Impact of Sewage Sludge Spreading on Soil and Crop quality – Results from a French thorough Survey

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Summary

The spreading of urban sewage sludge is a constant cause of controversy, in particular as this practice is known to supply amounts of potentially toxic trace metals to the soil, particularly cadmium. In order to clarify some parts of this debate, this article presents a synthesis of the results of many studies carried out in France on the impact of sewage sludge spreading: old experiments as well as more recent trials. This paper deals mainly with cadmium but sometimes some interesting results relating to other trace metals are reported. Presented data are relative to: i) the soil quality such as total trace metal contents and results of partial extraction aimed at determining the most phytoavailable fractions; ii) the composition of cereal grain, chiefly wheat. Depending upon the period, the experiments were completely different as far as the tonnages of sludge applied and the quantities of cadmium involved. Three categories of trials stand out: 1) During the 1970s and 80s, sludges with a high trace metal content, especially cadmium, were spread at one experimental farm on sandy soils and in the Vexin area on silty topsoils. In all cases, a notable long-lasting impact was found on the total Cd content of the topsoil and the composition of cereal grains. The quantities of applied Cd were very high, from 3,600 g ha^{-1} to 641,000 g ha^{-1}. 2) Sludges containing high amounts of industrial Ca were spread on acid soils in the Limoges region for more than 20 years. Increases of the cadmium content in the topsoil were only measured in fields where the cadmium input was the highest, of 300 to 600 g per ha. A clear increase in the Cd content of cereal grains has been noted as well. 3) During the 1990s and 2000s, numerous experiments with sewage sludge applications compatible with the French regulations were implemented. Local authorities launched many, relatively light, experiments in diverse areas and on diverse soil types. Research institutions carried out other, more exacting, trials. The quantities of Cd input varied from 0.6 to 270 g ha^{-1}. In these cases, no impact was measurable on the composition of cereal grains.

Key words

cadmium, cereal grains, phytoavailability, sewage sludge, soil, total content

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Introduction

Trace metals – cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb), zinc (Zn), etc. - are unwanted though often unavoidable constituents of urban sewage sludge. As some of these metals such as Cd, Pb, and Hg are potentially toxic and have no agronomic interest, their presence generates a certain worry in France as well in all Europe. This is perfectly understandable as some sludge is spread over soil destined for growing food. However, the term "sludge" covers a multitude of different compositions, depending on whether such residue comes from a small, rural, water-treatment plant or from a huge plant in an industrial conurbation. This illustrates the importance of knowing the exact origin of the sludge and its trace metal concentrations.

Going far beyond the European directive 86/278/CEE, the new French regulation of 1998 firmly urges the sludge producers to improve the quality of their by-product. The two main points of this new ordinance were (i) the lowering of the maximum metal contents in the sludge to be spread, for instance the maximum cadmium content of sludge was 20 mg kg^{-1} DM until the 01/01/2001, then 15 mg kg^{-1} until 01/01/2004 and only 10 mg kg^{-1} since this date; (ii) the limitation of the metal fluxes applied on farmlands over a 10-years period (see Table 3).

Fortunately, the efforts of the past 30 years in this field have borne fruit, as all total trace metal contents in sludge have steadily decreased over time, particularly for cadmium (Figure 1). Such efforts should continue without respite, to reduce the input of metals into agricultural land as much as possible.

Other questions are: what happens to these trace metals once they have been spread on soil? How much do the chemical forms in which these metals occur—and their carrier phases—change over time, when comparing newly spread sludge with 5- to 10-year-old residues (McGrath et al., 2000; Bergkvist et al., 2003)? Which constituents of the receiving soils, such as iron and manganese oxides, clay minerals and organic matter will immobilize such metals? In addition, will the indefinite progressive accumulation of such metals not have a harmful effect in the short- to medium term, i.e. 20-100 years, such as the transfer of metals into plants, i.e. into our food chain (phytoavailability)? Or will, on the contrary, such metals migrate rapidly toward groundwater, because of their high mobility, or toward surface water through runoff and transport of solid particles?

Finally, must we fear a "time bomb" effect at the medium- to long term, i.e. the release of initially immobilized metals under the influence of a major environmental change, such as the progressive acidification of soil abandoned by agriculture?

Hereafter, the results are presented of several experiments, most of which focused on soft wheat. They derive from studies carried out in France by various research institutions and/or Agriculture Chambers. Their common objective was to evaluate the impact of the spreading of sludge from urban waste-water treatment plants under different conditions of composition and volume:

An exhaustive paper on this subject was published in the "Courrier de l'Environnement de l'INRA" (Baize et al., 2006), presenting the protocols and main results of numerous French experiments, in particular the most recent ones, studying the impact of urban sewage sludge.

In the present paper, we refer especially to the older and better known studies that are most often mentioned. In addition, we discuss in particular cadmium (Cd) as this is one of the most toxic trace metals, as well as being among the most mobile and phytoavailable ones. Like mercury and lead, the presence of cadmium is highly dependent upon human agricultural and industrial activities. Except for some well-known and well-localized anomalies, its natural pedogeochemical concentrations generally are very low, around 0.10 to 0.15 mg kg^{-1} in surface horizons, though human input from various sources can easily triple or quadruple such values (Baize et al., 1999).

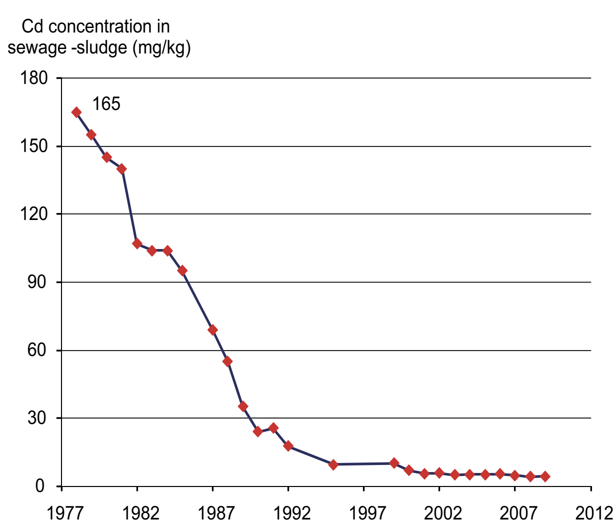

Figure 1. Mean annual cadmium concentrations from 1978 to 2009 in sludge from the treatment plant of waste-waters of Greater Paris (at Achères). This concentration has decreased strongly over time from 160 to 4.3 mg kg^{-1} DM

Three tools for impact assessment

In order to estimate the impact of sewage sludge application, three kinds of rather simple tools are often available:

(i) the total metal concentrations determined in topsoil samples;

(ii) the easily phytoavailable quantities as assessed by "partial" (also known as "single") extractions applied to the same topsoil samples;

(iii) the metal composition of plants cultivated on these soils, especially cereal grains (maize, barley, wheat), the latter being a major product in the human food chain that is of vital importance for the French economy.

Total trace-element contents in soil and soil-plant transfer

In order to be representative of a truly "total" content, an analytical process must be able to extract all chemical forms of the element to be assayed, including those in the silicate lattices. It is thus necessary to use X-ray fluorescence or very strong dissolution methods, such as that using the hydrofluoric acid (AFNOR, 1996) or the alkaline fusion.

Total contents are measurements of stocks at a given time. The repetition of such measurements over different periods at the same point monitors the content and identifies any changes. In contrast with partial or sequential extractions, total contents today are routinely and easily determined and pose no problems of interpretation, regardless of the soil sample properties such as pH, particle-size, presence of carbonates and of iron- and manganese oxides, etc. (e.g. Nirel & Morel, 1990; Tack and Verloo, 1995; McBride et al., 2006). Unfortunately, total-content methods do not distinguish the chemical species, which means that such analyses provide no information about the mobility of the element in the soil, nor of its availability or toxicity toward living organisms (e.g. Rieuwerts et al., 1998a).

As a general rule, there is no direct relationship between the total content of a trace element in the soil and the quantity of this element absorbed by a plant (e.g. Rieuwerts at al., 1998b; Meers et al., 2005). In addition to the chemical forms in which each trace metal occurs, plant absorption is determined by certain soil properties, such as the pH or the abundance of constituents that can easily retain metals, as well as by the physiological and genetic specifics of the plants. Once absorbed by roots, trace metals can accumulate in the roots or move to other parts of the plant, e.g. leaves, stems or grains, where they are stocked. Each plant species has its proper strategy concerning such trace metal absorption and transfer phenomena.

Trace-element determination in plant organs and "partial" extraction

In order to know whether urban-sludge spreading has an impact on the composition of plant organs that are consumed, it is necessary to assay trace metal contents directly in these organs such as wheat or maize grains, spinach leaves, potatoes, etc. In addition, it is needful to know the concentrations of such metals in the same crops during "normal" agriculture, i.e. without sludge spreading. To obtain such references, the AGREDE-QUASAR (1) (Baize et al., 2003) and GESSOL-La Châtre (2) (Baize et. al, 2009) research programmes were set up. These studies on the composition of agricultural products are indispensable for obtaining reliable answers to these questions and they must consider not only the plant species and the cultivar, but also past agricultural practices as well as soil types, which are highly varied in France.

The simplest method for defining trace-metal species in soil uses chemical reagents that dissolve part of the metals in an air-dried soil sample sieved at 2 mm. After this, the metals thus extracted are assayed. In the case of such "partial", also called "selective", extraction, a single reagent is used and the searched-for speciation is essentially "functional" (Ure, 1991; Bermond, 1999). The aim is to extract only the trace-metal forms of interest, i.e. the potentially "mobile" or "phytoavailable" ones, through a judicious choice of the reagent and the operating conditions.

However, this approach of assessing phytoavailability by using partial extraction faces major theoretical objections. It is somewhat presumptuous to try to simulate absorption by means of an instantaneous chemical reaction, *in vitro*, between a reagent and a dried and sieved soil sample. Absorption is a biochemical reaction that occurs at the interface between the soil solution and the roots of a plant, over a period that can take several months

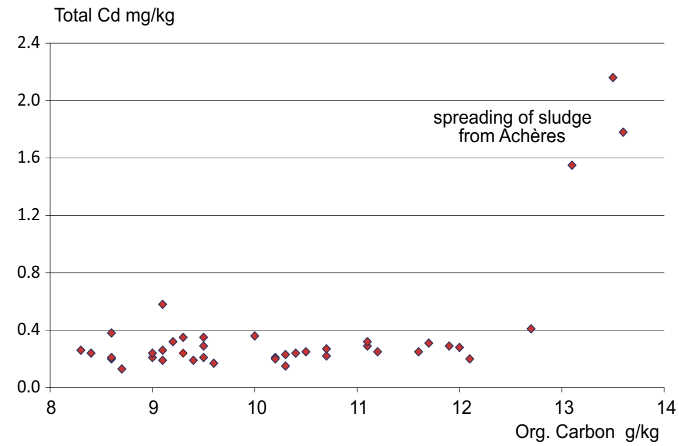

Figure 2. Cd versus soil organic carbon content for 39 surface tilled horizons of a particular soil series (Luvisols developed in loess in the Vexin area). There is no relation between the two variables, but the three sites where sludge was spread in 1970s show a clear cadmium contamination and carbon enrichment

while the plant grows. Another problem is that several methods exist for such assaying work, the protocols (and thus the results) of which are difficult to compare. No method seems to be universal, i.e. valid for all soil types, all elements, and all plants.

Spreading huge volumes of sludge with high trace-element contents during the 1970s and 1980s

Sludge from the Achères plant[1] spread in the Vexin area

Sludge with a high trace metal content from a large waste-water treatment plant at Achères (see Figure 1) was spread on nearby agricultural land because it could be an interesting source of fertilisers. At the time - 1975 to 1986 - no surveillance or regulations existed for this type of practice, when quite large tonnages were spread in some cases. This sludge had so good reputation as fertiliser and humus-rich amendment that it was then sold to the farmers. Twenty two non-experimental fields of the Vexin area were studied by Bernardon (1993), who calculated that during that period one to four sludge-spreading events contributed as high as 0.22 to 4.3 kg of Cd per ha, 6.5 to 40 kg of Pb ha^{-1} , 28 to 189 kg of Zn ha^{-1} , and 8 to 61 kg of Cu ha^{-1} .

Such an input obviously had a clear impact on the total trace metal contents of the receiving soils, especially on a particular soil series (luvisols developed in loess - Baize, 1997). Figure 2 shows that the three surface horizons of soil with a large volume of spread sludge have much higher cadmium content than that usually observed in surface horizon of this soil series (1.55 to 2.16 versus 0.15 to 0.40 mg kg^{-1}). The rare studies of wheat-grain quality, however, did not show a significant "before and after" difference in cadmium content (Bauvois et al., 1985; Tercé et al., 2002).

[1] At the time, the major waste-water treatment plant at Achères processed 90% of the waste water from Greater Paris.

Table 1. Bézu-le-Guéry field trial: total trace metal contents of the surface horizons, measured in 1983 and 1993 (Bauvois et al., 1985; Ducaroir and Cambier, 1994)

	Four plots with sludge (118 t ha^{-1} DM)			Four control plots without sludge		
	1983		1993	1983		1993
	min.	max.	mean	min.	max.	mean
Cd	2.47	3.38	3.06	0.09	0.21	0.12
Cr	56.6	67.7	70.4	27.2	37.6	27.0
Cu	42.0	52.5	49.2	7.0	9.2	7.3
Hg	0.29	0.35		0.04	0.06	
Ni	16.0	20.4	19.8	11.5	15.1	11.1
Pb	43.9	48.5	57.2	18.8	23.6	23.4
Zn	143	169	181	33	46	38

The experiment at Bézu-le-Guéry

Another example is the experiment run by the Agriculture Chamber of Aisne during the 1970s, when sludge from the Achères plant was spread over hydromorphic silty soil near Bézu-le-Guéry in the Haute Brie area (Bauvois et al., 1985). In all, 208 t ha^{-1} of sludge, or 118 t of dry matter (DM) per ha, was applied in two double-dose applications in 1974 and 1977. Knowing that the cadmium content of the sludge was 117 mg Cd kg^{-1} DM, the quantity of cadmium thus spread can be estimated at 13.8 kg ha^{-1}, corresponding to an increase in the concentration of about +3.5 mg kg^{-1} of the tilled surface horizon.

Such experimental conditions that applied huge quantities of metals had a major impact on the total cadmium contents of the surface horizon (and on the other metals – Table 1). The average cadmium content of winter barley grains, grown on four plots with spread sludge and harvested in 1983 (six years after the last spreading), was 0.79 mg kg^{-1}, whereas that of four control plots without spread sludge was only 0.33 mg kg^{-1} (Bauvois et al., 1985).

These spreadings had also marked effects for other trace metals, such as zinc (Figure 3).

Experiments at Couhins (INRA, experimental farm near Bordeaux)

This well-known French experimental site was set up in 1974. It resulted in many publications, the main ones being Juste and Solda, 1977; Legret et al., 1988; Gomez et al., 1992; Juste and Mench, 1992; Bermond et al., 1998. The trial plots were measured only 6 by 3 m, and every treatment was repeated in four plots. The initial objective was to compare control plots that received the following: i) only mineral fertiliser; ii) farm manure (with a Cd content of 0,70 mg kg^{-1} DM); iii) sewage sludge from the plant at Ambarès; and iv) sewage sludge from the "Louis Fargue" plant at Bordeaux.

The Ambarès (small town of 9000 inhabitants) sludge represented "standard" (at the time) urban sludge. It contained 60 mg of Cd kg^{-1} and was spread at the rate of 10 and 100 t ha^{-1} DM every two years from 1974 to 1988. The Louis Fargue sludge was selected for its extraordinary cadmium (and nickel) content because of the presence of a battery factory upstream from the treatment plant. This plant in fact processed more industrial than simple urban waste water. This sludge was spread three times, in 1976, 1978 and 1980 at rates of 10 and 100 t h^{-1}a DM. According to Legret et al. (1988), the maximum cadmium

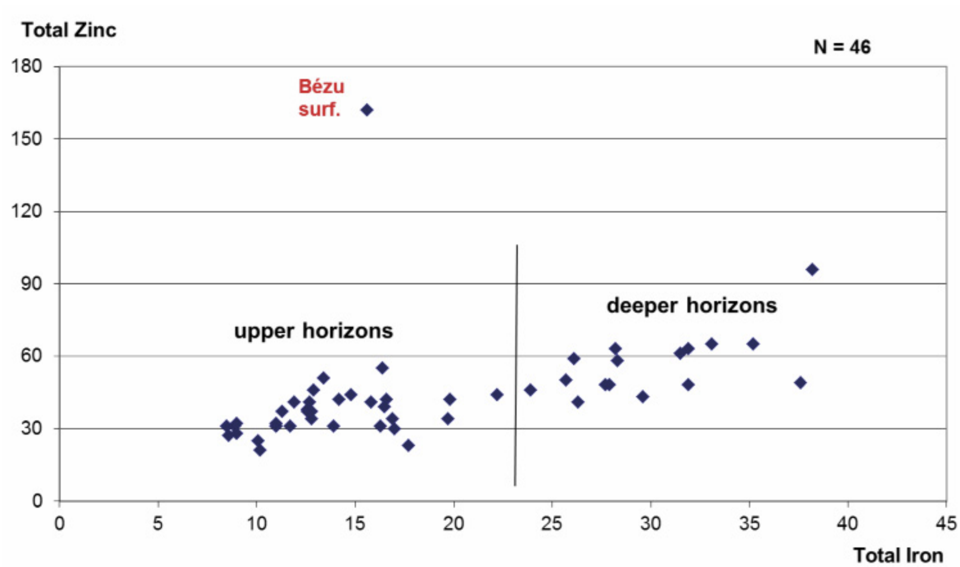

Figure 3. Albeluvisols developed in a loamy aeolian deposit (South-east of the Paris basin). Fourty six samples were taken at different depths from forest and cultivated soils. Upper horizons were taken at 0 to 45 cm depth – deeper ones at 50 to 110 cm depth. Strong natural correlation between total concentrations of Zn and Fe was observed. One sample, outside this correlation is the surface Ap horizon of the experimental plot at Bézu-le-Guéry amended with sewage sludge at high rates (Baize and VanOort, 2014)

Table 2. Couhins experiment – Major results. 50% of the cadmium brought during the 70s is not present today in the first metre of soil. It is gone away in the environment

	Quantity of Cd brought	Total Cd in soil		Cd content in maize grains 1976	
		Before spreading	After spreading	Control	After spreading
Plots with sludge from Louis Fargue	641 kg per ha	< 0.4 mg per kg	94.9 mg per kg	0.17 mg per kg	0.50 mg per kg

content was 2672 mg Cd kg^{-1} for an average of 1830 mg Cd kg^{-1}. Gomez et al. (1992) reported the main features and results of this experiment. They calculated the cumulative cadmium quantities thus spread as:

Ambarès - 100 t ha^{-1} x 8 applications → 27 kg Cd ha^{-1}

Louis Fargue - 100 t ha^{-1} x 3 applications → 641 kg Cd ha^{-1}

The soil of the experimental plots is highly particular. Over at least 1m depth, it contains only 4% clay and >80% sand. Its initial pH was 5.3. Such sandy-gravelly soils are locally known as “grave” (Arenosols according to the WRB). The tested crop was only irrigated maize. Soil properties and plant composition at different growth stages were regularly monitored.

Under these conditions, major impacts were observed in the plots that received a total of 300 t ha^{-1} of Louis Fargue sludge. Firstly, after the third application in 1980, a strong phytotoxic effect was observed on the maize yield which decreased by half. Secondly, the cadmium concentration of the surface horizon determined in 1989 was 94.9 mg kg^{-1} DM versus < 0.50 before the launching of the trial (Table 2). However, Gomez et al. (1992) calculated that about half of the cadmium input was no longer present in the first metre of soil! Part of this missing cadmium may have been evacuated laterally, linked with particles transported by runoff. The trial plots are indeed quite small and those serving as controls contained on average 1.3 mg Cd kg^{-1} in 1989, which is the evidence of a lateral transfer. Massive vertical losses through the soil should not be excluded either, in view of the sandy, quick filtering, and acid character of this soil.

In 1976, in the plots that received two Louis Fargue sludge applications (i.e. 200 t ha^{-1} DM), the mature maize kernels contained 0.50 mg Cd kg^{-1} *vs.* 0.17 mg Cd kg^{-1} in the control plots and those with manure application (Juste and Solda, 1977).

These experiments supplied extravagant quantities of cadmium (up to 641 kg Cd ha^{-1}) on an “extreme”, very sandy and rather acid, soil without any constituents that might fix the supplied trace metals. The impact of such spreading on this soil and the plants cultivated on it is thus not surprising. In the 1990s and more recently, the INRA researchers from Bordeaux have carried out many more experiments on the same plots, studying the best procedures for remediating such polluted soils by means of *in situ* immobilization (e.g. Boisson et al., 1998; Mench et al., 2000; 2002). These old experimental plots today constitute a fascinating research laboratory, even though they do not represent today’s reality of the spreading of sewage sludge on agricultural soils.

Spreading cadmium-rich sewage sludge in the limoges area

In the Limoges area, crop rotation commonly consists of four to five years of temporary grassland, followed by maize for silage fodder and a cereal with straw. The soils in this region are more or less acid, consequently a risk exists of transferring trace metals from topsoils to plants.

In the past, before 1998, the sludges produced by local wastewater treatment plants had high cadmium content, e.g. 20.7 mg kg^{-1} DM on average. The cadmium came mainly from industrial waste related to china manufacturing and decoration. Although since late 1999 such industrial effluents no longer affect sludge quality, the Agriculture Chamber of Haute-Vienne has still carried out a study of soil and soft-wheat-grain quality according to the protocol of the AGREDE-QUASAR programme (Courbe et al., 2002). A total of 36 sites were studied, corresponding to soils developed in metamorphic rocks (leptynite, migmatite and gneiss), and to soils derived from diorite.

Four types of site were distinguished in terms of cadmium quantities supplied by sludge spreading over the preceding 10-years period: 22 control sites without spreading; 2 sites that received less than 100 g Cd per ha, designated below as “small quantities”; 4 sites on diorite that received between 100 and 300 g Cd per ha, designated below as “moderate quantities”; and 8 sites that received between 500 and 600 g Cd per ha, designated below as “large quantities” i.e. much more than the cumulative flow now authorized by the French regulation of January 1998 (150 g ha^{-1} over 10 years). In fact, these fields probably received three times more cadmium over the past 30 years.

In this rural area of little-intensive agriculture, any total cadmium content of soil >0.40 mg kg^{-1} results probably from Man-induced contamination. Of the 36 analysed soil samples, 11 values exceeded this threshold of 0.40 mg kg^{-1} and three contained between 1 and 2 mg kg^{-1} (Figure 4). Eight correspond to plots that received “large quantities” and two others to soil over diorite that received “moderate quantities”.

In the wheat grains, 12 values are ≥0.11 mg kg^{-1} DM, the maximum concentration for cadmium in wheat grains as recommended by the Higher French Council for Public Health (CSHPF). Eight of these were measured in “large quantities” sites and three in “moderate quantities” sites, but this cadmium abundance in the wheat grains seemed to have no relationship with the soil pH.

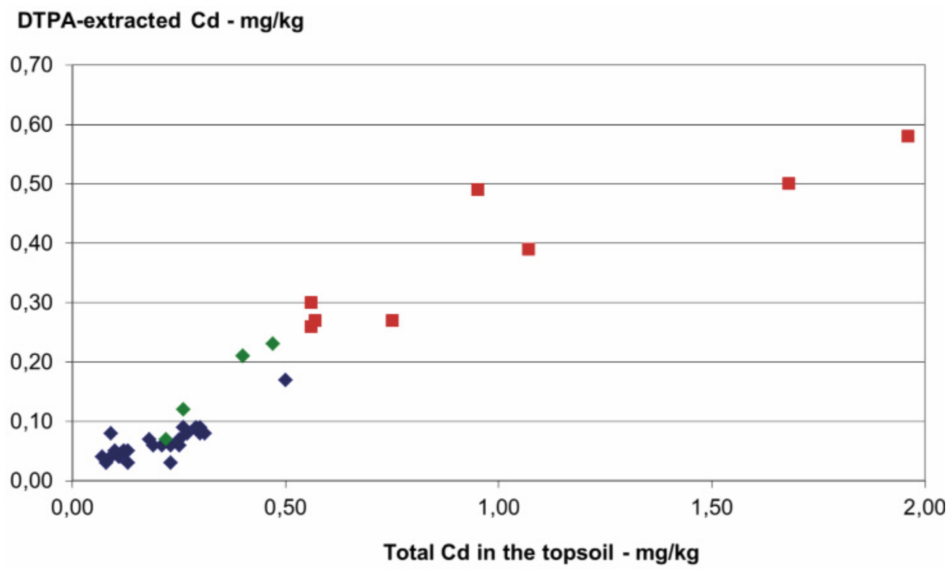

Figure 4. Soils of the Limoges area (36 ploughed horizons). Relationship between cadmium extracted with DTPA and total Cd in topsoil samples. "Large quantities" sites are shown by red squares, "moderate quantities" by green diamonds and "small quantities" and control sites by blue diamonds (Courbe et al., 2002)

The soil samples were subjected to partial extraction with DTPA[2]. The quantities of DTPA-extracted cadmium correlate well with the Cd contents measured in the grains (Figure 5 - correlation coefficient R^2 = 0.71). Beyond the threshold of 0.10 mg kg^{-1} extracted with DTPA, there is a high probability that the Cd concentration in the wheat grains will be equal to or higher than the recommended CSHPF value of 0.11 mg Cd kg^{-1} DM.

The four soils developed from diorite, which received moderate quantities of Cd, have rather low cadmium contents at 0.22 to 0.47 mg kg^{-1}, but Cd concentrations in the grains are as high as those of the eight "large quantities" sites over metamorphic rocks (0.08, 0.13, 0.14, 0.18 mg kg^{-1}). This means that the exogenous cadmium brought to the diorite sites is phyto-available for the wheat, notwithstanding a lower total content in the soil.

In conclusion, the study carried out in the Limoges shows that moderate spreading of sewage sludge containing cadmium quantities compatible with the new regulation (small quantities sites) does not show up in either the soil analyses or the wheat-grain analyses. However, any higher and continuous input of cadmium (moderate quantities sites over diorite and large quantities sites) clearly stands out in both soil and wheat-grain analyses.

The reader will have noted that this quite particular case belongs to the past. Though it is of scientific interest, it questions neither the rules laid down by French regulations, nor the reasonable spreading of urban sludge over agricultural land.

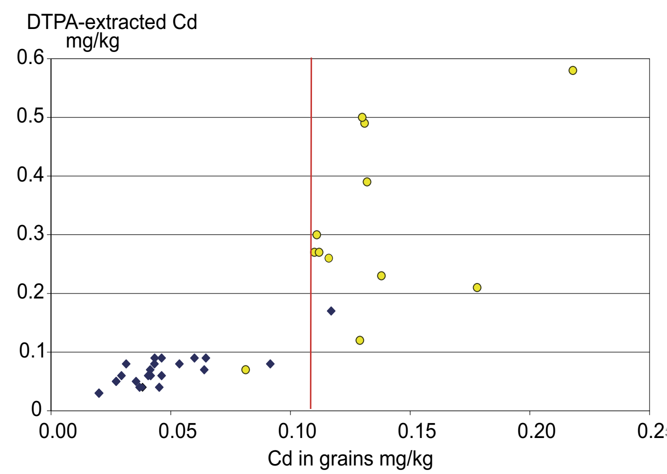

Figure 5. Soils of the Limoges area (36 ploughed horizons). Relationship between cadmium extracted with DTPA from soil samples and cadmium assayed in wheat grains. "Moderate and large quantities" sites are shown by yellow circles; diamonds represent the control sites and the two "small quantities" sites (Courbe et al., 2002). The red line corresponds to the French recommended maximal value

Spreading of sewage sludge over farmland complying with French regulations

AGREDE-QUASAR Research Programme (Baize et al., 2003)

The AGREDE-QUASAR study by INRA in collaboration with several Agriculture Chambers, took place over two periods. First, soil series were studied that had not received sewage sludge, or other urban waste. Located in rural areas, these soils have received steadily phosphate fertilisers and they were not sheltered from possible little atmospheric deposition. Only two cultivars of soft wheat were studied (Trémie and Soissons), and the sampling strategy was soil series oriented. The mature wheat ears were harvested over 1 m^2 and over a widely separated dozen fields for each soil type, after which the trace-metal contents of the grains were determined. At each grain-sampling site, the surface horizon of the soil was taken and characterized by in-depth analyses, including agro-pedological characterization, total trace metal contents, and partial extraction of the metals with three different reagents ($CaCl_2$ 0.01 M – $NaNO_3$ 0.1 M and NH_4NO_3 1 M). This provided references for various soil series with "sludge-free" agriculture.

[2] DTPA = diethylene triamine pentaacetic acid

Table 3. Estimated cadmium quantities reported from experiments presented or mentioned in Baize et al. (2006) (expressed as g ha^{-1}). Comparison with regulation fluxes and amounts supplied by phosphate fertilisers

	Study	Years	Number of spreading events	Cd quantities (g ha^{-1})	
Before 1990				minimum	maximum
Experiments	Couhins – sludge from Louis Fargue	1970s	3		641,000
	Couhins – sludge from Ambarès	1970-80s	8		27,000
	La Bouzule (1974-1981)	1970-80s	2	810	9,180
	Bézu-le-Guéry	1970s	2		13,800
Field reality	Vexin (Bernardon, 1993)	1970-80s	1 to 4	220	4,320
	Vexin – Vélannes (Tercé et al., 2002)	1970-90s	4		3,624
After 1990					
Limoges	Limoges – small quantities	1990s	over 10 years	< 100	< 100
	Limoges – moderate quantities	1990s	over 10 years	100	300
	Limoges – large quantities	1990s	over 10 years	500	600
Others	Barneau and Bouy - simple rate	1990s	2	64	112
	Barneau and Bouy - triple rate	1990s	2	228	268
	QUASAR programme (1999)	1990s	1 to 3	0.8	15
	Burgundy and Franche-Comté	1990s	1 to 4	0.57	21
	Agriculture Chamber of Somme	1990-2000s	4 in 10 years	7	27
	Ensisheim (maize)	1990s	5 in 10 years	25	36
	Colmar	2000s	3 in 5 years	11	23
	Poucharramet (wheat)	2000s	3	2	18
	City of Mayenne	2000s	3 in 5 years		17.6
Legal thresholds	European regulation 86/278/CEE 1986		over 10 years		1,500
	French regulation - *until 01/01/2001*	1990-2000s	over 10 years		300
	French regulation - *from 01/01/2001 on*	2000s	over 10 years		150
P-fertilisers	Amounts supplied by phosphate fertilisers		over 10 years	3	60

During the second period, in 1999, new sites covering the same soil series as before were studied following the same protocol, but which had received sewage sludge at rates in accordance with the current regulations at the time (Cd content of the sludge < 3.5 mg kg^{-1} DM, most often < 1.5).

In all, 163 sites were studied, 33 of which "with sludge", belonging to 11 contrasting soil types; these were soils developed over a large range of sedimentary rocks, including Quaternary silt, river terraces, and marine sediments such as Cretaceous chalk and Jurassic limestone. The sampled fields were located in nine departments of the northern half of France.

In the framework of this programme, the composition of wheat grains could be compared with the analytical characteristics of the soil (surface horizon) where the wheat grows. In the rare other French studies, the analysed grain batches were selected by "production region", and nothing is known about the type and properties of the soils in which the wheat was grown.

The quantities of sludge spread over the trial plots as part of this programme were entirely reasonable and respected the regulations (Table 3). Cadmium input was estimated to be between 0.8 and 15 g ha^{-1} for one or two spreading events.

Partial extraction with neutral salts was carried out on the soil samples, after which the extracted Cd, Cu, Pb and Zn were assayed.

The "with sludge" soil samples did not show any difference from the "sludge-free" ones. The pH was the main factor for explaining the quantities of extracted cadmium and zinc, the highest quantities being obtained for the lowest pH (<6.5). The reverse was true for copper, where the highest extracted quantities were obtained from certain samples with a pH of 8.0 or more. Most of the "with sludge" soil samples had a pH >8.0, but other soils that had the same range of pH values, but were "sludge-free", gave similar results (Baize et al., 2003).

During this study it was thus never possible to show a significant impact of sludge spreading on soil composition (based on partial extraction), or on that of the wheat grains. In the case of soil types with a natural tendency toward acidity (soils developed in sands or loess-like deposits), the highest cadmium contents of wheat grains occurred on soil with the lowest pH, <6.5 as it could be expected because pH is the most important soil factor controlling cadmium uptake (Kirkham, 2006).

Other experiments

A national inventory lists 136 agronomic experiments in France that tested the interest and impact of various residual organic and mineral substances. Not all covered trace metals and some did not concern large-scale farming. Baize et al. (2006) succinctly described eight experiments, all implemented after 1995, some elements of which are shown in Table 3. The soils tested are of different types and the experimental design can be rather simple or quite complex and thorough. The cadmium input varied strongly between trials and even within a single trial, though always remaining between 0.5 to 88 g ha^{-1}. None of the experiments showed a noticeable effect of sludge application on crop quality. Figure 6 provides a good example. Deléan and Kockmann (2003) studied a set of experimental fields in Burgundy and Franche-Comté belonging to various soil types: 15 parcels having never received sludge before were subjected to sludge application on the half of their surface, the other half being used as control plot. Thirteen parcels spread with one or several spreading during the previous five years were also investigated. Analyses of topsoils and wheat grains were carried out (n = 43).

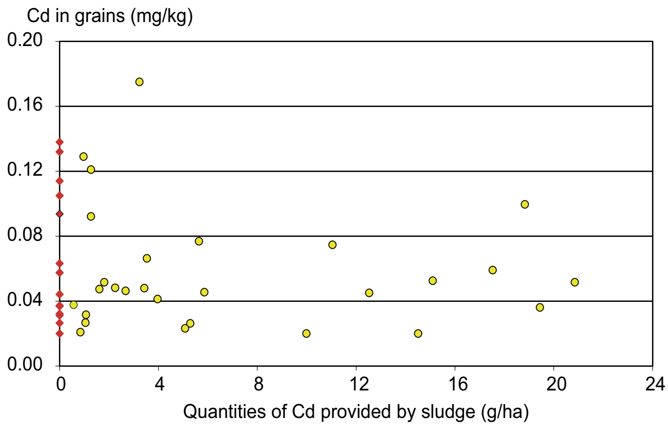

Figure 6. Trials in Burgundy and Franche-Comté (Deléan and Kockmann, 2003). Relationship between cadmium content in wheat grains and Cd quantities provided by sewage sludge application (R^2 = -0.016). Cadmium-input in 15 control plots are nil.

Obviously, no link exists between the cadmium content of grains and the Cd quantity provided via sewage sludge application. Differences observed in grains result much more from the soil properties, such as pH, texture or Fe and Mn oxihydroxides abundance, and from the cultivar grown.

Discussion

It is commonly very difficult to measure the impact of sludge applications on soil. Today, in France, the input of cadmium is very low to minute, which means that any Cd increases in the surface horizon often are smaller than the analytical uncertainty. In this respect, it is useful to remember that:

— To increase a trace-metal concentration by 1 mg kg^{-1} in a tilled horizon weighing between 3600 and 3900 tonnes ha^{-1}), the input of this trace metal must be 3.6 to 3.9 kg ha^{-1} .

— Therefore, the input of 100 g Cd per hectare theoretically will increase the average Cd content of the receiving tilled horizon by only about 0.028 mg kg^{-1} which is obviously of the same order as the analytical uncertainty.

Quantifying the impact of sludge spreading on the chemical composition of consumed organs of plants is not easy as well. This kind of analysis is difficult to perform and only a few laboratories are able to carry out such analyses with a good reliability.

Another, commonly ignored, difficulty is that many experimental fields are pedologically heterogeneous, at least as far as their initial trace metal content is concerned. This is often discovered afterwards. Other problems, equally difficult to identify, such as those related to sampling, sample processing, or analytical work, can again generate absurd post-sludge-application analytical results when compared with the pre-application ones. Generally, such results are clearly unrelated, being far too high or much too low, compared to the real input of trace metal quantities!

Where trace-metal input quantities were enormous (Couhins, Bézu-le-Guéry, La Bouzule, Vexin in the 1970s and 80s), quite clear impacts are visible. These appear in the total metal contents of soils, in the quantities of metals recovered by partial extraction, and in the composition of certain plant organs. Where cadmium input is small and compatible with the requirements of French legislation, it is impossible to demonstrate even the slightest impact on soil or crops. In fact, this is quite reassuring, at least in the short term! The main merit of 1998 French regulation of sewage sludge application has thus been to greatly decrease the trace-metal input into soils. For the most part, this aim was reached.

But, several difficulties remain. First of all, not enough time has passed to have a long-term view of the impact of sewage sludge application at reasonable rate. It is clearly difficult to extrapolate the results from short-term experiments - 4 to 15 years as a maximum - to evaluate the impact of such spreading on the long term. In addition, much more work has to be done on other food crops such as spinach, cabbage and salads, which accumulate trace metal more easily than wheat grains.

The best way to evaluate the possible transfer from soil to plants is to carry out direct analyses of specific organs of the harvested plants. However, such analyses are expensive and delicate, posing problems for many analytical laboratories. This is the main interest of partial extraction with neutral salts or complexing reagents (EDTA, DTPA) on soil samples, in order to make a best-possible estimate of phytoavailability or mobility of trace metals (Baize et al., 2009).

Finally, it should be stressed that the pH of the receiving soil is a major factor influencing the risk of trace metal transfer and especially of cadmium in soil toward plants; the lower the pH, the higher is the risk of phytoavailability and mobility. Fortunately, farmers can easily control this parameter in the medium term by regular application of alkaline calcic amendments.

In fact, limed sludge carries its own antidote against contained trace metals, by increasing the pH of the receiving soils and thus decreasing the potential for mobility and phytoavailability of potentially dangerous metals, at least in the short term. But, the problem is what happens when naturally acid agricultural land, amended with sewage sludge, is abandoned. A progressive re-acidification might lead to desorption and liberation of the metals.

As a last point, we should stress the role played by certain natural soil constituents as powerful trace-metal adsorbers. These include organic matter, clay minerals, and especially iron and manganese oxihydroxides. Different soil series do not present the same capacity of trace metal retention, according to the relative abundance of such constituents in all their horizons, not only in the topsoil. Their vulnerability thus is highly variable and merits to be taken into account in order to plan an optimal location of sewage sludge spreading.

Conclusion

First, a major point should be highlighted: the phrase "sewage sludge application on farmland" does not do justice to the possible colossal differences in metal-quantity input. Such differences depend upon different circumstances, such as the cumulative tonnage that was applied, and the sludge composition that has showed large variations over time and space. Table 3 shows the input of estimated cadmium quantities for the various trials described in this paper. Such quantities go from <1 g Cd ha^{-1} (sludge from small plants with minor trace metal content, spread in little quantities), to 4320 g Cd ha^{-1} (four applications of sludge from the Achères plant in the 1970s and 80s). Worse, in the case of the experiment at Bézu-le-Guéry, two "double-dose" applications supplied 13.8 kg Cd ha^{-1} and the maximum quantity was reached at Couhins Louis Fargue with 641 kg Cd ha^{-1}.

This illustrates the importance of always evaluating as precisely as possible the real input of trace-metal quantities from sludge applications, and to compare this with figures from other types of input, such as phosphate fertiliser, compost, or manure.

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