Use of sewage sludge – nitrogen availability and heavy metal uptake into rape

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Anwendung von Klärschlamm – Stickstoffverfügbarkeit und Schwermetallaufnahme durch Raps

1. Introduction

Since decades the application of sewage sludge on agricultural areas is under intensive discussion. Sewage sludge, a product of biological waste water treatment, can basically be used as a fertilizer or at least to improve the structure of the soil through additional supply of organic matter. Besides this generally agreed positive effect of sewage sludge applications to agricultural soils there is considerable concern about the accumulation of heavy metals and other pollutants and questions related to sanitary aspects (KIRCHMANN, 1994; KÖNIG and KRÄMER, 1985; SMITH, 1996). In this respect soil acts both as sink of pollutants and as source for the transfer of pollutants into the food chain. WILCKE and DÖHLER (1995) calculated that the limit values for Zn and Ni in agricultural soils of Germany will be reached within 600 to 800 years of continuos application of sewage sludge containing these heavy metals at the limit values for sewage sludge. Aiming at a sustainable soil management with respect to the use of organic wastes and residues it seems to be of high importance to gain additional information on i) the plant availability of nutrients

Zusammenfassung

Die Ergebnisse eines dreijährigen Gefäßversuches im Freiland zur Frage der Auswirkung von Klärschlamm (γ-sterilisiert und unbestrahlt) auf die Biomasseproduktion von Raps, die Schwermetall- und Stickstoffverfügbarkeit werden präsentiert.

Mobile Schwermetallfraktionen von Cd, Cu und Zn wurden signifikant durch die Klärschlammgaben erhöht, der Schwermetalltransfer in den Raps zeigte allerdings keine eindeutigen Ergebnisse.

Das Wachstum von Raps wurde im ersten und dritten Versuchsjahr positiv von den Klärschlammgaben beeinflußt. Die mittlere Ausnützung des Klärschlammstickstoffs sank von 7,4 % (1. Jahr), 1,8 % (2. Jahr) auf 1,1 % (3. Jahr), was einer Gesamtnutzung des Stickstoffs von 10,3 % entspricht. Klärschlammbestrahlung zeigte keinen signifikanten Einfluß auf die untersuchten Parameter.

Schlagworte: Klärschlamm, Raps, Schwermetalle, Stickstoff.

Summary

The results of a three years experiment with large pots in the field evaluating the effects of sewage sludge (sterilised by γ-irradiation or not sterilised) on rape growth, heavy metal- and N-uptake, using the ^{15}N-dilution technique, are presented.

Mobile fractions of Cd, Cu and Zn increased significantly in the substrate due to sewage sludge treatments. However, heavy metal transfer into rape plants did not respond clearly.

Rape growth was clearly enhanced in the first and third year due to sewage sludge applications. The average N-utilisation by rape from sewage sludge in a three years period decreased from 7.4 % (first year), 1.8 % (second year) to 1.1 % (third year), resulting in an overall utilisation of 10.3 % of sewage sludge – N_t by rape plants. Irradiation of sewage sludge did not result in any significant effect on the investigated parameters.

Key words: Heavy metals, nitrogen, rape, sewage sludge.

contained in sewage sludge ii) the mobility and accumulation of heavy metals introduced into soil and iii) the sanitary aspects of sewage sludge applications. The present paper attempts to contribute to some of these topics. The objectives were: i) to quantify the N-availability from sewage sludge under realistic climatic conditions, ii) to quantify the accumulation of heavy metals in soil fertilised with sewage sludge and their potential uptake into plants and iii) to evaluate the impact of γ-irradiation (sterilisation) on the nitrogen availability.

2. Materials and methods

2.1 Soil

The experiment was started in 1995 with large pots containing 30 kg of a soil/quartz sand substrate (1 : 1), which were sunk into the ground at level of the surrounding experimental field of the Austrian Research Centre Seibersdorf. The pots had a mean diameter of 42 cm and a height of 40 cm. The treatments were prepared with four replicates. The soil used in the experiment is a Chernozem from Seibersdorf. Chemical and physical parameters were determined according to BLUM et al. (1996) (Table 1).

The experimental soil has a quite high pH of 7.8, a high lime content (29 %) and a high soil organic matter content (4.9 %). Soil texture can be classified as loamy silt. The high organic matter content made the dilution of the soil with quartz sand necessary in order to amplify the effects.

2.2 Sewage sludge

The used sewage sludge originated from the sewage treatment plant of Mödling/Lower Austria. The main characteristics are presented in Table 1. The treatments in the experimental years 1995 and 1996 both originated from the same sewage treatment plant, but from different samplings. Therefore, the characteristics differ slightly (Table 1). The most important parameters, total nitrogen and organic matter contents were almost identical. The heavy metal concentrations did not exceed any limit value of the Lower Austrian regulations. The sewage sludge contained distinct amounts of pathogenic bacteria. For the experiment part of the sludge was sterilised by γ-irradiation in the ^{60}Co-irradiation plant of the Austrian Research Centre Seibersdorf. The applied radiation dose was 28.5 kGy. The application of an equivalent of 7.5 t sewage sludge per ha introduced considerable amounts of nitrogen (1995: 398 kg/ha, 1996: 407 kg/ha) and organic matter (1995: 4193 kg/ha, 1996: 4335 kg/ha) into the substrate. The amount applied is three times the portion, which may be legally used in agriculture in Austria, in order to amplify the effects.

2.3 Pot experiment

The treatments of the experiment in the year 1995 are shown in Table 2. On 8th of August air dry soil was mixed with the dried and ground sewage sludge, quartz sand and mineral nitrogen fertiliser in a concrete mixer. The pots received 30 kg of substrate. The experimental crop rape was

Table 1: Chemical and physical characteristics of the soil and the used sewage sludge
Tabelle 1: Chemische und physikalische Merkmale des Bodens und des verwendeten Klärschlammes

Parameter	soil	sewage sludge 1995	sewage sludge 1996	Parameter	soil	sewage sludge 1995	sewage sludge 1996
pH ($CaCl_2$)	7.78	6.06	6.24	CEC (meq/100g)	18.3	-	-
$CaCO_3$ (%)	29	2.6	4.8	N_t (%)	0.10	5.30	5.43
org. matter (%)	4.87	55.9	57.8	NH_4-N (mg/kg)	-	11900	2300
K_2O (%)	-	0.68	0.65	NO_3-N (mg/kg)	-	2500	< 100
K_2O (CAL, mg/kg)	230	3790	4290	Cd (mg/kg)	0.19	3.2	2.2
P_2O_5 (%)	-	6.26	5.6	Cr (mg/kg	14.3	70.6	37.3
P_2O_5 (CAL, mg/kg)	460	2660	460	Cu (mg/kg)	14.0	243	222
sand (%)	18	-	-	Hg (mg/kg)	0.042	1.7	1.3
silt (%)	58	-	-	Ni (mg/kg)	12.9	34.4	27.8
clay (%)	24	-	-	Pb (mg/kg)	16.0	106	79
EC (mS/cm)	-	3.93	4.34	Zn (mg/kg)	39.3	1160	880

All data are referred to dry matter. CAL: Calciumacetate-lactate extract, EC: electrical conductivity, CEC: cation exchange capacity sown on the 10th of August 1995 (20 kernels/pot) and the green rape plants were harvested on the 19th of September 1995. In order to test the N-availability in the sewage sludge in the second year, no additions were made to the treatments 1 to 3. To test a possible accumulative effect, irradiated sewage sludge was applied to treatment 5 (7.5 t/ha) and not irradiated sewage sludge to treatment 6 (7.5 t/ha) in the year 1996. $(^{15}NH_4)_2SO_4$ was added in the year 1996 to the treatments 4, 5 and 6. Rape was sown on 22nd of April 1996 (20 kernels/pot). The green rape was harvested on the 13th of August 1996. In the last experimental year (1997) no further sewage sludge was applied. All pots were treated with $(^{15}NH_4)_2SO_4$. To optimise the isotopic dilution method the N-application was split into two portions of 25 kg N/ha (25th of May and 12th of July 1997) and the solution was applied with a 50 ml syringe to avoid immediate gaseous losses. Rape was sown on 16th of May 1997 (20 kernels/pot) and rape was harvested on 28th of July 1997, 73 days after sowing.

2.4 Analysis

Rape biomass was weighed after cutting, dried, ground and then dried for 24 hours at 105° C. 2.00 g of the plant material were wet digested (20 ml HNO_3 and 4 ml $HClO_4$). Heavy metal contents were analysed by means of atomic absorption spectroscopy (Perkin Elmer) and ICP (Perkin

Table 2: Set-up of the pot experiment
Tabelle 2: Aufbau des Gefäßversuches

year 1995

Treatment (n = 4)	Sewage sludge (irradiated)	Sewage sludge (non irradiated)	$(^{15}NH_4)_2SO_4$ (4.820 at% ^{15}N excess)	$(NH_4)_2SO_4$
1	-	-	50 kg/ha	-
2	7.5 t/ha	-	50 kg/ha	-
3	-	7.5 t/ha	50 kg/ha	-
4	-	-	-	50 kg/ha
5	7.5 t/ha	-	-	50 kg/ha
6	-	7.5 t/ha	-	50 kg/ha

year 1996 (additional treatments)

Treatment (n = 4)	Sewage sludge (irradiated) [total 95 + 96]	Sewage sludge (non irradiated) [total 95 + 96]	$(^{15}NH_4)_2SO_4$ (4.820 at% ^{15}N excess)	$(NH_4)_2SO_4$
1	-	-	-	-
2	[7.5 t/ha]	-	-	-
3	-	[7.5 t/ha]	-	-
4	-	-	50 kg/ha	-
5	7.5 t/ha [15 t/ha]	-	50 kg/ha	-
6	-	7.5 t/ha [15 t/ha]	50 kg/ha	-

year 1997 (additional treatments)

Treatment (n = 4)	Sewage sludge (irradiated) [total 95 + 96]	Sewage sludge (non irradiated) [total 95 + 96]	$(^{15}NH_4)_2SO_4$ (4.820 at% ^{15}N excess)	$(NH_4)_2SO_4$
1	-	-	25 + 25 kg/ha	-
2	[7.5 t/ha]	-	25 + 25 kg/ha	-
3	-	[7.5 t/ha]	25 + 25 kg/ha	-
4	-	-	25 + 25 kg/ha	-
5	[15 t/ha]	-	25 + 25 kg/ha	-
6	-	[15 t/ha]	25 + 25 kg/ha	-

Elmer Plasma II). A part of the dried plant sample was transferred to the IAEA laboratories for nitrogen and ^{15}N analysis with a mass spectrometer (Micromass Sira 9) in combination with an elemental analyser (Carlo Erba 1500).

In the year 1995, 1996 and 1997 (autumn) composite substrate samples from the four replicates of each treatment were collected. The substrate samples were air dried and passed through a sieve (mesh size 2 x 2 mm). Subsamples were transferred to the IAEA laboratories for ^{15}N analyses. The heavy metal contents were analysed after wet digestion under reflux using aqua regia by means of AAS and ICP. Mobile fractions of Cd, Cr, Cu and Zn in soil were additionally determined in the year 1996 using 1M ammonium acetate (ECKER and HORAK, 1995; BLUM et al., 1996) as mild neutral extractant.

2.5 Calculations

Nitrogen uptake into rape derived from mineral fertiliser (Ndff) was calculated according to the isotopic dilution method:

$$\% \text{ Ndff} = [(\%^{15}\text{N a.e. rape}) / (\%^{15}\text{N a.e. fertiliser})] \times 100 \qquad (1)$$

The nitrogen fraction derived from unlabelled sewage sludge (Ndfss) was obtained by:

$$\% \text{ Ndfss} = [1 - (\%^{15}\text{N a.e. in rape treated with } (^{15}NH_4)_2SO_4 \text{ and sewage sludge}) / (\%^{15}\text{N a.e. in rape treated with } (^{15}NH_4)_2SO_4)] \times 100 \qquad (2)$$

Nitrogen derived from soil (Ndfs) was calculated as difference:

$$\% \text{ Ndfs} = 100 - \% \text{ Ndff} - \% \text{ Ndfss} \qquad (3)$$

Statistical analysis was performed using the programme WinStat 2.0.

3. Results and discussion

3.1 Soil

Table 3 shows the results of the aqua regia extractions of composite substrate samples collected in 1995. The heavy metal concentrations reached approximately 50 % of the initial values determined in the soil samples (Table 1), reflecting the 1 : 1 dilution with quartz sand. No distinct differences could be observed between mineral nitrogen treated pots (1, 4) and pots having received 7.5 t of sewage sludge per hectare (2, 3, 5, 6). No effect of the heavy metal input through the sewage sludge additions could be deduced. Therefore, it was decided to measure only the most relevant heavy metals in the year 1996, but to keep all replicates and to determine the mobile fractions. Table 4 shows the results of the aqua regia extraction of the substrate samples collected in autumn 1996.

Although we analysed 8 replicates per treatment no statistically significant differences between the treatments were observed for the heavy metals Cd, Cr, Cu and Zn. This result was not expected, because the amount of sewage sludge was high (up to 15 t of dry matter/ha) and the inputs of heavy metals were quite high compared to the original soil content. For example 0.64 mg Cd, 12.8 mg Cr, 55 mg Cu and 242 mg Zn were added to the pots receiving an equivalent of 15 t sewage sludge per hectare within two experimental years. These values represent 22 %, 6 %, 26 % and 41 % of the total substrate content for Cd, Cr, Cu and Zn, respectively. Substrate was analysed again in 1997 and the results confirmed the previous findings; a tendency to even smaller heavy metal values could be observed (Table 5).

The fact that the increase of heavy metal concentrations could not be detected may be due to two effects. The first is the high uncertainty of the heavy metal determination,

Table 3: Heavy metal contents of substrate samples (Chernozem : quartz sand = 1 : 1) after rape harvest in the year 1995
Tabelle 3: Schwermetallgehalte von Substratproben (Tschernosem : Quarzsand = 1 : 1) nach der Rapsernte im Jahr 1995

mg/kg dry matter

Treatment	Cd	Co	Cr	Cu	Hg	Ni	Pb	Zn
1	0.09	1.8	8.3	6.9	0.022	7.8	6.5	19.4
2	0.07	1.7	5.2	6.7	0.021	7.5	8.4	19.8
3	0.09	2.1	5.9	8.0	0.026	8.0	9.6	23.0
4	0.09	1.9	7.9	6.2	0.017	7.6	7.0	18.6
5	0.09	2.0	7.8	7.5	0.023	7.9	8.4	22.1
6	0.08	2.3	7.2	7.4	0.027	7.8	7.4	23.6

Table 4: Heavy metal contents of substrate samples (Chernozem : quartz sand = 1:1) after rape harvest in the year 1996
Tabelle 4: Schwermetallgehalte von Substratproben (Tschernosem : Quarzsand = 1:1) nach der Rapsernte im Jahr 1996

n = 4 (sewage sludge) or n = 8 (NH_4-N)

	Cd		Cr		Cu		Zn	
Treatment	mg/kg	St. dev.	mg/kg	St. dev.	mg/kg	St. dev.	mg/kg	St. dev.
1 + 4 (NH_4-N)	0.11	0.04	9.25	1.47	6.78	0.92	22.43	2.23
3 (Sewage sludge 7.5 t/ha)	0.12	0.01	9.05	0.08	7.35	0.50	25.00	1.41
6 (Sewage sludge 15 t/ha)	0.11	0.02	8.67	0.75	7.22	0.96	24.00	2.83
Average	0.11	0.06	8.80	0.77	7.03	0.79	24.31	3.15

Table 5: Heavy metal contents of substrate samples (Chernozem : quartz sand = 1:1) after rape harvest in the year 1997
Tabelle 5: Schwermetallgehalte von Substratproben (Tschernosem : Quarzsand = 1:1) nach der Rapsernte im Jahr 1997

mg/kg dry matter

Treatment	Cd	Co	Cr	Cu	Hg	Ni	Pb	Zn
1	0.10	1.6	5.4	5.7	0.019	5.1	7.0	18.9
2	0.11	1.7	5.4	6.2	0.027	5.3	7.2	19.7
3	0.11	1.8	5.6	6.9	0.029	5.2	8.0	21.5
4	0.12	1.8	5.5	6.0	0.017	5.3	7.0	18.2
5	0.11	1.8	5.3	6.7	0.024	5.3	8.3	21.8
6	0.13	1.8	6.2	7.2	0.025	5.9	7.9	23.4

which is reflected by the standard deviations of sometimes 10 % or more and the incomplete extraction of heavy metals by aqua regia. GERZABEK (1993) reported that acid extractions yield only 69 %, 49 %, 76 % and 87 % of the real total contents of Cd, Cr, Cu and Zn in soil samples, respectively. A second reason could be losses of mobile heavy metal fractions from the pots. Such an effect was also observed in a field pot trial by KAMEL (1990). Losses of heavy metals from experimental soils in this earlier study reached up to 17 % (Zn) of the initial value within 550 days. Results of long-term experiments with sewage sludge in Japan also indicated considerable losses of Cadmium and Zinc from sewage sludge additions (KAWASAKI, 1996). The conditions for heavy metal leaching in our experiment were more favourable than in field conditions due to the obvious increase of the hydraulic conductivity in the substrate by the quartz sand application, part of the observed effect, therefore, could be biased by the experiment itself. Nevertheless, it has to be taken into account that the mobile heavy metal fraction in sewage sludge generally is larger than in uncontaminated soils. This is supported by the results of the mobile heavy metal fraction determination in the second experimental year (1996, Table 6).

The portion extracted by ammonium acetate ranged between 0.015 % (Cr) and 7.3 % (Cd) of the aqua regia extracted contents. The less mobile element chromium did not show any response to the sewage sludge treatments, whereas Cd-, Cu- and Zn-concentrations in the ammonium acetate extract increased significantly with increasing sewage sludge applications. The concentration of extract-

Table 6: Mobile heavy metal fractions extracted by ammonium acetate (year 1996)
Tabelle 6: Mit Ammoniumacetat extrahierte mobile Schwermetallfraktionen (Jahr 1996)

n = 4 (sewage sludge) or n = 8 [$(NH_4)_2SO_4$]

	Cd		Cr		Cu		Zn	
Treatment	µg/kg	St. dev.	µg/kg	St. dev.	mg/kg	St. dev.	mg/kg	St. dev.
1 + 4 (NH_4-N)	7.04 a	1.09	1.41	0.27	0.031 a	0.010	0.040 a	0.018
3 (Sewage sludge 7.5 t/ha)	7.00 a	0.20	2.65	1.47	0.045 b	0.010	0.087 b	0.009
6 (Sewage sludge 15 t/ha)	11.00 b	3.01	1.37	0.21	0.060 c	0.008	0.095 b	0.017
Average	8.02	1.43	1.71	0.65	0.042	0.009	0.065	0.015
% of total	7.3		0.015		0.60		0.27	

Values in the same column followed by different letters are different at $p<0.05$ (Sheffé test).

able Cu and Zn increased by a factor of 1.94 and 2.38, respectively, between control pots and those receiving an equivalent of 15 t sewage sludge per hectare. It can be concluded that the ammonium acetate extraction is a potentially suitable and sensitive method to determine an increase of heavy metal mobility due to the application of organic residues. Similar trends of increasing mobile heavy metal fractions due to sewage sludge application were reported by KUMAZAWA (1996).

3.2 Heavy metals in rape

Table 7 shows mean values of the heavy metal concentrations in rape plants of the first experimental year (1995).

Rape plants did not exhibit any heavy metal contents, which might indicate a high availability in soil. The comparison with literature values (SAUERBECK, 1985) shows that none of the investigated elements exceeded ranges frequently observed in plant biomass. The measured concentrations in most cases were close to the lower limit observed in nature. The high pH of the substrate did not favour heavy metal plant availability, which could be the main reason for the observed low uptake. Generally only few differences between the mineral fertiliser pots and those amended with sewage sludge occurred (Tables 7, 8, 9). Only copper concentrations in rape plants (year 1995) were slightly but significantly increased by a single sewage sludge treatment of 7.5 t/ha. In the year 1996 small but significant differences between treatments were measured for Cadmium and Chromium (Table 8).

The 7.5 t/ha sewage sludge treatment showed the lowest values in both cases, although the mobile Cd- and Cr-fraction were quite similar in the mineral fertilizer and 7.5 t/ha sewage sludge treatment (Tables 6, 8). The 15 t/ha treatment was not significantly different from the control. According to the literature the application of organic manures or residues can reduce heavy metal uptake into agricultural crops. This is explained by the fact that even quite small quantities of organic matter applied to soil through compost (SCHERER et al., 1997) or sewage sludge (SMITH, 1996; ELLIOT and SINGER, 1988) can increase the adsorption

Table 7: Heavy metal concentrations in rape biomass in the year 1995
Tabelle 7: Schwermetallkonzentrationen in der Raps-Biomasse im Jahr 1995

mg/kg dry matter (mean and standard deviations)
n = 16 (sewage sludge) or n = 8 [$(NH_4)_2SO_4$]

Element	$(NH_4)_2SO_4$-treatment	7.5 t sewage sludge / ha
Cd	0.16 ± 0.02	0.14 ± 0.03
Co	0.099 ± 0.030	0.115 ± 0.038
Cu	3.53 ± 0.29 a	3.96 ± 0.48 b
Hg	0.024 ± 0.007	0.026 ± 0.013
Ni	1.16 ± 0.29	1.04 ± 0.30
Pb	0.23 ± 0.07	0.24 ± 0.07
Zn	28.9 ± 5.5	29.8 ± 3.7

Values in the same row followed by different letters are different at $p<0.05$ (Duncan's test).

Table 8: Heavy metal concentrations in rape biomass in the year 1996
Tabelle 8: Schwermetallkonzentrationen in der Raps-Biomasse im Jahr 1996

n = 4 (sewage sludge) or n = 8 [$(NH_4)_2SO_4$]

	Cd		Cr		Cu		Zn	
Treatment	mg/kg	St. dev.	mg/kg	St. dev.	mg/kg	St. dev.	mg/kg	St. dev.
1 + 4 (NH_4-N)	0.25 b	0.03	1.66 b	0.56	3.87	0.50	31.50	2.62
3 (sewage sludge 7.5 t/ha)	0.18 a	0.03	0.64 a	0.37	3.37	0.31	33.25	3.95
6 (sewage sludge 15 t/ha)	0.23 ab	0.02	1.22 ab	0.21	3.52	0.37	36.25	3.40
Average	0.23	0.03	1.30	0.38	3.66	0.39	33.12	3.32

Mean values in the same column followed by different letters are significantly different at $p<0.05$ (Sheffé test).

capacity of the soil and thus diminish heavy metal uptake through formation of stable organo-metallic complexes. The lack of a diminishing effect of Cd and Cr uptake into rape in the 15 t/ha sewage sludge treatments (1996) might be a bias of the experimental setup in this year. The 7.5 t/ha treatment was the only one not receiving ammoniumsulfate as mineral N-source. Ammonium is known to reduce the pH in the rhizosphere (MARSCHNER, 1986) and thus may have increased heavy metal availability in the 15 t/ha treatment. In the year 1997 again there was no clear response of heavy metal concentrations in rape plants to the different treatments, both higher (Cu, Hg, Zn) and lower heavy metal concentrations (Cd, Ni) were observed in some sewage sludge treaments compared to the control (Table 9).

Thus, no final conclusion on the impact of sewage sludge on heavy metal uptake into rape plants can be deduced from our study, although sewage sludge quantities applied were much larger than in practice. These findings generally are supported by results from a three years field trial in Austria on a similar Chernozem soil (KOCH and GERZABEK, 1996), where no clear impact of repeated sewage sludge treatments on heavy metal concentrations in field crops was observed. In future it may be important to consider the nutritional status of the plants and the processes taking place in the rhizosphere for the improvement of the prediction of heavy metal availability.

3.3 Rape yield and nitrogen uptake

Rape yields in the year 1995 were significantly enhanced by sewage sludge applications (Table 10).

Average rape shoot dry matter in sewage sludge treated pots was 150 % higher than in pots treated with mineral nitrogen solely, differences between irradiated and not irradiated sewage sludge were not statistically significant at the $p<0.05$-level. The growth effect can be attributed to the considerable inputs of available plant nutrients, especially N, P and K by sewage sludge (Table 1). Mineral nitrogen provided by an equivalent of 7.5 t sewage sludge per hectare exceeded N from $(^{15}NH_4)_2SO_4$ by a factor of 2.16. Due to large variations between replicates there were no significant differences of nitrogen contents in rape shoots between treatments, the nitrogen uptake per pot was significantly enhanced by the sewage sludge application (Table 10). Sewage sludge treated rape exhibited a 1.92 times higher N-uptake as compared to the control. Differences between irradiated and not irradiated sewage sludge treatments concerning N-uptake were not statistically significant.

Nitrogen in rape aboveground biomass originating from $(^{15}NH_4)_2SO_4$ was highest in the control pots (Table 10, Figure 1), as was the nitrogen utilisation from $(^{15}NH_4)_2SO_4$. The ammonium fertiliser contributed approximately 10 %

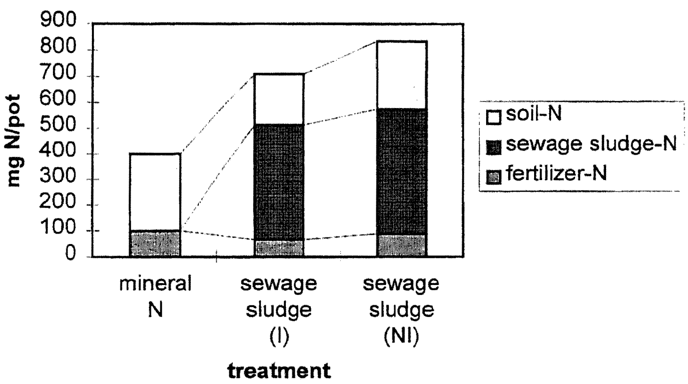

Figure 1: Contribution of N-sources to N-uptake into rape plants (year 1995); I: irradiated, NI: not irradiated

Abbildung 1: Beitrag der N-Quellen zur N-Aufnahme bei Rapspflanzen (Jahr 1995) I: bestrahlt, NI: unbestrahlt

Table 9: Heavy metal concentrations in rape biomass in the year 1997
Tabelle 9: Schwermetallkonzentrationen in der Raps-Biomasse im Jahr 1997

mg/kg dry matter
n = 4

Treatment	Cd	Co	Cu	Hg	Ni	Pb	Zn
1	0.12 b	< 0.1	3.1 ab	0.047 abc	0.40 de	< 0.1	13.9 a
2	0.11 b	< 0.1	3.2 b	0.040 abc	0.32 cd	< 0.1	15.4 ab
3	0.11 b	< 0.1	3.1 ab	0.037 ab	0.24 bc	< 0.1	17.6 bc
4	0.12 b	< 0.1	2.6 a	0.031 a	0.25 bc	< 0.1	15.4 ab
5	0.09 a	< 0.1	3.2 b	0.055 c	0.12 a	< 0.1	16.3 abc
6	0.10 ab	< 0.1	4.2 c	0.051 bc	0.16 ab	< 0.1	18.3 c

Mean values in the same column followed by different letters are significantly different at $p<0.05$ (LSD method).

Table 10: Rape yield and N-balance of $^{15}NH_4$-N and sewage sludge-N in the experimental year 1995
Tabelle 10: Rapsertrag und Stickstoffbilanz des $^{15}NH_4$-N sowie des Klärschlamm-N im Versuchsjahr 1995

n = 4

	$(^{15}NH_4)_2SO_4$	^{15}N + sewage sludge (SeSl) (irradiated)	^{15}N + sewage sludge (not irradiated)
N supplied from $(^{15}NH_4)_2SO_4$ (mg/pot)	795	795	795
N supplied from SeSl (mg/pot)	0	6320	6320
N_{min} (NH_4 + NO_3) from SeSl (mg/pot)	0	1717	1717
rape yield g DM/pot	22.63 ± 1.87 a	33.31 ± 7.00 b	35.18 ± 4.98 b
%N in rape crop	1.77 ± 0.18	2.13 ± 0.36	2.37 ± 0.61
N-uptake by rape; mg N/pot (%)	400.0 ± 31.5 a (100)	708.7 ± 31.0 b (100)	833.8 ± 289.8 b (100)
^{15}N at% excess (rape)	1.211 ± 0.135 b	0.449 ± 0.070 a	0.506 ± 0.075 a
N originating from $(^{15}NH_4)_2SO_4$ mg/pot (%)	100.4 (25.1)	65.9 (9.3)	87.5 (10.5)
N originating from sewage sludge mg/pot (%)	0	446.2 (63.0)	485.7 (58.3)
N originating from soil mg/pot (%)	299.6 (74.9)	196.6 (27.7)	260.6 (31.2)
N-utilisation from $(^{15}NH_4)_2SO_4$; %	12.6	8.3	11.0
N-utilisation from SeSl - N; %	-	7.1	7.7
^{15}N at% excess (substrate)	0.038	0.045	0.038

Values in the same row followed by different letters are different at $p<0.05$ (Duncan's test).

Table 11: Literature values of N-utilisation from sewage sludge in the first year after application
Tabelle 11: Literaturwerte der N-Verwertung aus Klärschlamm im ersten Jahr nach der Ausbringung

crop	country	%N utilisation	authors
sugarcane leaves	Argentina	7.9-19%	MAGNAVACCA et al. (1996)
maize (aboveground biomass)	Malaysia	5.8-7.8%	ISHAK et al. (1996)
wheat	China	1.7-15.2%*	calculated from JIANG et al. (1996)
rape	Austria	7.1-7.7%	this study

* Values obtained by using the difference method.

to the N-uptake into rape treated additionally with sewage sludge (Figure 1).

The largest N-portion found in these rape plants originated from sewage sludge (60.7 % on average). Sewage sludge nitrogen both substituted soil-N and fertiliser-N uptake and accounted for the increase of the total N-uptake as compared to the control. Within 40 days rape was able to use 7.4 % (mean of irradiated and not irradiated sewage sludge) of total nitrogen or 27.1 % of mineral nitrogen provided by the sewage sludge additions. Similar N-utilisation obtained by the ^{15}N-dilution technique was reported for other crops in literature (Table 11).

According to SMITH (1996) the sewage sludge used in our experiment can be classified as sludge with high mineralisable N (total N > 5 %, C:N < 8). PAUL and BEAUCHAMP (1995) reported ^{15}N-recoveries by maize plants from $(^{15}NH_4)_2SO_4$ and from dairy cattle slurry of 29 % and 15 %, respectively. Considering the short vegetation period in our experiment, the latter value support our findings.

The results of the experimental year 1996 differed significantly from the year 1995 (Table 12).

Due to wet soil conditions rape plants showed an unfavourable development after incomplete germination of rape seeds in spring. Therefore, no significant differences were obtained for rape yield. Nitrogen contents of rape plants were higher in treatments 4–6 due to additional N-supply through ammonium fertiliser and sewage sludge. N-uptake did not differ significantly between treatments. The ^{15}N-enrichment in rape biomass showed no statistically significant differences (Table 12). The reasons

Table 12: Rape yields and N-uptake in the second experimental year (1996)
Tabelle 12: Rapsertrag und Stickstoffaufnahme im 2. Versuchsjahr (1996)

n = 4

	1	2	3	4	5	6
rape yield g DM/pot	57 ± 11	84 ± 46	62 ± 43	69 ± 12	75 ± 53	63 ± 9
% N in rape DM	1.80 ± 0.20	1.89 ± 0.25	2.07 ± 0.28	2.30 ± 0.12	2.36 ± 0.17	2.17 ± 0.28
^{15}N at.% exc. in rape	0.025 ± 0.010	0.031 ± 0.017	0.038 ± 0.026	0.050 ± 0.017	0.075 ± 0.046	0.057 ± 0.016
mg N-uptake per pot	1025	1583	1283	1592	1766	1366
% Ndff	0.519*	0.643*	0.788*	1.04**	1.56**	1.18**
N-utilisation from $(^{15}NH_4)_2SO_4$, %	0.669*	1.28*	1.27*	2.08**	3.47**	2.03**
^{15}N at.% exc. in substrate	0.049	0.035	0.037	0.041	0.044	0.046

1: $(^{15}NH_4)_2SO_4$ in 1995
2: sewage sludge (SeSl) irradiated 7.5 t/ha and $(^{15}NH_4)_2SO_4$ in 1995
3: 7.5 t SeSl not irradiated per ha and $(^{15}NH_4)_2SO_4$ in 1995
4: $(NH_4)_2SO_4$ in 1995, $(^{15}NH_4)_2SO_4$ in 1996
5: SeSl irradiated 7.5 t/ha and $(NH_4)_2SO_4$ in 1995, SeSl irradiated 7.5 t/ha and $(^{15}NH_4)_2SO_4$ in 1996
6: SeSl not irradiated 7.5 t/ha and $(NH_4)_2SO_4$ in 1995, SeSl not irradiated 7.5 t/ha and $(^{15}NH_4)_2SO_4$ in 1996
* ^{15}N supplied 1995
* ^{15}N supplied 1996

are high losses of labelled mineral fertiliser nitrogen through leaching and/or volatilisation, which are most likely during the wet spring. This explanation is supported by the extremely low N-utilisation from labelled ammonium sulphate in the year 1996 of 2 to 3.5 % obtained from the isotopic dilution calculation (treatments 4–6, Table 12), which is a factor of 2.4 to 6 lower than in 1995. The contribution of ammonium sulphate nitrogen to the N-uptake of rape in 1996 was a factor of 6 to 24 lower as compared to the results in the year 1995. High nitrogen leaching losses and a factor of 6 lower mineral nitrogen fraction in sewage sludge applied in spring 1996 (Table 1) could be the explanation of the lacking effect of sewage sludge on yield and N-uptake of rape. Similar problems with the isotopic method used in the present experiment were reported by JIANG et al. (1996) and AZAM et al. (1996). The latter authors reported a trend of increasing ^{15}N contents in plants grown on sewage sludge treated plots. This trend (not significant) was also obtained in pots of treatments 1 to 3 (Table 12). This result could suggest that sewage sludge treated pots kept a higher portion of mineral fertiliser nitrogen. The input of organic matter to the soil increased probably the microbial biomass, which biologically fixed some of the mineral nitrogen. Negative effects of the heavy metal input on soil microbial activity can only be observed in long-term studies (SMITH, 1996) and thus are not likely in our experiment.

In the year 1997 the isotopic dilution experiment again was successful and clear effects of the sewage sludge additions in the previous years could be detected. Rape shoot yield was in some cases significantly higher in the sewage sludge treated plots as compared to the control (Table 13).

Nitrogen uptake also clearly showed the residual effect of the sewage sludge additions in the years 1995 (treatment 2 + 3) and 1995 + 1996 (treatments 5 + 6). N-uptake into rape aboveground biomass increased from the control plots, the pots having received sewage sludge in 1995 (7.5 t/ha) to the 15 t sewage sludge per hectare treatments. The increase of N-contents in rape shoots was only significant for the repeated sewage sludge treatment (treatments 5 + 6, Table 13). Both, for N-uptake and N-contents no significant differences could be observed between irradiated and non-irradiated sewage sludge.

The contribution of fertiliser nitrogen to the N-uptake into rape plants was nearly constant for all different treatments in the year 1997 (Figure 2).

The N-utilisation from ammoniumsulphate (average: 33.1 %) was clearly higher than in the year 1995, which can be attributed to the split application in the experimental year 1997. In the solely ammoniumsulphate treated plots

Table 13: Rape yields and N-uptake in the third experimental year (1997)
Tabelle 13: Rapserträge und N-Aufnahme im 3. Versuchsjahr (1997)

n = 4

	1	2	3	4	5	6
N supplied from $(^{15}NH_4)_2SO_4$ in 1997 (mg/pot)	795	795	795	795	795	795
N supplied from SeSl (mg/pot)	0	6320	6320	0	12783	12783
rape yield g DM/pot	37.07 ± 4.00 ab	44.16 ± 5.00 c	41.64 ± 6.30 bc	32.24 ± 3.02 a	40.78 ± 2.85 bc	40.84 ± 1.17 bc
% N in rape DM	1.11 ± 0.30 a	1.33 ± 0.12 ab	1.50 ± 0.25 bc	1.51 ± 0.24 bc	1.74 ± 0.11 c	1.72 ± 0.28 c
mg N-uptake per pot	414.2 ± 131.4 a	585.5 ± 74.2 ab	622.4 ± 142.8 bc	488.6 ± 108.9 ab	625.5 ± 153.1 bc	698.7 ± 102.7 c
^{15}N at.% exc. in rape	2.619 ± 0.217 c	2.378 ± 0.188 c	2.121 ± 0.178 b	2.459 ± 0.132 c	1.800 ± 0.118 a	1.834 ± 0.139 a
N from $(^{15}NH_4)_2SO_4$, mg/pot (%)	224.9 (54.3)	228.7 (39.1)	273.9 (44.0)	249.2 (51.0)	233.3 (37.3)	265.5 (38.0)
N from sewage sludge, mg/pot (%)	0 (0)	36.9 (6.3)	102.7 (16.5)	0 (0)	182.0 (29.1)	194.2 (27.8)
N from soil, mg/pot (%)	189.3 (45.7)	319.9 (54.6)	245.8 (39.5)	239.4 (49.0)	210.2 (33.6)	239.0 (34.2)
N-utilisation from $(^{15}NH_4)_2SO_4$, %	35.3	34.8	34.5	31.1	29.3	33.4
N-utilisation from sewage sludge, %	0	0.58	1.62	0	1.42	1.52
^{15}N at.% exc. in substrate	0.192	0.128	0.184	0.168	0.130	0.120

1: $(^{15}NH_4)_2SO_4$ in 1995 and 1997
2: sewage sludge (SeSl) irradiated 7.5 t/ha and $(^{15}NH_4)_2SO_4$ in 1995 and 1997
3: 7.5 t SeSl not irradiated per ha and $(^{15}NH_4)_2SO_4$ in 1995 and 1997
4: $(NH_4)_2SO_4$ in 1995, $(^{15}NH_4)_2SO_4$ in 1996 and 1997
5: SeSl irradiated 7.5 t/ha and $(NH_4)_2SO_4$ in 1995, SeSl irradiated 7.5 t/ha and $(^{15}NH_4)_2SO_4$ in 1996 + 1997
6: SeSl not irradiated 7.5 t/ha and $(NH_4)_2SO_4$ in 1995, SeSl not irradiated 7.5 t/ha and $(^{15}NH_4)_2SO_4$ in 1996 + 1997
Values in the same row followed by different letters are different at $p<0.05$ (Duncan´s test).

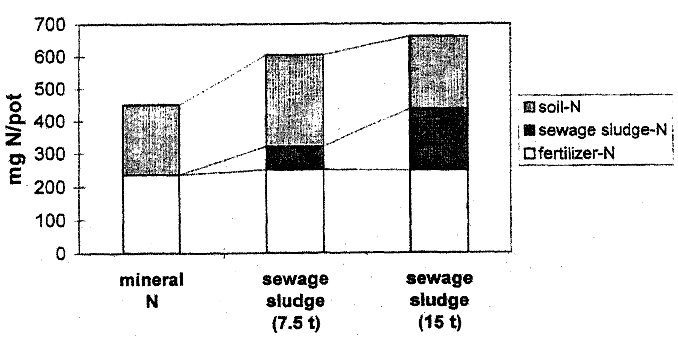

Figure 2: Contribution of N-sources to N-uptake into rape plants (year 1997)
Abbildung 2: Beitrag der N-Quellen zur N-Aufnahme bei Rapspflanzen (Jahr 1997)

fertiliser-N was the main source of plant uptake, the pots treated in the year 1995 with 7.5 t sewage sludge per hectare provided approximately the same amount of N from ammoniumsulphate and soil. N-uptake from soil into rape was partly substituted by sewage sludge nitrogen in pots having received sewage sludge in the years 1995 and 1996. The average contribution of sewage sludge – N to the uptake was 11.4 % and 28.5 % for the 7.5t/ha and 15 t/ha treatment, respectively. The high N-portion originating from sewage sludge observed for the 15 t/ha treatment as residual effect supports the exceedingly high values of app. 60.7 % detected in 1995 shortly after the first sewage sludge application. The absolute N-utilisation from sewage sludge was quite low and did not exceed 1.5 % (Table 13), which is distinctly lower than in the first year (Table 10). Using the difference method the following average N-utilisations from the sewage sludge applications can be calculated: 7.4 % (first year), 1.8 % (second year), 1.1 % (third year). In total approximately 10.3 % of sewage sludge-N_t could be used by the rape plants during a three years period.

4. Conclusions

- Heavy metal concentrations in the substrate (aqua regia extraction) did not change significantly after the addition of equivalents of 7.5 and 15 t sewage sludge dry matter per hectare. Mobile fractions of Cd, Cu and Zn measured in the year 1996 increased significantly in the substrate due to sewage sludge treatments. However, no clear trends in heavy metal uptake into rape due to the sewage sludge treatments were observed.
- Dry matter yield of rape and nitrogen uptake were significantly enhanced due to sewage sludge treatments in the first and third year. In the year 1996 rape yields, nitrogen contents and the ^{15}N-enrichment in rape biomass showed no statistically significant differences between treatments due to high nitrogen losses during spring 1996 and a slow development of rape plants. The yield increase and enhanced N-uptake in the third year can be interpreted as residual effect of the sludge applied in the previous years.
- Irradiation of sewage sludge did not result in any significant effect on rape biomass production, N-availability in the sewage sludge or heavy metal uptake by plants.
- The ^{15}N dilution method can be distinctly improved by splitting the labelled fertiliser.

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