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Abstract: A survey of 78 lakes and streams in 9 sensitive regions of the Federal Republic of Germany and 1 in western France was conducted in March-April 1983 and December 1981, respectively. The waters show high to extreme acidification. Acidification correlates well with the regional deposition of sulfur from the atmosphere. In areas east of the Rhein-Ruhr industrial district acidification levels rank among the highest reported in the world. In all regions the chemical composition of one or more water samples indicates that adverse biological effects might occur.

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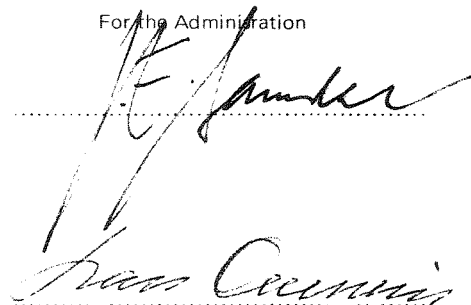


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REGIONAL SURVEY OF FRESHWATER
ACIDIFICATION IN WEST GERMANY (FRG)

by

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PREFACE

This regional survey of freshwaters in the Federal Republic of Germany is part of a continuing effort by the acid rain research group at NIVA to map acidified and non-acidified softwater regions of Europe and compare the situation with results from regional surveys in Norway. The data reported here comprise a set of 9 samples collected in December 1981 in the Vosges mountains of France and 69 samples collected by a number of individuals in March-April 1983 in the FRG. All samples were analyzed at NIVA's laboratory. NIVA's contribution to these surveys was funded by the Norwegian Ministry of the Environment.

This study was carried out with the kind cooperation of the following individuals and their institutes: R. Buerk at Bruchsal, M. Hauhs at Goettingen, W. Meinel at Kassel, and K. Thiele at Grafenau. We also acknowledge the assistance of our institutes in Hohenheim, Oslo and Mainz, respectively.

SUMMARY

In recent years several independent reports of anthropogenic acidification of surface waters in the Federal Republic of Germany (FRG) have appeared (Hecker 1982, Krebs 1979, Krieter 1983, Mattias 1982). To put these studies into a regional frame and to obtain an overview of the extent of acidification in the FRG water samples were collected from softwater streams and lakes not receiving wastewater. The samples were collected in cooperation with several individuals and institutions in the FRG and sent to the Norwegian Institute for Water Research (NIVA) in Oslo for analysis.

The results documented high to extreme acidification and high levels of sulfate and nitrate in all the softwater areas sampled. The degree of acidification correlates well with the regional deposition of sulfur from the atmosphere. In areas east of the Rhein-Ruhr industrial district acidification of surface waters reaches levels as high as and higher than those reported from other regions of the world receiving acid deposition from the atmosphere. In all regions the chemical composition of one or more water samples indicates that adverse biological effects might occur.

1. INTRODUCTION

In recent years several independent reports of anthropogenic acidification of surface waters in the Federal Republic of Germany have appeared (Hecker 1982, Krebs 1979, Krieter 1983, Mattias 1982). To put these studies into a regional frame and to obtain an overview of the extent of acidification in the FRG a synoptic survey of lakes and streams in 9 separate areas was organized with the cooperation of individuals and institutions in the FRG. The water samples were collected in the spring 1983 and sent to the Norwegian Institute for Water Research for chemical analysis.

2. METHODS

All the water samples come from softwaters. The catchment areas are mostly forested; the influence of agricultural fertilizers and industrial or municipal wastewaters is minimal. Sampling was conducted during the last phases of snowmelt. None of the sites has significant primary source of geologically-derived sulfate in the catchment. The influence of road-salts cannot be excluded in all cases, but the effect on the acidification measured in the streams and lakes is negligible. The samples were analyzed for pH, conductivity, Ca, Mg, Na, K, total-Al, NH_4 , NO_3 , SO_4 , Cl, HCO_3 , and total organic carbon (TOC) according to methods in routine use at NIVA. (See Wright and Henriksen 1978 and Henriksen 1983 for analytical details.) A total of 69 samples were analyzed. The analytical data and sample sites are listed in Appendix 1.

3. RESULTS

The degree of acidification expressed as the sum of sulfate and nitrate concentrations in $\mu\text{eq/l}$ is shown as group averages in Figure 1. These values are approximately the

same as those obtained from loss of alkalinity calculated using the procedure given by Wright (1983). The regional pattern of total sulfur deposition from the atmosphere (OECD 1976) in the FRG is clearly reflected in the observed levels of acidification in each of these 9 areas surveyed. Acidification is greatest in the forested regions of the northern part of the Mittelgebirge represented here by the data from the Kaufunger forest and the Knuell hills. Both these lie on sandstones and are downwind from the major emission sources in the industrial Rhein-Ruhr area. Acidification is also high in the central part of the Mittelgebirge represented here by the streams on the quartzites at Taunus and Hunsrueck and the Odenwald (on sandstone). In Taunus and Hunsrueck 2 samples of groundwater contained high concentrations of sulfate with Ca the major cation.

Regions of the FRG such as the Black Forest and the Bavarian Forest from which heavy damage to forests has been reported do not exhibit the highest levels of acidification of surface waters in our study. Although the pH levels in the waters sampled are as low as 4.0, the concentrations of the strong-acid anions sulfate and nitrate are about 250 $\mu\text{eq/l}$ as compared to the levels above 1000 $\mu\text{eq/l}$ observed in streams farther north.

Nitrate comprises a rather high fraction (10-38 %) of the sum of strong-acid anions. The fraction is highest in areas with the lowest acidification, while in the areas with greatest acidification nitrate comprises less than 11 %. In Scandinavia and acidified areas of North America nitrate levels are usually less than 10 % of the sum of nitrate plus sulfate (Wright 1983).

The estimated regional pattern of sulfur deposition (wet and dry) is also shown in Figure 1 (OECD 1976). The central part of the FRG receives an annual deposit of

5-10 g S/m²/yr, while in the Rhein-Ruhr area and to the east deposition rates exceed 10 g S/m²/yr. The southern Mittelgebirge receive less atmospheric sulfur (2.5-5 g S/m²/yr. The estimated total S deposition pattern matches well our observed levels of acidification in surface waters.

The acidification levels observed in the FRG are among the highest reported in the world. In southernmost Norway in lakes and streams in which acidification has eliminated thousands of fish populations acidification levels reach about 200 µeq/l (Wright 1983). This level is about the same as measured in the samples from the northern part of the Black Forest. In lakes and streams on the Swedish west coast acidification levels are somewhat higher, 200-300 µeq/l, about the same level as the Odenwald samples. And lakes very near to the large sulfur emission source at Sudbury, Ontario, Canada, show about 400-500 µeq/l sulfate plus nitrate (Dillon et al. 1980). Our samples suggest that acidification in the FRG reaches twice that level.

One or more samples from each of the areas had low pH level (below 5) and correspondingly high level of total Al (Fig. 2). Both these parameters are of importance for salmonid fishes, insects and other aquatic organisms. Experiments with brook trout conducted by Baker and Schofield (1982) indicate a pH-Al region that is toxic (Fig. 2). Water samples from nearly every area sampled fall into this toxic region.

4. DISCUSSION

The surface waters in forested areas of the FRG are generally thought to be unpolluted and of high quality. Forest streams are considered to be the last pristine aquatic biotope. Our survey shows that many of these

waters have pH and Al levels far exceeding the toxic level for salmonid fish and many other organisms (Fig. 2). Many fish food organisms are even more sensitive. Damage to these ecosystems may be extensive.

This survey is only a first look at regional acidification of surface waters in the FRG. Clearly more extensive surveys are necessary to fully assess the extent of freshwater acidification in the FRG. The chemical composition of these forest streams provide information as to the ability of the forests themselves to withstand the inputs of strong acids from the atmosphere (Krieter 1983). The stream water also gives insights into the health and stability of the forest ecosystem and in particular the soils and may even provide clues about future trends.

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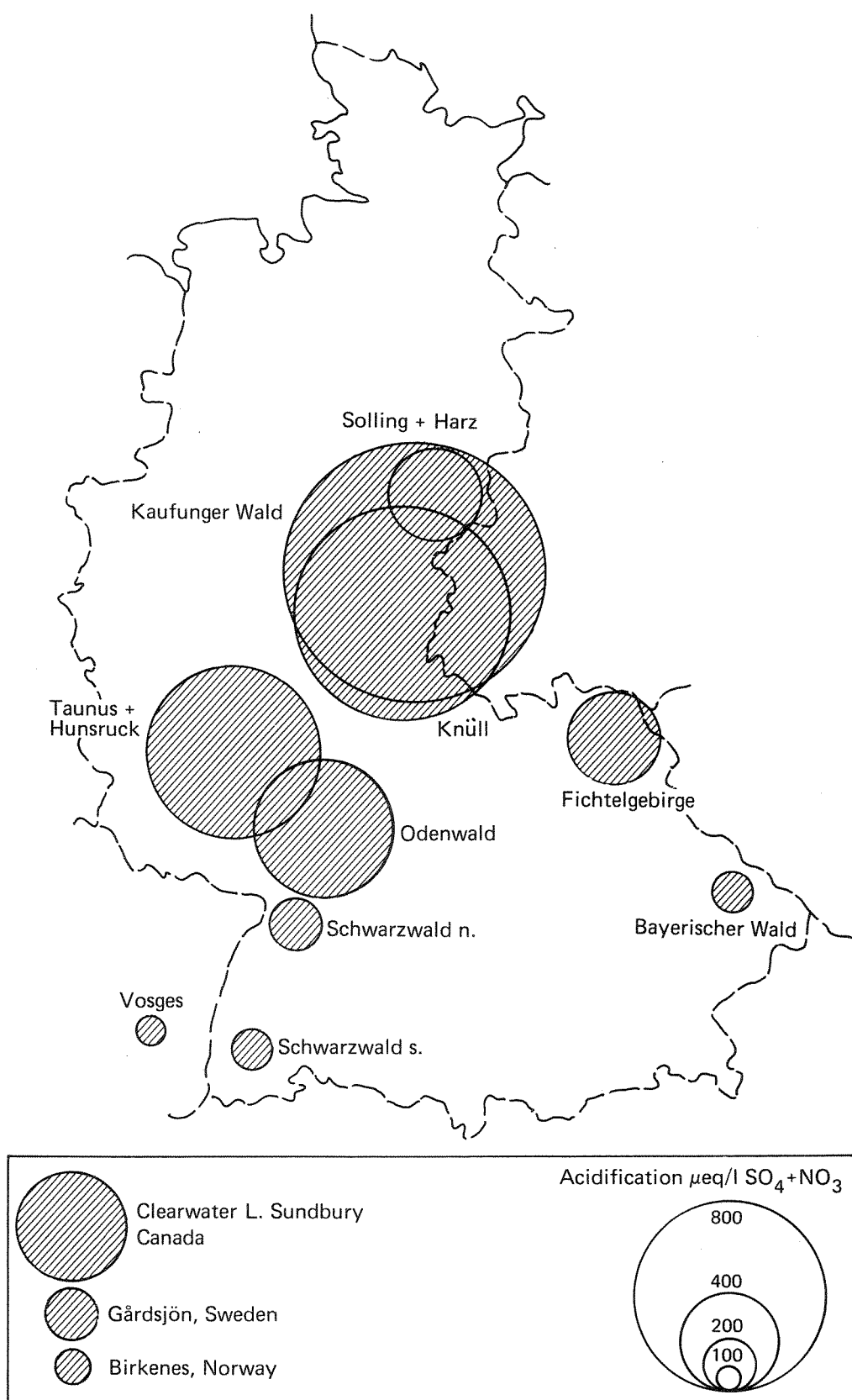


Fig. 1. Surface water acidification and total sulfur deposition 1983. The circles depict mean levels of the sum of sulfate plus nitrate in stream and lake samples collected in each area. The heavy lines indicate the total S deposition (wet and dry) estimated by OECD (1976).

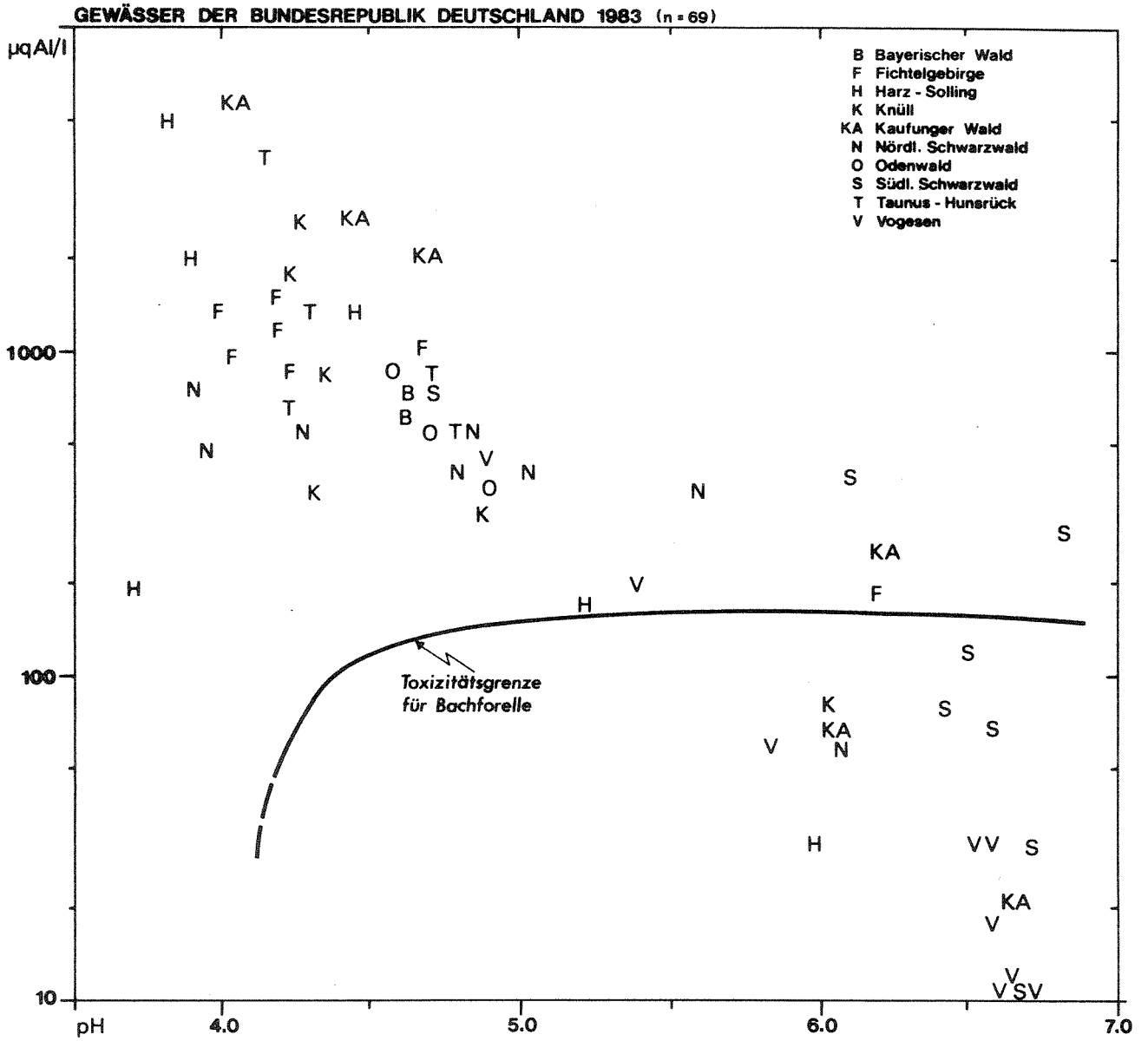


Fig. 2. Total Al ($\mu\text{g/l}$) levels and pH measured in the surface water samples. Also shown is the critical level for toxicity to brook trout as determined by Baker and Schofield (1982).

APPENDIX 1. Major-ion chemistry of lakes and streams in
BDR and southwestern France. Units:
Units: $\mu\text{eq/l}$ except Al ($\mu\text{g/l}$) and TOC (mg/l).
COND = conductivity mS/m at 25°C .

Station codes (STNUM)

BAYERW	Bayerischer Wald
FICHTE	Fichtelgebirge
KAUF.W	Kaufunger Wald
KNUELL	Knüll
N.SCHW	Nord Schwarzwald
ODENWA	Odenwald
SO-HAR	Solling-Harz
S.SCHW	Süd Schwarzwald
TA-HUN	Taunus-Hunsrück

SINUM	LOK	A M D R N G	NAVN	PH	COND	ECA	EMG	ENA	EK	ENH4	EN03	ECL	ES04	ALK-E	AL	I0C
BAYERW	1	830410	GR-OHE	4.62	3.67	80.3	42.8	27.0	12.3	.7	72.1	16.9	124.9		740.	5.1
BAYERW	1	830411	GR-OHE	4.63	3.55	76.8	41.1	28.3	10.7	1.7	80.7	16.9	124.9		660.	3.5
FICHTE	1	830323	MAHRIN	6.35	13.80	475.0	410.5	162.7	40.9	1.4	228.5	180.5	707.9	01.9	150.	2.2
FICHTE	2	830323	EOER	6.19	7.53	239.5	124.2	224.0	39.4	7.7	85.0	265.2	166.6	67.1	180.	4.0
FICHTE	3	830323	F.ZINN	4.65	8.03	147.7	56.8	259.3	32.7	7.1	100.0	242.6	291.5		900.	3.9
FICHTE	4	830323	M.WAIN	4.20	11.50	156.7	53.5	259.3	32.7	7.1	121.4	332.9	333.1		1400.	4.3
FICHTE	5	830323	F.NAAB	4.19	7.48	129.7	42.0	68.3	28.9	7.7	80.0	45.1	291.5		800.	6.6
FICHTE	6	830323	F.SEE	4.22	12.20	163.7	50.2	482.8	24.5	15.7	52.1	445.7	291.5		1200.	6.8
FICHTE	7	830323	KRAITZ	3.97	16.90	193.6	62.5	643.8	27.1	1.4	83.5	67.0	353.9		1000.	10.0
FICHTE	8	830323	MOOSKR	4.01	13.70	137.2	56.8	495.9	28.1	7.7	40.7	488.0	291.5		240.	4.2
KAUF-W	1	830321	STEINB	6.19	15.30	537.0	420.3	258.4	16.1	7.7	128.5	349.8	957.5		10.	3.0
KAUF-W	2	830321	KALITW	6.80	26.80	928.1	890.2	639.4	20.7	7.7	94.2	519.1	1707.2		120.	2.8
KAUF-W	3	830321	OTHERS	7.45	48.70	2874.2	896.6	752.5	26.3	1.4	192.8	1241.2	2498.4		1800.	4.4
KAUF-W	4	830321	ENDSCH	4.55	13.90	453.6	287.1	147.5	40.1	7.7	85.7	163.6	1020.2		70.	2.1
KAUF-W	5	830321	NIESTE	6.01	14.20	588.8	381.7	150.5	48.6	7.7	107.1	138.2	1041.0		2500.	4.6
KAUF-W	6	830321	INGELW	4.42	16.10	464.1	315.1	167.0	38.9	7.7	135.7	177.7	1041.0		5800.	7.7
KAUF-W	7	830321	SCHWAR	4.04	19.20	323.3	243.5	160.9	36.6	7.7	157.1	208.8	1145.1		20.	2.4
KAUF-W	8	830321	WENGEß	6.64	16.40	616.8	412.9	343.6	49.6	7.7	89.2	335.7	812.0		2500.	4.6
KNEJEL	1	830401	KNEJEL	4.28	13.10	260.5	192.5	102.7	33.0	7.7	7	112.8	874.4		850.	4.9
KNEJEL	2	830401	KNEJEL	4.33	8.45	221.6	64.2	61.3	40.1	7.7	90.7	81.8	437.2		335.	2.0
KNEJEL	3	830401	KNEJEL	4.87	14.70	598.8	344.7	141.4	47.3	7.7	90.7	149.5	895.3		350.	4.8
KNEJEL	4	830401	KNEJEL	4.30	16.40	350.3	436.0	225.8	89.0	7.7	149.9	239.8	832.8		1750.	6.4
KNEJEL	5	830401	KNEJEL	6.02	14.90	504.0	276.4	114.4	54.0	2.9	72.1	169.4	978.2		80.	2.4
KNEJEL	6	830401	KNEJEL	4.27	14.90	668.7	369.3	143.1	71.3	1.4	95.7	172.1	874.4		480.	16.8
N.SCHW	1	830323	O.DURR	3.93	7.49	121.3	56.0	24.4	18.2	1.4	48.6	56.4	166.6		540.	7.4
N.SCHW	2	830323	M.DURR	4.28	5.87	140.2	36.7	36.1	26.1	7.7	92.8	53.6	160.3		540.	5.9
N.SCHW	3	830323	N.DURR	4.79	4.65	168.2	51.8	33.9	27.4	7.7	85.7	56.4	181.1		710.	9.6
N.SCHW	4	830323	WILDSE	3.91	8.39	86.8	23.0	30.0	16.1	7.7	72.5	53.0	166.6		430.	5.0
N.SCHW	5	830323	BROTEN	4.78	4.27	145.2	49.4	34.4	26.8	7.7	64.3	50.8	170.7		430.	5.0
N.SCHW	6	830323	EYACHL	5.03	4.27	164.2	51.3	35.2	25.8	7.7	70.0	53.6	160.6		350.	4.7
N.SCHW	7	830323	EYACHM	5.57	4.14	181.6	62.5	36.1	27.9	7.7	65.0	53.6	183.2		60.	1.0
N.SCHW	8	830323	O.TROS	6.08	5.41	225.5	107.8	41.3	33.5	7.7	84.3	53.6	187.4		530.	1.4
OJENWA	1	830401	DUPRAZ	4.71	9.40	310.9	205.6	84.8	56.5	7.7	81.4	112.8	541.3		800.	1.7
OJENWA	2	830401	DURRA3	4.57	10.30	316.4	203.2	71.8	55.7	7.7	104.2	101.6	563.0		280.	2.1
OJENWA	3	830401	SCHAB1	5.95	11.20	442.1	209.8	171.4	82.8	4.3	157.1	234.1	583.0		350.	1.7
OJENWA	4	830401	SCHMB3	4.89	10.80	380.2	191.7	184.0	55.7	7.7	128.5	253.9	499.7		60.	1.9
OJENWA	5	830401	WESCC1	7.45	17.10	858.3	301.6	304.5	55.0	7.9	207.1	248.2	437.2		80.	2.9
OJENWA	6	830401	WESCC3	7.32	16.50	713.6	364.4	313.2	71.1	23.6	199.9	253.9	687.1		150.	7.9
SO-HAR	1	830329	SOLLIN	5.22	8.66	214.1	287.9	118.8	36.6	7.7	70.0	138.2	499.7		5100.	5.4
SO-HAR	2	830315	HILLS	3.82	28.70	178.6	411.3	269.7	47.0	2.1	399.8	332.9	1519.9		2000.	5.4
SO-HAR	3	830320	KELLER	4.47	6.95	182.1	79.8	140.9	25.1	7.7	199.9	189.0	478.9		1200.	1.6
SO-HAR	4	830320	LB-QUE	6.00	5.31	182.1	148.1	63.5	20.5	7.7	65.7	90.3	249.8		30.	9.9
SO-HAR	5	830320	LB-PES	6.00	10.90	66.4	28.8	47.4	10.2	5.7	81.4	73.3	208.2		190.	10.9
SO-HAR	6	830320	ROFENB	3.72	10.90	286.4	89.0	142.7	30.2	7.7	18.6	149.5	149.9		260.	6.5
S.SCHW	1	830104	KIRN.U	6.84	4.27	170.2	46.1	111.8	28.4	7.7	20.7	118.5	143.7		410.	9.0
S.SCHW	2	830104	KIRN.S	4.69	5.24	117.3	45.2	154.0	16.1	7.7	6.4	174.9	164.5		720.	9.0
S.SCHW	3	830104	TITTI.I	6.49	4.95	206.6	44.4	153.1	17.9	7.7	35.0	129.8	124.9		30.	3.3
S.SCHW	4	830104	TITTI.J	6.70	6.04	251.0	61.7	193.1	18.4	7.7	35.0	183.4	139.5		10.	1.3
S.SCHW	5	830104	FELDU	6.68	1.93	93.3	24.7	40.0	4.3	7.7	17.1	11.3	50.0		65.	1.5
S.SCHW	6	830104	FELDI	6.59	1.67	82.3	24.7	37.8	3.8	7.7	17.1	11.3	50.0		80.	3.3
S.SCHW	7	830104	WINDFA	6.44	3.23	121.8	31.3	97.0	8.7	7.7	29.3	79.0	85.4		10.	2.0
S.SCHW	8	830104	MORGEN	6.91	25.60	1042.9	674.5	462.0	39.1	1.4	235.0	733.5	791.2		800.	6.2
TA-HUN	11	830411	SEITBER	4.70	13.90	369.3	345.5	194.0	44.5	7.7	71.4	197.5	832.8		1300.	7.1
TA-HUN	12	830411	GOLDST	4.30	11.00	182.1	153.0	123.5	38.9	7.7	44.3	146.7	624.6		580.	3.4
TA-HUN	30	830411	HANGWA	4.79	11.90	419.2	222.1	140.1	40.7	7.7	135.7	149.5	687.1		600.	6.7
TA-HUN	34	830411	KESSSEL	4.24	24.40	918.9	616.9	261.0	33.0	7.7	299.9	231.3	1457.4		3700.	4.3
TA-HUN	35	830411	GRUNDW	4.14	15.10	237.5	116.0	96.1	31.5	7.7	121.4	146.7	895.3		1003.3	1.5
TA-HUN	100	830411	GRUNDW	6.76	20.30	1097.8	419.5	269.7	32.0	7.7	96.4	400.6	395.6		1002.2	1.4
TA-HUN	110	830411	GRUNDW	6.68	20.10	1097.8	130.0	304.5	32.7	7.7	98.5	403.4	353.9		1002.2	1.4

VAIN	A M J	NAVN	PH	COND	ECA	EMG	ENA	EK	ENH4	EN03	FCL	ES04	ALK-E	AL
1	811222	CORBEA	4.91	2.90	60.4	24.7	52.6	9.5		52.1	39.5	102.0	.0	460.
2	811222	BLANC	6.76	3.63	162.7	51.8	80.9	12.0		28.6	64.9	108.3	106.5	10.
3	811222	LJPE	6.57	3.25	121.3	60.9	66.6	11.0		37.1	59.2	91.6	74.4	35.
4	811222	BLANCH	5.36	2.60	95.3	47.7	47.8	6.4		46.4	36.7	93.7	10.9	65.
5	811222	LONGEM	6.65	3.95	173.2	86.4	211.8	13.3		45.7	208.8	122.8	107.5	20.
6	811222	LISPAC	5.40	3.34	81.8	43.6	97.0	10.2		37.1	93.1	112.4	6.4	200.
7	811222	RETOUR	6.57	3.64	180.6	94.6	182.7	8.4		45.0	183.4	108.3	143.5	20.
8	811222	ROIR	6.76	3.65	159.2	50.2	78.7	11.0		29.3	62.1	118.7	96.1	10.
9	811222	LAUCH	6.76	3.77	162.2	67.8	93.7	9.5		32.1	50.8	75.0	186.6	10.