Agronomic and environmental impacts of a single application of heat-dried sludge on an Alfisol

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Abstract

A field experiment was conducted on Alfisols in South-West France to assess the agronomic and environmental impacts of a single application of heat-dried sludge pellets at 11.1 Mg dry matter ha^{-1}. The sludge pellets, with a moisture level of 9.5%, were spread on an irrigated crop of maize (*Zea mays* L.). This treatment was compared with inorganic fertilization (urea and diammonium phosphate mixed with KCl). Soil properties, yield and the composition of maize and the quality of drained water were monitored over 1 year to detect any changes resulting from sludge application. Amongst several determined soil properties, only two were significantly modified by the sludge application: The nitric nitrogen stock of the soil was higher in the inorganic fertilized plot, whereas Olsen-P soil content was higher in the sludge-amended plot. Agronomic recovery rates of N and P added by sludge were high: For the first crop following application, total amounts of N and P supplied by the sludge had the same efficiency as approximately 45% of the N and P amounts supplied by inorganic fertilizer. This ratio was 7% for the N uptake by the second maize crop. The quality and quantity of maize were equally good with both types of fertilization. During the 2 years following sludge spreading, N leaching remained as low in the sludged plot as in the inorganically fertilized one. The Cu, Zn, Cr, Cd, Pb and Ni composition of the drainage water was affected by neither of the types of amendment. From the heavy-metal contents of the soil, water and maize monitored over 1 year in the field experiment and from literature data for cow manure and atmospheric emissions, a theoretical balance between crop soil heavy-metal input and output over one century was drawn up. The long-term impact of cow manure on Zn, Ni and Cr in soil is higher than that of the studied heat-dried sludge. Obviously, sludge tended to cause a strong increase in soil Cu storage, valued for these soils, which are otherwise very Cu deficient.

Keywords: Sewage sludge; Thermal process; Luvisols; Drainage water; *Zea mays*; Heavy metals; Fertilization

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1. Introduction

Domestic wastewater treatment has been improved to reduce water pollution. As a result, the quantity of wastewater residues, i.e., sewage sludge, has increased worldwide. In 2000, annual sludge production reached around 7×10^6 dry metric tons (Mg) in the United States (Epstein, 2003), 17×10^6 Mg in Europe (Gavalda et al., 2001) and approximately 3×10^6 Mg in New Zealand and Australia (Cameron et al., 1997). The problem of the disposal of such by-products has increased with human population in cities (Krogmann et al., 1997). Spreading sewage sludge on land recycles valuable plant nutrients (Peterson et al., 1993) and avoids burning the main element of sludge: carbon. By enhancing organic matter in crop soils, sewage sludge may improve the physical properties of soils. According to Clapp et al. (1986), many experiments conducted in various countries around the world have shown that sewage sludge supply improved bulk density, aggregation, porosity, hydraulic conductivity and water retention. Therefore, sludge has been used as an organic amendment for the reclamation of degraded soils (White et al., 1997; Epstein, 2003). Moreover, soil microbial activity can be stimulated by sludge (Brendecke et al., 1993; Moreno et al., 2003). In other areas where intensive cropping caused nutrient loss, the addition of sludge (with low metal levels) generally promoted biomass production (Labrecque et al., 1998; Bidegain et al., 2000; Binder et al., 2002).

Before being recycled as fertilizer, sludge must be treated to reduce its pathogen concentration and water content. The effects of composting, liming and anaerobic treatment have been widely studied (Coker, 1983; Krebs et al., 1998; Darees Boucher et al., 1999), while little research has been done on the effects of thermal conditioning on sludge agronomic value. However, such a dewatering process produces dry pellets that are convenient because they can be readily spread without any odour nuisance and with the same tools as those used for manure. Thus, this new method of conditioning is particularly appealing. However, the fertilizing potential of the pellets has to be verified. Likewise, no experiments have yet been carried out on the soils in this area where sludge has been spread.

The aim of this work was to quantify the crop-yield response to heat-dried sludge for irrigated maize and its impact on the environmental and agronomic properties of Alfisols under actual field conditions. Sludge conditioned in this way was tested in a field experiment on a maize crop and was compared with an inorganic fertilizer. Soil properties, yield and composition of maize and drained water quality were monitored over 1 year (two for nitrate leaching) to detect any changes caused by spreading heat-dried sludge. The data, collected for 1 year, were extrapolated to a century to check whether sludge application could cause heavy metal accumulation in these crop soils. To do this, the hypothesis took into account the present legal agricultural practices. Heavy metals leaching and uptaking were also assumed to be constant over one century.

2. Materials and methods

2.1. Experimental site and setup

The field experiment was conducted at the Ecole Nationale Supérieure Agronomique de Toulouse (ENSAT) field station in the large alluvial corridor of the Garonne, 35 km South-West of Toulouse (France). The experimental site is located in the Midi-Pyrenees area at Poucharramet (43°24’N, 1°11’W). The site’s average altitude is 100 m above sea level, and it is subjected to an oceanic climate with both Atlantic and Mediterranean influences. The mean annual temperature is 13.2 °C, and the mean annual rainfall is 800 mm.

The soils are Alfisols. They belong to the Epiaqualfs according to American soil taxonomy and to redoxi–luvisols according to the ISSS (1998). These soils are common in temperate regions. All the quaternary alluvial deposits of South-West French rivers have the same pedogenetic evolution towards Alfisols, locally called “Boulbènes”. Their first characteristic is a great textural differentiation in the profile, showing a surface horizon depleted of clay and an accumulation of clay in a 0.6-m-deep argic horizon. Mainly formed by illuviation, the argic horizon is compact and has a low internal permeability. As a result, water stagnates in the upper horizon during winter while the soils are actually dry in summer. Such difficult agricultural conditions have led farmers to extensively pipe-drain and irrigate the Boulbènes

Table 1
Physical and chemical soil characteristics in the experimental site in April 1997 (mean and S.D.; n=24 upper soil horizon) and limit values

	Soil data	Limit values permitted in France for sewage sludge application[a]
pH_{water}	5.94±0.21	
Total organic Carbon (g kg^{-1} soil)	7.23±0.76	
Total N (g kg^{-1} soil)	0.79±0.04	
Total P (g kg^{-1} soil)	238±24	
Olsen P (mg kg^{-1} soil)	27±5.1	
Cation exchange capacity (cmol$_c$ kg^{-1} soil)	6.01±0.73	
Total iron (mg kg^{-1} soil)	16165±1115	
Total manganese (mg kg^{-1} soil)	261±51	
Total copper (mg kg^{-1} soil)	7.43±0.61	100
Total zinc (mg kg^{-1} soil)	28.4±1.5	300
Total chromium (mg kg^{-1} soil)	15.7±0.6	150
Total lead (mg kg^{-1} soil)	11.3±1.1	100
Total nickel (mg kg^{-1} soil)	8.24±0.54	50
Total cadmium (mg kg^{-1} soil)	0.14±0.02	2
Particle size distribution		
Sand % 32±3		
Silt % 53±3		
Clay % 15±1		

[a] Limit values allowed by French legislation (since January 1998).

(Zimmer et al., 1991). The upper horizon with the low clay level is acid (Table 1). Its low organic matter content, usually around 1%, is paralleled by deficient physical and chemical fertility. Thus, biosolids could obviously be supplied to such soils whose heavy-metal contents are far beneath the permitted French and European limits (Table 1). The site is situated inside nitrate-vulnerable zones, thus, nitrate leaching was also monitored.

The sludge studied in this experiment was from the wastewater treatment plant of Toulouse (population 600,000). It was produced by aerobic biological treatment of municipal wastewater. Raw liquid sludge produced by this plant was previously tested in a former experiment (Guiresse et al., 1995). The authors reported good nitrogen and phosphorus sludge efficiencies on maize crops. The number of inhabitants in Toulouse has been growing, as has sludge production. Hence, a new dewatering process has been set up to reduce the final biosolid volume. With a flash thermal process, sludge is dried at 105 °C, resulting in loss of enough volatile matter to cause measurably lower high heating values before the sludge would enter the active combustion phase (Krogmann et al., 1997). As a large proportion of sludge carbon is volatile, nitrogen emission may also occur during heating. Finally, the sludge pellets had a water content of 9.5% (Table 2).

Three treatments were set up: a control plot received no fertilizer, plot I was fertilized with an inorganic fertilizer, and plot S was fertilised with the heat-dried sludge. Each treatment, except for the control, was carried out in triplicate and each replicate was around 2000 m^2. The three replicates of each treatment were distributed within a plot, which is hydraulically isolated by a pipe drainage system, making it possible to monitor the quality of the drained water. The two plots used in the present experiment were chosen because their hydrodynamic properties were very close. Although the drainage pipes were not equally spaced: 10 m apart in plot S and 25 m in plot I, Guiresse et al. (1989) revealed that flow drained from plot S was barely above that of plot I because the outflow rates are more relevant to the properties of soil than to pipe row spacing.

Table 2
Physicochemical characteristics of the sludge pellets (mean and S.D.; n=6) and limit values

	Sludge data	Limit values permitted in France for sewage sludge application in agriculture[a]
Water content (% fresh weight)	9.5	
Characteristics on a dry matter basis		
Ash (%)	20.1±0.5	
Organic matter (%)	79.9±0.5	
Total organic carbon (%)	48.1±1.1	
Total N (%)	4.49±0.2	
NH_4^+–N (%)	0.84±0.04	
Total P (%)	1.91±0.17	
Total Ca (%)	1.77±0.06	
Total Mg (%)	0.31±0.01	
Total K (%)	0.23±0.02	
Total copper (mg kg^{-1})	188±27	1000
Total zinc (mg kg^{-1})	330±48	3000
Total chromium (mg kg^{-1})	24.9±3.8	1000
Total lead (mg kg^{-1})	49.5±6.5	800
Total nickel (mg kg^{-1})	16.1±2.0	200
Total cadmium (mg kg^{-1})	1.57±0.25	10

[a] Limit values allowed by french legislation (since January 1998).

The sludge pellets were spread on May 15th, 1997 at 11.1 Mg ha^{-1} (dry matter basis) with a tow-behind spreader equipped with two spinning wire agitators and immediately incorporated into the top 0.2 m of soil with a cultivator. This supply of exogenous organic matter represented 28% of the organic matter naturally present in the soil. While the pellets were being spread, six samples were collected and analyzed according to the same protocols as those used for the plant analyses described below. In agreement with the sludge composition (Table 2), the total amounts of nutrient elements provided by sludge were 500 kg N ha^{-1}, 213 kg P ha^{-1} and 36 kg K ha^{-1}; heavy-metal input via sludge and fertilizer is given in Table 3. In plot I, inorganic fertilization was supplied by urea and 17 triple phosphate (diammonium phosphate mixed with KCl and urea): 240 kg N ha^{-1}, 37 kg P ha^{-1} and 71 kg K ha^{-1}. Treatments were arranged in three complete blocks. *Zea mays* L. cv. Dunia was sown on May 20th, 1997 at 87,000 plants ha^{-1} in rows spaced 0.8 m apart. Immediately after maize grain harvest (October 17th, 1997), the stubble was incorporated into the top soil, and ryegrass was sown without any fertilization to reduce nitrate leaching during the winter. On May 18th, 1998, the second maize crop was sown cv. DK512 with the same density and row spacing, but without any fertilization, to assess the residual agronomic effects of heat-dried sludge.

2.2. Sample collection, analysis and calculations

Three composite soil samples were collected for each replicate of each treatment. They were taken from the tilled horizon (0–0.20 m) with a drill before the sludge was spread (April 1997), during maize growth (July 1997) and just after grain harvest (October 1997). Additional soil samples were taken in September for the monitoring of nitrates in soils. Nitric nitrogen (NO_3^-N) was extracted with 1 M KCl solution (Mulvaney, 1996). In prior studies carried out at the same site by Guiresse et al. (1995), NH_4^+ represented less than 1% of the nitric nitrogen, thus, in the present experiment, only soil NO_3^- was monitored. Soil samples were air dried and passed through a 2-mm sieve. The following soil properties were analyzed: pH (1:1 soil/H_2O), organic carbon (C) content with a dichromate–sulfuric acid mixture (Nelson and Sommers, 1996), cation exchange capacity with ammonium acetate and total N by the Kjeldhal method (Bremner, 1996). Total soil zinc (Zn), copper (Cu), chromium (Cr), iron (Fe), cadmium (Cd), lead (Pb), nickel (Ni), manganese (Mn) and phosphorus (P) content was determined in *aqua regia* digests: 3 g soil was mixed with 21 ml HCl and 7 ml HNO_3 during 12 h, then heated at 150 °C during 2 h, according to ISO normalization (ISO 11466), and finally analyzed by ICP-MS (Elan 6000, Perkin Elmer SCIEX). A mix of ICP grade indium and rhenium in 5% nitric acid solution (Merck, France, F67023 Strasbourg) was used as internal standard. Available forms of heavy metals were studied by extraction with diethylene triamine penta-acetic acid (DTPA) at pH 7.3 and determination by atomic absorption spectrometry (Perkin Elmer Analyst 100, France). European standard material CRM 141 R (calcareous loam soil) was provided by Promochem F67123 Molsheim. Available Olsen-P was extracted with 0.5 M $NaHCO_3$ (pH 8.5) mixed to soil during 30 min, filtered and then determined by ascorbic acid colorimetry (Kuo, 1996).

Aboveground, a biomass of maize was sampled at physiological maturity on September 18th. In each replicate, three composite samples were made up with three whole plants. After separating the leaves and stalks from the grains, the samples were weighed, chopped, dried at 80 °C, reweighed for moisture calculation and ground. On the subsamples, total *N* was measured by micro-Kjeldhal digest and steam distillation. The other mineral elements were determined after calcination at 550 °C. The ashes were

Table 3
Heavy metal input (g ha^{-1})

	Zn	Cu	Pb	Cr	Ni	Cd	Cu+Zn+Ni+Cr
Sludge	3660	2090	550	280	180	20	6210
17 Triple phosphate	48	17	0.2	29	5	3	99
Maximum cumulative inputs allowed over a 10-year period[a]	45000	15000	15000	15000	3000	15000	60000

[a] Limit values allowed by French legislation (since January 1998).

taken up in hydrochloric acid. Cationic macroelements were analyzed with the atomic absorption spectrometer, microelements, by ICP-MS, and P, by ceruleomolybdic reaction using a Unicam SP 1800 spectrocolorimeter (Unicam, Philips, France).

Treatment effects were analyzed using variance analysis (ANOVA). Multiple comparisons were made by Tukey's test, and the differences were considered significant when the probability given by the matrix of pair-wise comparison was below 0.05 ($P<0.05$).

Fertilization efficiency was assessed using the difference between N or P taken up by maize in control and in amended plots. Thus, the fertilizing efficiency of the inorganic fertilizer and the sludge can be quantified with $RE_{N(\text{or P})}$: apparent recovery efficiency of the applied N or P [kg N(or P) taken up kg^{-1} N(or P) applied] in agreement with Binder et al. (2002).

$$RE^{n}_{N(or\ P)} = \left[UN^{n}(or\ P)_{+N(or\ P)} - UN^{n}(or\ P)_{+0N(or\ 0P)}\right] / N \quad (1)$$

where UN^n (or P) is the plant N(or P) accumulation in aboveground biomass (kg ha^{-1}), N(or P) is the amount of inorganic or sludge-N(or P) applied (kg ha^{-1}), and the subscripts +N(or P) and +0N(or 0P), respectively, refer to treatment with and without N(or P) application. The exponent n refers to the number of years after sludge application: RE^1_N and RE^2_N were calculated for the first and the second crops, respectively, following spreading, while for phosphate, the calculation was only done the first year, i.e., RE^1_P.

Soil drainage water was also monitored: The outflows were measured hourly at the two outlets of the pipe drainage network from plots I and S. Outflows were then cumulated over 1 day to get daily values. At the same time, daily composite samples were collected in 500-ml polyethylene Nalgene flasks (125 ml were automatically pumped every 6 h) from May 1997 to February 1999. The first daily subsamples were filtered at 0.2 μm, then major cations and anions were detected by ionic chromatography (Dionex DX-100). Phosphate and ammonium concentrations were beneath the detection limit (<1 mg l^{-1}) in all samples. After filtration, second daily subsamples were acidified (2% double-distilled HNO_3), and Fe, Mn, Cu, Zn, Cr, Ni, Pb, Cd were analysed with ICP-MS during the first drainage season following the sludge application.

3. Results and discussion

3.1. Soil properties

The cation exchange capacity remained low in the upper horizon (from 5.67 to 6.85 $cmol_c\ kg^{-1}$ soil) over the whole experiment. In the same way, the soil pH was not affected by sludge application and stayed at around 6.2 in plot S while it increased slightly from 5.9 to 6.4 between April and October in the control plot (Table 4). This change was probably a seasonal phenomenon due to the crop because, in the long term, the natural evolution of these soils is the acidification of the top horizon (Gavalda et al., 2001).

Table 4
Agronomic properties of the soil (0–30 cm) before and after sludge spreading (means and SD for 3 replicates)

	April 1997	October 1997
pH_{water}		
Control plot	5.87±0.068 A	6.36±0.087 B
Inorganic fertilized plot	5.80±0.161 A	5.86±0.154 A
Sludged plot	6.17±0.096 B	6.38±0.122 B
Total organic carbon (g kg^{-1} soil)		
Control plot	6.89±0.113 A	7.54±0.225 AB
Inorganic fertilized plot	7.02±0.599 A	8.02±0.520 AB
Sludged plot	7.65±0.960 AB	8.26±0.472 B
Total N (g kg^{-1} soil)		
Control plot	0.755±0.010 AB	0.692±0.013 B
Inorganic fertilized plot	0.779±0.037 A	0.755±0.045 AB
Sludged plot	0.811±0.038 A	0.825±0.019 A
Cation exchange capacity ($cmol_c\ kg^{-1}$ soil)		
Control plot	5.78±0.459 AB	6.03±0.235 AB
Inorganic fertilized plot	5.67±0.759 A	5.83±0.677 AB
Sludged plot	6.63±0.309 B	6.85±0.231 B
Olsen P (mg kg^{-1} soil)		
Control plot	20.0±1.00 A	26.7±5.13 AB
Inorganic fertilized plot	27.1±4.34 AB	28.0±3.50 B
Sludged plot	30.3±3.88 B	37.2±3.87 C
DTPA-extractable Cu (mg kg^{-1} soil)		
Control plot	1.02±0.00 AB	0.90±0.01 A
Inorganic fertilized plot	1.27±0.15 B	1.19±0.17 AB
Sludged plot	1.07±0.13 AB	1.24±0.15 B
DTPA-extractable Zn (mg kg^{-1} soil)		
Control plot	0.50±0.00 AB	0.45±0.03 A
Inorganic fertilized plot	0.71±0.09 B	0.58±0.06 AB
Sludged plot	0.71±0.12 B	0.74±0.11 B

Means with letters in common are not significantly different ($P>0.05$).

Another seasonal modification seemed to affect organic matter: The levels of carbon insignificantly increased, while nitrogen insignificantly decreased in the three plots from spring to autumn. Analyses conducted on humic and fulvic acids by Guiresse et al. (2004) during this experiment showed that the structure of the humic substances was not affected by sludge.

The average total heavy metal contents of the soil, reported in Table 1, remained low in the three plots during this experiment. Heavy-metal input from sludge was around 10% of the maximum cumulative input over a 10-year period according to French legislation (Table 3). The greatest amounts of metals supplied by sludge and inorganic fertilizer were Zn and Cu, which are essential micronutrients for plants. Soil total Zn and Cu concentrations remained around 28 and 7 mg kg^{-1}, respectively, without any significant increase. Such levels are not far from deficient values according to Alloway (1992), Baize et al. (1997) and Morera et al. (2001). Moreover, the available Cu and Zn levels were not increased by sludge application (Table 4): DTPA-extractable Cu at about 1.0 mg kg^{-1} and Zn at about 0.7 mg kg^{-1} soil were similar to values obtained by White et al. (1997) on degraded semiarid soil. Although Cr and Ni, which are biologically useful, were respectively almost 10 and 4 times more abundant in sludge than in inorganic fertilizer (Table 3), total soil concentrations were not enhanced by sludge addition and remained low: 16 and 8 mg kg^{-1}, respectively. The same remark can be made for other potentially harmful metals (Pb and Cd), whose total soil contents stayed at about 10% of the limit values: around 11 and 0.14 mg kg^{-1}, respectively. DTPA-extractable Cr, Ni, Pb and Cd were beneath the detection limits in soils from both plots.

In contrast, the soil P-extractable content was enhanced by sludge supply because Olsen P was higher in plot S than in the others after spreading (Table 4). As Frossard et al. (1996) and Rydin (1996) showed, the heat-drying process did not prevent sludge-P from being released into the soil or from being taken up by plants, as detailed in the next part.

The last soil parameter, which actually differed between sludge or inorganic fertilization, was nitric nitrogen. Indeed, 200 kg ha^{-1} NO_3^-N was found in the soil of plot I in July and 50 kg ha^{-1} in September, while very little was detected in plot S, like in the control (Fig. 1). Moreover, nitrate leaching must be taken into account.

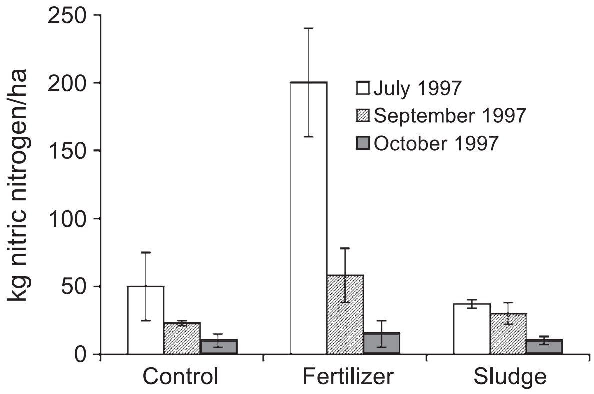

Fig. 1. Evolution of soil nitric nitrogen in the upper soil (0–30 cm).

3.2. Soil drainage water quality

In these drained and irrigated soils, leaching mainly occurred during an intense drainage period lasting from 1 to 3 months in winter. Over the past 20 years, the mean drainage is around 150 mm/year. The winter of 1997–1998 was not very wet, hence, the main drainage was reduced to 40 mm in January 1998. Besides the main drainage event, minor flows occurred throughout the year, especially during irrigation: All the flows were taken into account to evaluate N leaching in both plots I and S. Two days after sludge spreading, 18 mm of rain led to the first drainage flows, whose NO_3^- concentrations were around 55 mg l^{-1} in both plots. At the beginning of June 1997, water drained from plot S was more concentrated (70 mg l^{-1}), while water from plot I remained constant. But from autumn 1997 onwards, the trend was reversed: Drainage water from plot S was slightly less concentrated than that from plot I. Cumulated outflow during the first summer (1997) and the 1997–1998 winter was around 80 mm, for a flow-weighed mean nitric concentration of 55 mg l^{-1} (minimum was 30, and maximum, 90). For this period, the amount of leached nitrogen was 10 and 11 kg N ha^{-1} for plot I and plot S, respectively (Table 5). Globally, the slightly higher N leaching in plot S was due to the slightly higher outflow, the drain pipe being narrower. Winter flow rates and N leaching

Table 5
Cumulated drainage and N leaching

Period	Plot	Cumulated drainage (mm)	N leaching (kg ha^{-1})	Flow-weighted mean concentration (mg NO_3^- l^{-1})
Summer 1997	Plot I	39	5	56
(06/01/97–09/01/97)	Plot S	45	6	57
Winter 1997–1998	Plot I	41	5	55
(12/09/97–04/27/98)	Plot S	40	5	52
Summer 1998	Plot I	59	3	24
(05/18/98–09/27/98)	Plot S	62	3	20
Winter 1998–1999	Plot I	32	2	22
(12/03/98–02/05/99)	Plot S	32	2	30

were greatly reduced by the ryegrass because the amount of N leaching in previous winters was between 10 and 60 kg ha^{-1} (Guiresse et al., 1995). Moreover, the ryegrass biomass harvested in May 1998 was 3.1 Mg DM ha^{-1} in plot S, while it was 4.28 Mg DM ha^{-1} in plot I. Obviously, the ryegrass took up less N in plot S than in the others, indicating that the large sludge supply the previous spring did not increase soil winter nitrogen release and, hence, was not an added source of leachable N.

The trace metal element concentration of drained water was monitored over the first main rainy event after sludge supply. This drainage event happened from December 12th 1997 to January 16th 1998. Indeed, the flow occurred after six dry months, where only irrigation maintained soil moisture. Likewise, sludge pellets disappeared from the soil surface after October 1997. Given the release of trace metals that could occur following the mineralization of the sludge, analyses were focused on this drainage event. During this period, outflow rates did not follow a permanent regime. During rain, they quickly increased to a maximum: 1600 l h^{-1} ha^{-1} in plot S and 1000 l h^{-1} ha^{-1} in plot I. Such flow rates occurred only during an hour, when the water table was in the upper horizon, where hydraulic conductivity is maximum. After the rain stopped, outflow gradually decreased with the discharge of the water table within the soil profile. At the same time, the chemical composition of the drained water also changed according to the path taken by the water inside the soil. High flow rates leached elements that were more abundant in the upper horizon, like K^+, although water drainage during the discharge stage rather contained elements of the lower horizons, like Na^+ and Cl^-. Exactly the same remark can be made about metals. Gavalda et al. (2003) studied the pattern of heavy metals (total and available contents) through the whole profile. Some metals were more available in the upper than in the lower horizons, e.g., Cu and Mn, hence, their concentration peaks coincided with those of flow rate. Other elements, especially Pb and Cr, exhibited such a low mobility over the whole profile that their concentrations in the drainage water remained very low, whatever the flow rate. In contrast, levels of available Zn, Ni and Cd did not show a gradual decrease with depth, and their levels in the drainage water changed along the drainage event. These results were true for both amended plots, where the curve of drained water composition plotted against time was very similar. No significant difference appeared between mean drained water levels of Ni, Cu, Fe or Mn for the two plots (Table 6). Mean Pb and Cr contents were slightly higher in water drained from plot S than from plot I, but the opposite was noted for Zn and Cd. However, the amounts of heavy metals involved were so low that it was difficult to conclude if they were due to the type of amendments used or to slight changes in the hydrodynamic and geochemical characteristics of the two plots. It can be noted, however, that the levels in drainage water of the eight micronutrients monitored in the present work were between 2- and 20-fold lower than those found by Planquart et al. (1999).

3.3. Crop yields and recovery of elements

The total aboveground biomass produced the first year (1997) in plot S was not significantly higher than

Table 6
Heavy-metal leaching in drained water over the drainage event from 24/12/97 to 16/01/98

	Mean concentration (μg l^{-1})			Flow-weighted mean concentration (μg l^{-1})		Total amount leached (mg ha^{-1})	
	Plot I	Plot S	Difference	Plot I	Plot S	Plot I	Plot S
Fe	86.5	79.1	NS	87.5	82.8	16889	17619
Mn	0.04	0.04	NS	0.04	0.04	8	8
Cu	3.02	2.81	NS	3.06	2.72	590	579
Zn	1.69	0.74	*	1.62	0.77	314	163
Ni	1.78	1.66	NS	1.81	1.8	349	383
Cr	1.28	3.4	*	1.18	3.01	228	641
Pb	0.03	0.05	*	0.03	0.06	6	14
Cd	0.02	0.01	*	0.02	0.01	4	1

NS: no significant difference.
* Significant difference (P<0.05).

in inorganic fertilized plot I. Both plots gave maize yields about 30% above the control. As regards the macronutrient levels (Table 7), foliar N contents in plot S were lower than in plot I. This slight deficiency affected grain biomass, which was 8% lower in plot S than in plot I. Foliar N content was positively correlated (r^2=0.81, P<0.05) to grain yield, whereas P content was negatively correlated, indicating that N was a limiting factor but P was not. As a result, the efficiency of added N was higher than that of added P. Moreover, the apparent recovery efficiency of N from inorganic fertilizer (FRE_N^1) was 0.6, while that of sludge (SRE_N^1) was 0.27. These recovery efficiencies are greater than those of Smith and Tibett (2004) with undigested liquid sludge and anaerobically digested, mechanically thickened sludge. The ratio (SRE_N^1)/(FRE_N^1) allowed to approach the rate of available N released the first year by sludge and as taken up by maize as an inorganic fertilizer: SRE_N^1/FRE_N^1=0.45. This means that the efficiency of sludge-N was almost half of that of inorganic fertilizer-N one. Thus, the heat-dried sludge was more N-efficient than digested or limed sludge because the ratios for the latter, reported by Coker (1983), were about 0.13 and 0.28,

Table 7
Yield and composition of maize (per gram dry weight, means and S.D. for three replicates)

	Control plot	Inorganic fertilized plot	Sludged plot
Total aboveground biomass (Mg ha^{-1})	14.7 (2.02) A	20.9 (1.74) B	22.6 (2.17) B
Maize grain yield (Mg ha^{-1})	5.5 (1.01) A	9.9 (1.73) B	9.2 (2.08) C
Grain N content (% DM)	0.98 (0.045) A	1.42 (0.071) B	1.35 (0.121) B
Foliage composition			
Dry matter basis			
N%	0.34 (0.055) A	0.85 (0.113) B	0.61 (0.071) C
P%	1.44 (1.121) A	0.85 (0.079) B	1.01 (0.137) C
K%	1.38 (0.104) A	2.02 (0.163) B	1.69 (0.184) C
Ca%	0.34 (0.070) A	0.39 (0.045) A	0.41 (0.107) A
Mg%	0.12 (0.010) AB	0.13 (0.011) A	0.11 (0.015) B
Ash%	6.73 (0.422) A	7.71 (0.563) B	7.34 (0.469) AB
Fe (mg kg^{-1})	119 (56.6) A	120 (43.2) A	154 (74.2) A
Mn (mg kg^{-1})	38.6 (5.13) A	39.3 (7.13) A	32.2 (9.01) A
Cu (mg kg^{-1})	0.78 (0.523) A	3.33 (0.633) B	3.14 (1.054) B
Zn (mg kg^{-1})	3.89 (0.690) A	3.94 (0.489) A	5.32 (0.552) B
Ni (mg kg^{-1})	0.26 (0.196) A	0.30 (0.135) A	0.33 (0.231) A
Pb (mg kg^{-1})	0.22 (0.050) A	0.37 (0.123) AB	0.45 (0.071) B
Cd (mg kg^{-1})	0.013 (0.0058)A	0.043 (0.0087)B	0.028 (0.0075) C

Means with letters in common are not significantly different (P>0.05).

respectively. The model previously proposed by Labrecque et al. (1998) was verified and improved by our experiment:

$$\text{Sludge available N in first year} = \text{Sludge inorganic N} + 0.33\ \text{Sludge Organic N} \quad (2)$$

According to the composition of sludge (Table 2), Eq. (2) gives 227 kg available N in the first year. In the same time, the above ratio (SRE_N^1/FRE_N^1=0.45) multiplied by the total sludge-N gives 225 kg N for the same parameter.

The decrease of water in the pellets was paralleled by a decrease in ammonium due to volatilisation. According to the literature (Coker, 1983; Labrecque et al., 1998) and to our results (2), inorganic N content is the main parameter involved in pellet N-efficiency towards crops. In a heat-dried sludge tested by Labrecque et al. (1998), a water content of 7.3% corresponded to 0.43% of inorganic N content. In the pellets that we studied, 0.83% NH_4^+–N remained. As a result, this slightly less dewatered sludge was more N-efficient towards maize.

One year after spreading, the residual effects of sludge were evaluated by harvesting unfertilised areas: 51 kg N ha^{-1} were taken up by maize in plot I and 85 kg in plot S. The difference was due to the residual effect of the pellets: 34 kg ha^{-1} in regards to 500 kg previously supplied by sludge, which leads to SRE_N^2=0.068%.

Because P was not a limiting factor in this experiment, the apparent efficiency of the inorganic fertilizer was low (FRE_P^1=0.13) like that of sludge (SRE_P^1=0.06). Thus, the P-efficiency of sludge was almost half that of inorganic fertilizer (SRE_P^1/FRE_P^1=0.46). These results were very close to those of Pommel (1995), who reported that heat-treated sludge mixed with a loamy soil induced a relative efficiency of 0.4 compared with monocalcium phosphate.

Because the microelements were more abundant in leaves than in grains (except Zn), only leaf contents are presented in Table 7. Gigliotti et al. (1996) also detected Zn, Cu and Ni in larger amounts in maize leaves and stalks than in grains and roots. Gardiner et al. (1995) and Planquart et al. (1999) obtained the same results concerning, respectively, Cu, Ni and Cd in barley and Cu and Zn in colza. Amongst trace metals, foliar Cu concentration was the most closely correlated to grain yield, indicating that this element could be deficient. Nevertheless, grain Cu concentration was about 1.5 mg kg^{-1} (without any difference in the three plots), which was fivefold lower than previous results obtained in maize by Henning et al. (2001), in wheat and bean by Bhogal et al. (2003), and in pea by Krebs et al. (1998). Only Zn was more concentrated in the grains (18 mg kg^{-1}) than in the leaves. Although foliar Zn levels were significantly higher in plot S than in either of the others, they were between two- and fivefold lower than those reported by Henning et al. (2001) for maize and by Barbarick et al. (1997) for wheat. Concurrently, no other trace metals were enhanced by sludge amendment, as seen in Table 7, and were lower than those reported by all the above authors. It can be noted that the potentially most harmful element (Cd) was 10-fold less abundant in maize grains (0.003 mg kg^{-1}) than in leaves and nearly 1000-fold less than in many vegetables analysed by Jinadasa et al. (1997) in Australia. It was important to check metal levels because, in this area, the whole maize plant is frequently cropped as fodder for dairy cows. In the long term, heavy metals from the milk could accumulate in the food chain. From the data collected in this experiment, we were able to estimate the long-term heavy metal budget in the soil amended with the studied heat-dried sludge.

3.4. Long-term heavy-metal budget

A few assumptions must be made to establish a balance between the heavy-metal input in the soil (atmospheric emissions and fertilizers) and the output (crop uptake and leaching) over a century to answer the question of whether the amount of metals stored in the soils would decrease or increase.

3.4.1. Assumptions about amounts of heavy metals stored in soil (HMS)

Because it is the first horizon involved in transfers, only the upper, tilled horizon was taken into account to calculate the amounts of heavy metals stored in the soil: From 0 to 0.35 m depth, the total soil mass was estimated to be 5250 Mg ha^{-1}. Actually, many field experiments have revealed that the metal load remains in the upper horizons and does not move through the profiles (Williams et al., 1987; Alloway, 1992; Baize et al., 1997), although it depends on time scale and soil nature. The soil contents (SC) of heavy metals used here were those obtained with *aqua regia* digests from samples taken in the whole experimental field before sludge application (April 1997).

$$HMS\ (\mathrm{g\ ha^{-1}}) = SC \times 5250\ \mathrm{mg\ kg^{-1}} \times \mathrm{Mg\ ha^{-1}}$$

3.4.2. Assumptions about heavy-metal input by fertilizers

To simplify the pattern and assess heavy metal input by fertilizers over a century ($HMiF_{100}$), three different amendment processes have been considered: heat-dried sludge, inorganic fertilizer and cow manure. It was assumed that, in a given case, all the fertilization was exclusively supplied by one of the three types of fertilizers. Their composition was also assumed to be constant for 100 years.

3.4.2.1. Assumptions about sludge. The maximum cumulated amount of sludge application allowed by French legislation is 30 Mg DM ha^{-1} over a 10-year period. Thus, over one century, the total metal input was calculated from 300 Mg DM ha^{-1}. The composition of sludge was as described in Table 2.

3.4.2.2. Assumptions about inorganic fertilizer. In the field experiment, 240 kg N ha^{-1} was supplied by inorganic fertilizers; it is a common dose applied to maize in South-West France. One third of the total N is, often, 17 triple phosphate and the other two thirds urea. Because urea had a very low metal concentration (Gavalda et al., 2001), only the composite phosphate was involved in this model. Its annual metal input (Table 3) was assumed to be repeated yearly for one century.

3.4.2.3. Assumptions about cow manure. Although the composition of the manure changes along the year for a given dairy farm, it is possible to assess the average dry matter and N content. Dry matter is of about 25% fresh weight, and N content is of about 0.55% fresh weight (Gavalda et al., 2001). In all European areas sensitive towards nitrate pollution, the maximum load of manure-N is 170 kg N ha^{-1} year^{-1} (Amlinger et al., 2003). This amount of organic N corresponds to 7.7 Mg DM manure ha^{-1} and was assumed to be repeated every year for a century. Moreover, metal levels in cow manure can be variable. The mean composition reported by Juste et al. (1995) was used in our model: In mg kg^{-1} DM cow manure, the values were Cu 28, Zn 150, Ni 21, Cr 11, Pb 10 and Cd 0.7. The products of these values by 770 Mg gave the cumulated metal input over one century due to cow manure.

3.4.3. Assumptions about heavy-metal uptake by crops

Mean metal uptake by maize harvested in plots S and I in 1997 was taken into account in this balance for the three fertilizers studied. Total metal output by crops over 100 years ($HMoM_{100}$) was calculated from maize composition and yields, which were assumed to be constant. Evolutions of trace metal concentrations in crops and binding with time are not taken into account.

3.4.4. Assumptions about heavy-metal output in drained water

Heavy-metal output in leachate taken for the long-term balance was the mean value between plots S and I for mean climatic conditions; mean annual drained water was taken to be 150 mm, in agreement with Guiresse et al. (1989). As the drainage network was at 0.8-m depth, these data concerned more than just the upper horizon, enabling the calculation of the total heavy metals leached ($HMoL_{100}$). Losses in run off and deep seepage (beneath pipe drain) were assumed to be insignificant.

3.4.5. Assumptions about heavy metals from atmospheric deposition

We did not measure heavy-metal deposition from the atmosphere in this field experiment, and any estimation from literature data is problematic; levels vary over a wide range in space and time. Even in the present case, without any close big cities or industrial sites, atmospheric deposition can change from 0.8 to 250 g ha^{-1} for Pb and from 0.7 to 3.6 for Cd (Baize et al., 1997). Bourrelier and Berthelin (1998) proposed values for rural areas in South-West France of 60 Cu, 30 Zn, 35 Ni, 125 Pb, 3 Cd and 0 Cr in g ha^{-1} year^{-1}. We assumed that these values remained steady over one century to assess heavy metal atmospheric emissions reaching the soil ($HMiA_{100}$).

3.4.6. One century heavy-metal budget

The differences between the heavy metal input and output over one century indicates the evolution of HMS as follows:

$$\Delta_{100}\mathrm{HMS} = HMiF_{100} + HMiA_{100} - HMoM_{100} - HMoL_{100} \quad (3)$$

Δ_{100}HMS was calculated for Cu, Zn, Ni, Cr, Pb and Cd (Table 8). Whether the maize was cropped for grain or for fodder, the results were very close, except for Zn, which was exhausted easier when the whole plants were harvested and inorganic fertilizer used. Zn was also the only element whose soil storage decreased; for the other five elements, Δ_{100}HMS was positive for the three types of fertilization. Metal amounts added to the soil by various fertilizers were not all in the same range. First, some results were in the same range as the coefficient of variation (CV) of HMS, indicating that the metal increase was not significant with respect to natural variability. This was the case for Cu, Ni and Cr due to inorganic fertilizer. Second, other increases were above the CV of HMS but not very far from it: Ni and Cr supplied by sludge and Cr by cow manure. Third, soil storage of Cu, Zn and Ni were significantly enhanced when sludge or cow manure was applied. This result is not very worrying because these elements are useful for crops, and their initial soil contents were very low. In contrast, the last two elements (Cd and Pb) have a high potential toxicity and their amounts in the soil increased over one century at rates very close for the three fertilizers.

4. Conclusion

In this experimental study, the plot amended with heat-dried sludge produced the same maize yield as the plot receiving inorganic fertilizer. The ratio inorganic fertilizer-N replacement value was 0.45 the first season following sludge application and 0.07 1 year later. This high N efficiency might be due to sufficient humidity (9.5%) of the pellets to keep a relatively high level of ammonium. The sludge-drying process usually takes into account the fact that a low moisture content inhibits microbial activity. However, maintaining a certain level of residual water in the pellets gives a more efficient N-fertilizer. Furthermore, except for a slight nitrogen deficit, the macro- and micronutrient contents of the maize dry matter were not significantly different between the two types of input: sludge or inorganic fertilizer.

Although heat-dried sludge is a good N-fertilizer for maize, it did not increase nitrate leaching or the nitric nitrogen stock of the soil. This point must be underlined because sludge is spread in many areas sensitive for nitrate pollution. It is important to note that even 11.1 Mg ha^{-1} heat-dried sludge (in dry weight) did not enhance nitrate leaching during the two following winters.

Heat-dried sludge was also a valuable source of P for maize because, in comparison to 17 triple phosphate, about 50% of the sludge-P was available for the first crop. However, P was not limiting factor in these Alfisols, and the apparent efficiency of 17 triple

Table 8
Evolution over one century of heavy metals stored in the upper soil (0–30 cm)

Heavy-metal soil storage HMS				Δ_{100}HMS for sludge-amended soil				Δ_{100}HMS for inorganic fertilized soil				Δ_{100}HMS for cow-manure-amended soil			
	Mean g ha^{-1}	S.D.	CV %	Fodder maize		Grain maize		Fodder maize		Grain maize		Fodder maize		Grain maize	
				g ha^{-1}	%	g ha^{-1}	%	g ha^{-1}	%	g ha^{-1}	%	g ha^{-1}	%	g ha^{-1}	%
Cu	39008	3203	8	+56444	+145	+60279	+155	+1753	+4	+5588	+14	+21573	+55	+25408	+65
Zn	149100	7875	5	+77960	+52	+84878	+57	−16081	−11	−9163	−6	+94599	+63	+101517	+68
Ni	43260	2835	7	+7429	+17	+7809	+18	+3078	+7	+3458	+8	+18782	+43	+19162	+44
Cr	82425	3150	4	+7140	+9	+7142	+9	+2545	+3	+2547	+3	+8131	+10	+8133	+10
Pb	59325	5775	10	+26872	+45	+27343	+46	+12043	+20	+12515	+21	+19721	+33	+20193	+34
Cd	735	105	14	+726	+99	+767	+104	+530	+72	+571	+78	+792	+108	+834	+113

phosphate was low: 0.13. Nevertheless, soil Olsen-P content was higher in the sludge-amended plot than in the others after the first crop, indicating that the pellets improved soil P status even in a single application. Sludge application levels are currently based on their N content, while P should also be taken into account, particularly when soils are not P-deficient and subject to runoff and, hence, loss of P towards rivers.

In this experiment, a single application of heat-dried sludge did not have any effect on the heavy-metal content of maize. In comparison with the literature, the content was very low, reflecting the already low soil metal levels. Although nothing was visible over 1 year, an extrapolation of legal practices showed that, over one century, soil Zn, Ni and Cr stocks would increase slower in sludged soil than in cow manure amended soil, according to our results issue from a specific sludge application added with data from literature. It must be underlined that the soil storage of the two most potential toxic elements, Cd and Pb, increased for the three types of fertilization considered: inorganic, heat-dried sludge or cow manure. The impact of inorganic fertilizer was significant but lower than the two others. Heat-dried sludge had a strong impact on soil-stored Cu, but these soils were so Cu deficient that even if their content became 1.5-fold higher after a century, it would not represent an environmental risk. Such a conclusion is surprising because no experimental studies report an increase of Pb and Cd soil storage due to inorganic fertilizer. Actually, it is very difficult to monitor soil metal levels for long-term fertilization practices. Numerous authors have reported significant differences between theoretical soil concentrations, calculated from inputs, and field data. Lateral redistribution of the trace metals can probably explain such differences. Monitoring of fertilized and manure- or sludge-amended land must be carried, on but also that of soil water, rivers and sediments. To assess the environmental hazards of these agricultural practices, ecotoxicological assays could be conducted on solutions collected in actual field conditions, which might be very different from the aqueous extracts of soil–sludge mixtures studied by, for instance, Chenon et al. (2003). Such investigations are currently underway in the Garonne valley in France.

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