	5.1	4.5	5.3	4.8
D28	Cölbe	245	743	4.2	5.2	2.0	0.70	6.4	4.8	4.4	5.5	4.6
D29	Stadtallendorf	263	715	3.1	6.1	1.6	0.72	5.2	4.6	4.2	5.5	4.3
D30	Alsfeld	292	794	3.0	5.5	1.8	0.57	7.3	4.5	3.5	5.2	4.2
D31	Wartenberg	288	640	8.7	4.6	2.2	0.78	6.5	4.2	3.1	4.7	3.9
D32	Fulda	380	681	3.8	4.2	1.0	0.50	5.6	4.5	3.2	4.6	4.1
D33	Gersfeld	399	902	4.2	7.4	1.9	0.85	6.2	5.5	4.6	6.0	4.8
D34	Gersfeld	453	882	5.0	6.3	1.3	0.89	5.3	5.5	4.0	5.2	4.5
D35	Wasserkuppe	942	924	4.0	5.9	1.1	0.58	6.7	6.7	4.2	5.9	5.3
D36	Hilders	582	888	3.7	7.1	1.3	0.62	7.3	5.3	4.2	6.0	4.2
D37	Ehrenberg (Rhön)	660	965	3.4	4.5	1.3	0.50	5.6	5.6	3.4	5.4	4.9
D38	Vöhl	348	659	5.5	6.1	1.7	0.65	4.1	3.8	3.6	4.1	3.8
D39	Edertal	196	694	2.8	5.7	1.5	0.52	6.4	4.1	3.0	4.9	3.7
D41	Knüllwald	352	778	2.8	5.9	1.5	0.49	4.8	4.7	3.5	4.6	4.2
D42	Bad Hersfeld	201	703	3.1	4.9	1.1	0.45	5.5	4.3	3.3	4.1	3.7
D43	Hohenroda	349	805	4.0	6.1	2.2	0.72	5.9	4.6	3.8	4.8	4.4
D44	Breuna	232	718	4.3	8.1	1.8	0.67	9.3	4.9	4.8	6.3	4.8
D45	Calden	265	652	5.8	5.9	2.0	1.01	5.2	4.2	4.3	5.6	5.1
D46	Kassel	145	735	4.0	7.3	1.9	0.66	5.2	4.4	4.5	6.3	4.6
D47	Grossalmerode	594	835	3.3	6.9	1.5	0.62	5.4	5.5	4.2	5.8	5.1
D48	Wehretal	175	722	3.9	4.8	1.7	0.60	5.9	4.5	3.6	4.7	4.6
D49	Herleshausen	314	705	4.8	4.9	1.6	0.53	4.5	4.5	3.6	4.3	3.8
D50	Wahlsburg	107	863	4.3	7.0	1.5	0.92	5.5	4.9	4.4	5.5	5.3