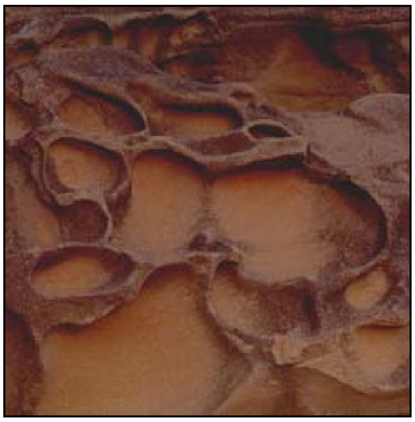
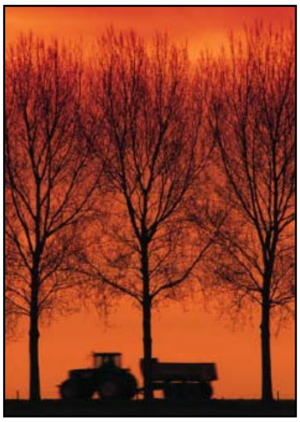
Background values in European soils and sewage sludges

Results of a JRC-coordinated study on background values

Edited by
B. M. GAWLIK and G. BIDOGLIO





PART III

Conclusions, comments and recommendations

B. M. Gawlik , G. Bidoglio

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
Joint Research Centre

2006

EUR 22265 EN

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EUR 22265 EN
Luxembourg: Office for Official Publications of the European Communities

ISBN 92-79-02122-2
ISSN 1018-5593

Printed in Italy

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Abstract

This report summarises the conclusions of two independent, JRC coordinated studies concerning background values of relevant organic micro-pollutants in sewage sludges as well as background values of some trace elements and organic matter in (uncontaminated) soils, in view of a revision of the Sewage Sludge Directive /278/EEC.

The first study, which was conducted by DIN e.V. (R. Leschber) and the second study, which was conducted by BGR (J. Utermann, O. Düwel, I. Nagel), were presented and discussed during two JRC workshop sessions 15-16 April 2004.), done in close collaboration with DG ENV. Attendees of these events were invited via the official channels involving the Environmental Departments and Authorities of the Member States. Numerous additional comments were received and the draft versions of the aforementioned reports were adopted accordingly.

In the following, the conclusions of the JRC concerning the main findings of the two studies as well as recommendations and observations are presented. It has to be stressed that these recommendations and observations are not necessarily those of the authors of the respective studies.

List of abbreviations

Throughout this report the following abbreviations and symbols are used.

AOX	adsorbable organic halogenated compounds
BGR	Bundesanstalt für Geowissenschaften und Rohstoffe
CEN	European Committee for Standardization
COST	European Co-operation in the field of Scientific and Technical Research
DBP	dibutyl phthalate
DeBDE	decabromo diphenyl ether
DEHP	di-(2-ethylhexyl) phthalate
DG	Directorate-General
DIN	Deutsches Institut für Normung
dm	dry mass (dry matter)
EN	European norm
EU	European Union
IES	Institute for Environment and Sustainability
ISO	International Standardization Organization
JRC	Joint Research Centre
LAS	linear alkyl sulfonates
NP	nonyl phenol
NPEO	nonylphenol ethoxylates
PAH	polyaromatic hydrocarbons
PBDE	polybrominated diphenylethers
PCB	polychlorinated biphenyls
PCP	pentachlorophenol
STP	sewage treatment plant
TE	toxicity equivalent chlorinated biphenyls
PCDD	polychlorinated dibenzodioxins/furans

1 Common introduction to the reports Part I, II and III

The European Commission has realized that since the "*Sewage Sludge Directive*" 1986 (EEC) was set into force, a rapid development in the field of the agricultural use of sewage sludge has taken place. On the one hand, the Directive confirmed that those European countries, which had set up legal regulations earlier, were on the right way and, on the other hand, it gave the frame for recycling secondary raw materials with a remarkable content of nutrients and soil improving properties for all EU countries.

Although the Directive set up only guide/limit values for heavy metals, the question whether there might be also harmful effects caused by organic micropollutants has been discussed from this time until now, being initiated and promoted by the COST 681 Action of the European Commission and follow-up activities. The results are revisions of existing national regulations in some countries thus setting up more stringent limit values for heavy metals and introducing new limitations for some organic micropollutants. However, there was no uniform way in handling these problems. Subsequently, in autumn 1999 the European Commission started discussions with governmental representatives of the EU countries as well as with experts/delegates from European economic, technical and scientific organizations. This led to the so-called "*3^rd^ Draft-Working Document on Sludge*" of April 2000, in which general aspects of a long-time improvement of the agricultural use of sludges were laid down.

The document covering proposals for future action contains several Annexes, of which Annex IV includes a table referring to limit values for concentrations of organic compounds and dioxins in sludge for use on land. Since the publication of the 3rd Draft these data have been subject of intensive discussions in the EU and at national conferences.

The following series of reports give some basic information about selected organic micropollutants in sludges as well as about the establishment of background values for some trace elements in soils, susceptible to receive sewage sludge.

2 Background information on the conclusion

Soil protection is becoming increasingly important at European scale. Soil monitoring will be the basis for a European soil protection strategy. The European Commission is aware of the fact that the existing information basis is incomplete. Thus, the Commission made in 2002 the following statement:

"*The actions to be taken in the near future to address soil protection are based on existing information, which is recognised as incomplete. For the long-term protection of soils it will be necessary to ensure the development of a more complete information basis, monitoring and indicators to establish the prevailing soil conditions, and to evaluate the impact of diverse policies and practices*.

One step into this direction was undertaken in the context of the revision process of the Sewage Sludge Directive, when DG ENV asked DG JRC to co-ordinate two studies on background values on trace elements in European soils and organic micro-pollutants in sludges, respectively. Both studies were outsourced by the JRC to renowned specialists in the fields and draft versions of the respective findings of these studies were circulated among the responsible authorities in the Member States as well as in the TWGs concerning the Thematic Strategy for Soil Protection.

Additionally, DG Environment and the JRC organised two scientific workshops on "*Organic compounds in sewage sludge*" and "*Heavy metal background values in EU soils*" in Ispra (Italy) on Thursday 15 and Friday 16 April 2004, respectively.

The main objective of these two workshops was to discuss the draft reports prepared on behalf of the JRC by the external contractors prior to their public release. During the intense discussions the scientific basis and evidence presented in the draft reports was assessed and recommendations were made for their improvement.

Furthermore, upon the circulation of the draft reports additional information and data were received. Together, with the authors of the study as well as the contributing services (Table 1), the JRC reviewed the studies in function of the comments received. Obviously, not all remarks were automatically included in the reports, but were carefully discussed. Where – according to our feeling criticism was justified – the necessary changes were introduced into the text.

The majority of the comments received asked for a clear distinction between the findings of the study and the recommendations made by the JRC. This has lead to this additional part III, which forms also the official opinion of the JRC on the subject.

In any case, the results of this project make a contribution on the above formulated EU policy on soil protection. However, the necessary database has still to be improved. Therefore the continuation of the survey, both for organics and trace elements is urgently advised. The strategies and approaches, as presented in this project should be persecuted, even extended.

Table 1 – Institutions and services contributing to the project on background value. The support is gratefully acknowledged

Country	*Institution/service*	*Contact person(s)*
Austria	- Federal Environment Agency, Vienna	*A. Freudenschuß,*
Belgium	- Ghent University, Department of Geology and Soil Science, Ghent	*E. van Ranst*
	- Direction générale des ressources naturelles et de l'environnement, Jambes	*J. Defoux*
Czech Republic	- Central Institute for Supervising and Testing in Agriculture (CISTA), Brno	*M. Sáňka, P. Němec*
Denmark	- University of Copenhagen, Institute of Geography, Copenhagen	*H. Breuning Madsen*
Estonia	- Geological Survey of Estonia, Tallinn	*V. Petersell*
Finland	- Geological Survey of Finland (GTK), Espoo	*T. Tarvainen*
France	- Institut National de la Recheche Agronomique (INRA), Olivet Cedex	*D. King*
Germany	- Federal Institute for Geosciences and Natural Resources (BGR), Hanover	*I. Nagel, O. Düwel, J. Utermann*
Ireland	- Irish Agriculture and Food Development Authority (TEAGASC), Johnstown	*D. McGrath*
Italy	- Agenzia Regionale per la Prevenzione e Protezione Ambientale del Veneto (ARPAV), Castelfranco Veneto	*P. Giandon*
	- Dipartimento di Scienze dell'Ambiente e del Territorio (DISAT), Milano	*D. Magaldi*
Lithuania	- Geological Survey of Lithuania, Vilnius	*V. Gregorauskiene*
The Netherlands	- Alterra, Green World Research, Wageningen	*D. J. Brus*
Norway	- Norwegian Institute of Land Inventory (NIJOS), Aas	*A. Arnoldussen*
Portugal	- Laboratory for Agricultural Chemistry Rebelo da Silva (LQARS) Lisboa	*R. Mano*
Romania	- Research Institute for Soil Science & Agrochemistry (RISSA), Bucharest	*A. Manea*
Slovakia	- Soil Science and Conservation Research Institute, Bratislava	*J. Čurlík*
Sweden	- Swedish University of Agricultural Sciences, Uppsala	*J. Eriksson*
United Kingdom	- c/o European Commission, DG JRC IES, Ispra	*R.J.A. Jones*
	- National Soil Resources Institute, Cranfield University, Silsoe	*P.J. Loveland*
	- Macaulay Land Use Research Centre (MLURI), Aberdeen	

3 Background of trace elements and organic matter in European soils

3.1 General remarks and annotations

Within the framework of this study it was attempted to compile a Europe-wide evaluation of heavy metal and organic matter contents of European top soils using existing data of various origin. The evaluation programme was accomplished thereby in several steps. The data published in part II of this report outline the current situation of available data in Europe (status 2004), whereby the main focus is set on the latest data request (Phase II, 2nd part).

The participating countries were asked to evaluate available data according to previously established criteria. The evaluated data are provided together with the corresponding meta-information. The evaluation was carried out in relation to three different bases of reference:

- soil parent material (MAT 11 level) and land use,
- soil pH,
- soil texture.

A detailed description of the available data was given. A comparative evaluation of the data was carried out under consideration of harmonisation aspects. As a first result it could be seen that the available data of the participating countries vary, both in quality and quantity. However, a differentiated view on trace element and organic matter contents in European soils is possible, although only to a certain extent. The degree of spatial coverage in the mapped countries differs, depending on the considered element and statistical parameter.

The study showed also that even after repeated data requests no complete and exhaustive data base for the whole of Europe could be obtained. Major data gaps still exist. Three principal reasons can be recognized for this:

- Relevant data are not collected yet.
- Data exist in fact, but could not be provided.
- Data are not comparable.

3.2 Specific remarks on the quality of data

The comparability of data was one of the key issues on the way to obtain a Europe-wide database on heavy metal and organic matter contents. In the course of the evaluation essential aspects of data harmonisation were attempted to take into consideration. Options and limits became obvious, such as the following:

- Provided **heavy metal** data were harmonised to *aqua regia* basis by using conversion algorithms. Despite of relating restrictions and inaccuracies this procedure seems a passable way and is put up for discussion.
- For **soil pH** a reference method was proposed (ISO 10390). Not all of the available data are based on that procedure. The transformation of data determined by different measurement procedures was renounced, as no original data were available. For further investigations the appliance of one measurement procedure exclusively is proposed. Already existing data could be transformed by one of the available conversion algorithm, which should be determined in advance.
- **Soil organic carbon** data are comparable up to a certain extent. Organic matter contents were calculated from measured organic carbon contents by using the factor 1.72. It should be discussed, if thereby a sufficient accuracy at European scale can be obtained. Otherwise more suitable conversion factor(s) should be stipulated.
- Due to missing criteria to compare / convert different **sampling strategies** the influence of this aspect could not be taken into account yet.
- According to the given information many of the evaluated data do not or no longer contain samples from **contaminated sites**. With regard of this harmonisation aspect data comparability was improved. Nevertheless upon a common procedure to eliminate pertinent data should be agreed.
- The national evaluations are based on different sample sizes. The comparability and spatial **representation** of the obtained results has to be checked. The need of criteria to ensure a minimum level of spatial coverage of the legend unit persists.

In summary it can be concluded that some little steps towards a Europe-wide data harmonisation have been done. However, the main standardisation problems still have to be solved. Initiatives like the CEN Standardisation Project HORIZONTAL or other activities are certainly a step into the right direction. The establishment of more structured way to coordinated better the data collection, .e.g. using existing reference facilities such as the JRC should be considered.

The significance of the chosen bases of reference has to be valued differently. It seems little target-orientated to draw conclusions about usual contents of heavy metals by means of pH or texture solely. Whereas the evaluation approach regarding parent material and land use points the right way. Questionable remains the reference level (MAT 11). For organic matter contents it seems more suitable to refer the values according to SMUs and land use.

3.3 Proposals of thresholds or guidance values

The issue of establishing new limit or guidance values upon the existing data base was heavily discussed during the meeting organized by the JRC. The pro’s and con’s raised did not always include sheer scientific arguments, but also economic or political aspects, which are however to be considered for a consensus building process. Despite the controversy of discussions, the JRC proposes the values below as a starting point for further discussion. Although we are aware of the scientific limitations raised before, one cannot ignore existing data or start from scratch. Furthermore, the algorithmic conversion as proposed by Utermann and co-workers, has a scientific background, which is accepted in the community given the lack of alternatives.

Furthermore, we are confident that the remarkable work of gathering and compiling the various data set, will establish for the first time a reference for background values of trace elements in European soil. As any reference, this can be improved, but to our opinion this is a good starting point.

3.3.1 Data base for ranges of trace elements within pH-classes

Tables 2a and 2b were established based on the observed 50 and 90 percentiles presented in Annex 1 of Part 2 of this report. Based on these data, a series of threshold values are proposed.

Table 2a - Ranges of heavy metal contents (aqua regia basis) within the pH-classes

	< 5		*> 5 – 6*		*Proposed values*
Cd	**pH H_2O**	**pH $CaCl_2$**	**pH H_2O**	**pH $CaCl_2$**	
50. P.	0.1 – 2.0	0.2	0.2 – 1.8	0.1 – 0.3	0.5
90. P.	0.4 – 3.5	0.4 – 0.8	0.4 – 4.0	0.5 – 0.9	
Cr					
50. P.	4 – 44	19 – 27	9 – 47	20 – 43	50
90. P.	11 – 126	41 – 61	17 – 128	34 – 147	
Cu					
50. P.	4 – 21	9 – 16	5 – 21	6 – 26	40
90. P.	7 – 89	26 – 36	10 – 60	29 – 66	
Hg					
50. P.	0.01 – 0.13	0.12 – 0.15	0.02 – 0.12	0.08 – 0.13	0.2
90. P.	0.03 – 0.42	0.26 – 0.35	0.05 – 0.23	0.17 – 0.27	
Ni					
50. P.	4 – 43	3 – 21	7 – 42	9 – 31	30
90. P.	9 – 62	21 – 52	15 – 82	22 – 54	
Pb					
50. P.	11 – 76	25 – 46	9 – 42	10 – 27	50
90. P.	16 – 99	49 – 128	18 – 84	41 – 73	
Zn					
50. P.	17 – 69	27 – 64	17 – 89	28 – 81	100
90. P.	33 – 198	71 – 132	37 – 154	72 – 139	

Table 2b - Ranges of heavy metal contents (aqua regia basis) within the pH-classes (cont'd)

	> 6 – 7		*Proposed values*	*> 7*		*Proposed values*
Cd	**pH H_2O**	**pH $CaCl_2$**		**pH H_2O**	**pH $CaCl_2$**	
50. P.	0.1 – 0.8	0.2 – 0.4	1	0.1 – 0.8	0.3	1.5
90. P.	0.4 – 2.6	0.7 – 1.8		0.4 – 1.2	0.5 – 0.7	
Cr						
50. P.	11 – 45	21 – 36	75	15 – 41	30 – 35	100
90. P.	18 – 97	34 – 54		23 – 54	50 – 54	
Cu						
50. P.	6 – 34	6 – 21	50	8 – 40	17 – 22	100
90. P.	14 – 75	29 – 39		13 – 80	33 – 36	
Hg						
50. P.	0.03 – 0.11	0.10 – 0.13	0.5	0.03 – 0.08	0.10 – 0.16	1
90. P.	0.06 – 0.42	0.16 – 0.35		0.06 – 0.32	0.28 – 0.32	
Ni						
50. P.	9 – 48	10 – 27	50	12 – 53	24 – 25	70
90. P.	17 – 90	26 – 43		20 – 74	36 – 43	
Pb						
50. P.	10 – 39	10 – 28	70	10 – 32	17 – 27	100
90. P.	19 – 49	39 – 104		16 – 51	41 – 64	
Zn						
50. P.	22 – 88	29 – 84	150	24 – 85	60 – 73	200
90. P.	47 – 168	105 – 185		42 – 150	107 – 132	

3.3.2 Data base for ranges of trace elements within texture classes

Similarly, the data in Table 3 create a liaison between the trace element contents and the respective texture class.

Table 3 - Ranges of heavy metal contents (aqua regia basis) within the texture classes

		No texture	*Coarse*	*Medium*	*Medium Fine*	*Fine*	*Very Fine*
Cd	50. P.	0.1 - 0.5	0.2 - 0.8	0.2 - 0.8	0.1 - 0.8	0.2 - 0.8	0.2 - 1.1
	90. P.	0.3 - 1.6	0.3 - 2.0	0.4 - 2.1	0.3 - 1.2	0.4 - 1.7	0.4 - 0.6
Cr	50. P.	2 - 19	7 - 30	17 - 35	17 - 39	19 - 58	21 - 60
	90. P.	28 - 46	13 - 73	26 - 124	28 - 66	27 - 85	31 - 59
Cu	50. P.	3 - 28	5 - 21	9 - 30	9 - 50	9 - 55	12 - 60
	90. P.	32 - 56	13 - 50	20 - 78	18 - 107	22 - 119	26 - 44
Hg	50. P.	0.09 - 0.24	0.02 - 0.10	0.04 - 0.13	0.03 - 0.17	0.04 - 0.14	0.03 - 0.13
	90. P.	0.18	0.05 - 0.40	0.06 - 0.43	0.05 - 0.35	0.07 - 0.34	0.08 - 0.27
Ni	50. P.	0.1 - 6	4 - 22	12 - 32	11 - 25	10 - 43	22 - 50
	90. P.	0.3 - 14	11 - 68	26 - 67	34 - 53	18 - 68	31 - 54
Pb	50. P.	8 - 30	8 - 24	10 - 42	12 - 48	14 - 45	12 - 61
	90. P.	17 - 86	17 - 77	19 - 124	21 - 121	21 - 116	25 - 53
Zn	50. P.	11 - 41	21 - 57	25 - 75	40 - 77	36 - 98	33 - 127
	90. P.	59 - 105	47 - 141	65 - 159	62 - 135	58 - 191	68 - 144

3.4 Recommendations

The aforementioned evaluation on the status of trace elements and organic matter contents in European soils gives a reasonably good overview on the respective background values in European Soils. However, it cannot be denied that the evaluation is limited by the restricted comparability of the underlying data. The approach used is certainly not ideal, but considering the lack of uniform trans-national datasets, i.e. obtained with the same analytical methodology (sampling, pre-treatment, digestion and determination), the concept is appropriate and 'fit-for-purpose'.

In view of the compiled information and the status in the various Member States that responded to the request for baseline data, the establishment of the following threshold values for heavy metal concentrations in soil to which sewage sludge is likely to be applied, is proposed. This is in consideration – at least in part – of the chemical behaviour of these metal elements in soil by relating the threshold values to ranges of soil pH obtained according to a standardised protocol.

Table 4 –Proposal for threshold values in function of soil pH.

Concentrations are expressed in mg/kg d.m.

Element	*Directive 86/278/EEC (6 < pH < 7)*	*5 ≤ pH < 6*	*6 ≤ pH < 7*	*pH ≥7*
Cd	1 – 3	0.5	1	1.5
Cr	*/*	50	75	100
Cu	50 - 140	40	50	100
Hg	1 – 1.5	0.2	0.5	1.0
Ni	30 – 75	30	50	70
Pb	50 – 300	50	70	100
Zn	150 – 300	100	150	200

From these figures, two main features become apparent. Firstly, the proposed threshold values increase with higher soil pH-values. This is because the solubility of heavy metals in the soil solution tend to decrease with a decreasing proton activity (increased pH). In other terms, the higher the soil pH, the more likely it will be that heavy metals remain immobilised in the soil. This is then also expressed in the bio-availability of heavy metals, for instance for plant uptake. However, there are exceptions to this behaviour, e.g. in the presence of complexing agents such as humic and fulvic acids (main constituents of soil organic matter) the resulting complexes may feature a different pH-behaviour than the "free" cation.

Secondly, the proposed values are significantly lower than the values listed in Directive 86/278/EEC. This particular proposal is driven by the fact that over past decades the control of release of these elements e.g. via industrial, municipal and domestic pathways has been improved. The introduction of suitable control mechanisms by EU-Legislation and the subsequent implementation in national regulations has brought about a net improvement of the environmental quality of the related waste streams, at least in case of heavy metal release. This is nicely illustrated for instance by the decrease of Hg emissions in case of incineration and co-incineration of non-hazardous waste.

The above listed threshold values should also ensure the safety of crops. Although there is no generally common behaviour for the various heavy metals, there is a relationship between the concentration of heavy metals in soil and the overall concentration is some plants. Depending on the element and plant of interest this relationship is more or less strongly expressed in tissue concentrations. However, based on the pathway for uptake (roots or leaves) and the solubility/mobility of the heavy metals in the soil compartment, various scenarios for the enrichment in plants can be expected. For instance, mercury features a rather low solubility in soil and is consequently not really available for the plant (usually below 0.04 mg of Hg / kg dm.)[1]. Cadmium for instance is usually up-taken from soil and consequently its concentration is usually higher in roots than in the stem, leaves or fruits of a plant[2]. Concerning lead, uptake by deposition is more significant than uptake from soil.

These examples illustrate the difficulty to assess a common behaviour for the relationship between heavy metal content and plant uptake. Furthermore, there is some disagreement in the scientific community concerning the plant-behaviour of some elements, e.g. lead. Thus, UK for instance considers lead as being so insoluble that it is not taken up by plants. Indeed, agricultural field trials indicates that a soil limit of 300mgPb/kg d.m. might be sufficient to provide a large margin of safety as regards human dietary intake of Pb. However, from a consumer protection point of view a decrease of allowable heavy metal levels is desirable, as other risks can be minimised as well.

Besides, the sheer scientific reasoning, the values proposed , are likely to find acceptance in the Member States. It remains a point of discussion to which extend the linkage between soil pH and heavy metal threshold values is sufficient to reflect the complex situation of interaction between heavy metals and the various soil properties. However, the establishment of a more differentiated system for threshold values is a step into the right directions.

At this stage is should be stressed that the establishment of pH-bound threshold values will cause some discussion on the issue of measurement uncertainty, e.g. for pH measurements. This issue has to be addressed in the revision process.

Furthermore, the proposed values are NOT considering issues of “cultural heritage”. Thus, it is known that some European soils feature higher values in Pb, due to antique lead mining activities, which can be dated back to the Roman Ere. The discussion to which extend this has to be considered in the revision process of Directive 86/278/EEC remains rather political one. From an ecotoxicological point of view, lead concentration in soil should be kept low.

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[1] *A. J. Maclean (1974). Can. J. Soil Sci. 54, 287-265.*

[2] *P. Schachtschabel, H.-P. Blume, G. Brümmer, K.-H. Hartge, U. Schwertmann (1992). Scheffer/Schachtschabel – Lehrbuch der Bodenkunde, 13. durchgesehene Auflage, Enke Verlag Stuttgart Germany, ISBN 3-432-84773-4.*

3.5 Proposal for further actions

Based on the comments received and experience made we recommend a systematic collection and evaluation of further existing data in a follow-up investigation programme ("*Long Term Action*"). The data gaps should be minimised under consideration of previously determined standards, e.g. from CEN and ISO standardisation projects such as Project HORIZONTAL. The data flow between regional, national and European institutions has to be improved. The survey should be embedded within the upcoming European Soil Framework Directive.

We see also a need for a further improvement of comparability of available data by applying conversion algorithms. As a matter of fact, one cannot ignore existing data and these algorithms appear as a useful tool to valorise the existing data treasure.

Efforts with respect to standardisation should be strengthened. In view of future data collections a consistent proceeding should be guaranteed. Therefore sampling procedures as well as analytical methods, definitions and nomenclatures should be harmonised urgently.

Last but not least in this context one should consider also the establishment of a quality management system at EU-level, which should involved major stakeholders and renowned reference centres.

4 Evaluation of the relevance of organic micropollutants in sludges

4.1 Introduction

There are some indications that most of the traditionally known organic contaminants in sludge are not expected to pose major health problems to the human population when sludge is re-used for agricultural purposes. In comparison, metal contamination of sludges is much more important with respect to human health.

The chemical properties of organics of health concern – hydrophobic and not water soluble - results in a low bioavailability to plants. Plant growth is dependent on the water solubility of nutrients and minerals and water is the transporting vector. Organics with a low water solubility are unlikely to be taken up by plants.

The presence of organic environmental pollutants, like dioxins and PCBs in agricultural crops is more the result of atmospheric deposition than direct absorption from contaminated soil. The analytical procedures for many organics are complicated and expensive – dioxins are a good example – which is an additional factor to be kept in mind when discussing monitoring of organics in sludges. Monitoring must also pay attention to the origin of sludge because the level of organic contamination may be very different when for example comparing municipal sewage sludge (mostly households) with sludges of industrial origin or sludges from storm- and run-off waters.

The conclusions presented hereafter are to be seen as an attempt to build a bridge between scientific knowledge, public sensitivity and economic feasibility. However it can not be denied that in particular research is demanded when addressing the issue of emerging pollutants. Although the analytical techniques have undergone a tremendous development and it nowadays possible to identify and quantify organic compounds in their metabolised forms, these techniques are NOT accessible for routine monitoring purposes. The availability of cheap, fast and reliable testing systems remains therefore a prerogative, if new legislation should address emerging pollutants, e.g. pharmaceutical metabolites, organo-metallic compounds or compounds deriving from personal care products.

One of the criticism forwarded during a first presentation of the results of the report's part I, was that the evaluation for the various compounds was not based sufficiently on a toxicological risk assessment. We agree partly with this statement, but the discussions here in Ispra with the respective desk officers from the national authorities showed also that public perception or financial constraints with regard to existing measurement infrastructure are at least as important as this argument. The problems resulting from this constellation were nicely highlighted by a remark of a colleague from Northern Europe, who claimed that tests with water melons exposed to PCB-containing sludge did not result into an accumulation of PCBs in the fruit. A part from the fact, that this fruit might not be the most representative crop for Northern Europe, the respective study did not address the issue PCB–metabolites that might well enter the food chain.

The example shows further how difficult it is to build this necessary consensus. Nevertheless the conclusions and recommendations presented hereafter are trying to prepare exactly this.

4.2 AOX

The observations made in Part 1 of this reports as well the need for cheap, yet reliable analytical methods are good arguments to propose the use of AOX in a legislative context. The experiences reported from the German example are indeed promising and the use of AOX might be a elegant way to avoid costly monitoring of polychlorinated compounds, once a low level has been reached over a longer period of time.

4.3 PCB

In Europe, the use of PCB has been stopped and consequently their environmental concentration, particularly in sewage sludge and soils has decreased. It actually reflects less the present input by emissions but rather a background pollution caused by recycled material with residual PCB concentrations. The introduction of limit values for sewage sludge in some EU countries has improved the situation. For these countries, we feel that there is no urgent need to monitor PCB in this matrice and one may argue about the usefulness of limit value.

However, with the enlargement of the EU, there might be a different situation for the New Member States, because of the little information available on the existing PCB load. In case of PCBs, a revised European Directive should therefore set a framework requiring rather a general registry of sewage sludges for agricultural utilization. In addition, it should give advice for the detection of point sources in cases of higher pollution in a catchment area. National regulations may fill in that frame by special limitations caused by local requirements from the toxicological point of view and the persistency of PCB in the environment.

The question of how many PCB congeners should be addressed needs to affronted onn European scale. Major differences exist and the use of standardised methods, as currently being produced by CEN under a multi-matrix, horizontal umbrella (Project HORIZONTAL) should be implemented during a revision of the existing Directive

4.4 PCDD/Fs

PCDD/F feature a similar toxicity compared to PCBs. However, their environmental concentrations are quite low, so that the mean daily intake by food is low. Sewage sludges contribute only to a low extent to environmental pollution which is dominated by atmospheric fallout. Due to restrictions and changes in chemical processes and combustion technology a decrease in the environmental pollution by PCDD/F is the result. Nevertheless the public sensitivity towards this compound class is high and some countries have set-up limit values for the agricultural use of sewage sludge.

Data of countries, which are monitoring PCDD/Fs show that concentrations in sludges have reached a level, where no problems are expected for soil if sludge is applied following existing regulations. As PCDD/Fs are strongly adsorbed to soil particles a leaching into groundwater is unlikely. Therefore periodical monitoring of a high frequency, which are cost-intensive, do not seem to be necessary. In cases where the origin of sludges is unclear or pollution can be expected further testing should be prescribed by the responsible authorities.

Similarly, as an in case of the PCBs a general framework should be set aiming first at the creation of a monitoring database and then on a occasional control mechanism.

4.5 PBDE

PBDE are dissipated into the environment by incineration of municipal waste, deposition of products containing flame retardants (computers, furniture etc.). Only the lower brominated congeners are released by volatilisation of the compound. Industrial and urban effluents are significant sources for contamination of surface water. In general PBDE are very stable in the environment and can be found in marine ecosystems by long-term transport far away from the source. Their behaviour in the environment can be compared with PCB. Concentration in surface water is below 1 ng/l, in effluents of STP between 1 – 5 ng/l. Due to the strong adsorption on fine particles, sediment concentrations increase up to 30 mg/kg dm. Nearby manufacturers using flame retardants for fire protection, sediment concentration can increase up to 3200 µg/kg dm. This is more common for such sites in the USA.

Contamination of sewage sludge in Europe varies from < 1 to 400 µg/kg dm. The most important congeners in sewage sludge are DePD and PeBD. In untreated soils < 0,2 µg/kg dm PeBDE could be found. After application of sewage sludge to soil the level goes up to 1 µg/kg dm. Strong adsorption can be assumed for PBDE, especially for the higher brominated congeners. Adsorption on suspended particles in sediments is two or three times higher because of the greater surface of the particles. Degradation depends on the conditions. PBDE are very sensitive to photolysis (degradation on soil surface is possibly important). Leaching into groundwater could not be observed due to the strong adsorption.

In biota bioaccumulation of PBDE takes place with a lot of organisms. NOEC concentration > 1000 mg/kg dm are far from real concentrations analysed in terrestrial media. With regard to human toxicity some questions still exist.

PBDE are clearly a class of emerging pollutants, which should subject of intense investigations. However, the available information about the relevance of PBDE in sewage sludge is not enough to postulate a EU-wide limit value at this stage.

4.6 LAS

LAS contamination has not decreased during the past years. A tendency for higher LAS values in sewage sludge could be assumed in STP of extended cities. The amount of LAS in sludges depends on the treatment technique. LAS concentrations in sewage sludges anaerobically digested are in the range of between 2000 to 18 000 mg/kg dm, aerobic treatment reduces the LAS level so that these sludges show concentrations between 20 and 1000 mg/kg dm.

In sewage sludge amended soils LAS values could be detected in the range of nearly 1 – 1,5 mg/kg dm after sludge application. Under good agricultural practice and normal conditions LAS will be eliminated rapidly due to relative low half-live values. DT 90 will be reached within nearly 90 days. A movement into groundwater seems not to be possible because of adsorption and degradation processes of LAS. Plant uptake plays a minor role and results in no negative effects after consuming plant parts. LAS is not carcinogenic, teratogenic and mutagenic. It has a low acute and chronic toxicity and does not act as a major ecotoxicological hazard in the terrestrial environment.

It can be concluded that if sewage sludges are applied to agricultural land with reference to the nutrient demand (N and P) of the respective cultures and sufficient withholding periods there will be no need for stringent restrictions of LAS concentrations in sludges.

In case of a stringent limitation it would be necessary to enlarge existing STP by introducing an effective aerobic stabilization step after or instead of anaerobic digestion. Another consequence could be that producers of LAS would have to change their detergent formulations.

4.7 NP and NPEO

The concentration of nonyl phenol and nonyl phenol ethoxylates in sludge varies in a wide range (1 to 600 mg/kg dm). This is caused by the sewage sludge treatment. In treatment plants using anaerobic digestion often higher contaminations by NP and NPEO could be observed. Higher contamination in sewage sludge corresponds to higher contamination of particular bound residues and in the liquid phase in effluents.

NP and NPEO are unlikely to be a problem in agricultural soils treated with sewage sludge due to degradation and medium adsorption. However, aquatic organisms (especially Daphnids, Scenedesmus and fish) react very sensitive to nonyl phenol causing estrogenic effects in a range of 10 to 20 µg/l and more. Therefore a limitation of NP and NPEO in sewage sludge could restrict higher contamination in STP effluents and could be a measure to protect water organisms against endocrine effects.

4.8 PAHs

PAH are a group of micropollutants which make a general judgement very difficult. The concentrations examined in sludge determined with regard to national regulations vary over a broad range. However, it must be taken into account that the agricultural use of sewage sludge following legal regulations with orientation to the nutrient demand and sufficient withholding times will contribute only to a limited extent to the PAH concentration in soil. Some evidence was produced that PAH are better biodegradable in agricultural soils which have received sludge, possibly caused by nutrient input, sufficient oxygen supply and adaptation of soil micro flora. Limits for a guidance substance such as benzo[a]pyrene as a representative substance might therefore be appropriate.

4.9 Phthalates

Phthalates in general and DEHP and DBP in particular are compounds, which are widely used in industry and in households causing an extended dispersion in the environment.

Contamination in surface water varies between 0,1 to 100 µg/l with a medium value of 0,5 to 3 µg/l. In effluents of STP up to 150 µmg/l for DEHP and 10 µg/l DBP had been found. This resulted in relatively high concentrations of 0,1 to 10 mg/kg dm in sediments.

Because of the strong adsorption to solids the expected removal after aerobic treatment in STP is > 90 %. Therefore, in sewage sludge contamination of DEHP and DBP may rise up to 300 to 500 mg/kg dm in maximum. The median value is very often below 30 mg/kg dm.

Contamination of soils by DEHP and DBP is low due to the relatively short degradation time under aerobic conditions. Thus, DEHP and DBP will be measured in µg range after sewage sludge application following legal regulations. With the exception of landfills where plastic material and very often some solvents are present, mobility of phthalates is negligible and unlikely to affect the ground water.

Due to the low information about effects to fresh water organisms and the tendency to bioaccumulate in biota a restriction of the phthalates should be discussed. It should be kept in mind that especially DBP and also DEHP react as weak estrogenic active substances and have the ability to bioaccumulate in fat tissues.

4.10 Comments on other organic pollutants:

A number of compound groups were not considered in this report, which is in part to lack of data, but also to the major analytical difficulties in determining them. These “Emerging Contaminants" correspond in most cases to irregulated contaminants, e.g. surfactants, pharmaceuticals and personal care products (PPCP), or gasoline additives, but also organo-metallic compounds. Typical representatives are organo-tins, musk ketone and musk xyxlenes, polyelectrolytes, antibiotic metabolites and cancer treatment agents.

As mentioned above, the underlying database is too scarce and the existing analytical methods are too cost-intensive to propose a regulatory framework for sewage sludges in theses cases. However, the authors of this report which to underline the potential risk that may arise from them and we strongly underline the need for coordinated and European-wide research with the aim to improve the current situation.

4.11 Final remarks and comments on organics

Problems and questions, which have arisen when performing this study on special organic micropollutants and cumulative parameters, have been pointed out in the respective chapters. In the following some general aspects are mentioned thus showing that solutions of the problems of organic micropollutants will not be easy.

The EU commission has given a good example with its still existing Sewage Sludge Directive 86/278/EEC by defining a span between guide and limit values for heavy metals. Upon the enlargement of the EU, it seems to be necessary to propose a flexible approach in particular for countries which have not yet reached the status of the most central European countries in collecting and treating wastewater. Therefore, strict limits for the organic micropollutants may not be the most satisfactory solution.

It should also kept in mind that often existing the national limit values are not necessarily based on a toxicological and ecotoxicological basis. Although the situation with respect to some parameters is relatively clear, there is on the other hand a number of gaps in knowledge to come to criteria for precautionary values for certain substances /substance groups.

To minimize the risk of soil pollution it seems necessary to limit the maximum load of sewage sludge dry mass per hectare and year(s) at a level at which the sludge nutrients correspond to the demand of crops. Detailed requirements for the technique of application of sludge to soil will also be helpful as safe withholding times and hygienic requirements in the case of sludge application to grassland and sensitive cultures. This should not be required only for sludge application but in the sense of a more global approach for other secondary raw materials as fertilizers and soil improvers or mixtures thereof.

In this context it should be taken into account that sewage sludge in comparison with all current materials has been the best analysed and characterized matrix for decades. In the past its examination helped to detect harmful environmental substances and provided successful restriction measures and inhibitions for them. To continue with this mode of procedure it is necessary to find a way which is effective and economic especially with regard to the technological possibilities and examination facilities in the New Member States and Acceding Countries.

To our opinion, the new regulation should envisage a distinction between limits/limit ranges for organic micropollutants which should be regularly analysed and those where this may be open to the control boards/administrations of the national governments. If necessary, the local authorities could then still opt to establish restrictions.

In this context AOX is seen as a possible tool to control halogenated compounds. This could be achieved by setting alarm values, which if attained, leads to a more specific analysis for chlorinated organics. There is good experience with the determination of this cumulative parameter and a EN Standard is currently finalised. 500 mg/kg dm have proven to be an effective tool to prevent an extensive use of halogenated compounds in household and industry and it seems that this would be tolerable also by the Accession countries so that a proposal for a guide/limit span could be 600 – 400 mg/kg dm.

Parallel to this restriction, it must be discussed if PCB and PCDD/F control with limits in some countries should be expanded to a European regulation. This is certainly an emotional discussion given the public sensitivity for these compound classes. Nevertheless, it may be a better way to leave these controls open to decisions of the European countries where regulations exist or have been planned to fill out the European frame. We agree on the general principle to have PCB control but it must not be necessary to have the same sampling/analysing frequency as for AOX. A limit of 0,8 mg/kg dm for 7 congeners seems reasonable from the experience made in some countries. In case there will be a decision for a guide/limit regulation it may be recommended as 0,6 – 1,0 mg/kg dm. The analytical determination of PCB poses no problems because the preparation of a horizontal standard in Europe is under way with a high priority.

Based on the reflections made, it is felt that PCDD/F should be left out from a European regulation because of the very costly procedure to determine concentrations at what could be defined as background values. In cases of pollution detected by general examination programs for catchment areas or the like it is open to the local authorities to set up measures of prevention and control.

PCDB, the representative group of brominated flame retardants belongs as well to the halogenated substances. Future investigation programs will contribute to a better knowledge about the occurrence, behaviour and fate in the environment. Data of today and risk estimations do not indicate that there is an urgent need of setting limits, the more as there is no analytical standard procedure for sludges and soils existing at present.

In three European countries restrictions for the PAH content in sewage sludge exist. As pointed out there is no uniform opinion how many of these substances should be put on a list for limitations. Due to the different chemical character of these substances and a different toxic potential there is no sound reason for setting up a cumulative limit value for 3, 6, 9 or more PAH. It is well known that the main sources of pollution are exhaust gases from combustion processes which contribute to soil pollution. Therefore, it is proposed to set up a range for guide/limit values especially for benzo[a]pyrene, the carcinogenic guide substance for PAH. A range 1 – 3 mg/kg dm is recommended. If it seems necessary to expand the number of PAH, the substances known as carcinogenic, too, should be taken into consideration. With view to the unknown situation in the New Member States and Acceding Countries this problem and the level of limit should handled with care.

Organic micropollutants used in detergent formulations like LAS and NP/NPEO are another group of interest. Although Denmark has set up stringent limits for LAS, one may argue whether there is a stringent need for and EU-wide limit in sludges. There is a large number of publications confirming that LAS, although present in sewage sludges at relatively high concentrations, poses no environmental impact when sludges are applied to land following the regulations existing. There is a good biodegradation in agricultural soil, residual concentrations found in some cases are far away from toxicological levels. Setting up limits for LAS would mean that the majority of sewage treatment works had to be additionally equipped with aerobic stabilization steps or that the respective industry for the production of washing and cleaning agents had to change the processes of production.

The situation is different for NP/NPEO. NP is classified as a "*priority hazardous substance*" and therefore restrictions also for sewage sludge seem necessary. As pointed out production and uses underlie existing inhibitions. To be sure that there will be no harmful effects on soil caused by NP in sewage sludge the setting up of a range of guide/limit values seems a reasonable tool. The already proposed value in Annex IV of the 3rd Draft Document on Sludge of 50 mg/kg dm could be the lower guide value of a greater span taking into account the situation in the different European countries (50 – 100 mg/kg dm).

The last group of substances which are discussed here, the phthalates are represented by diethyl-hexylphthalate (DEHP). Due to their widespread use phthalates are found in all environmental compartments. The analytical techniques for their determination have been improved in the last years, so that earlier findings of pollution loads must be estimated with care. A so called horizontal approach is under discussion, however, it has been stated that a lot of pre-normative work has to be done. So it seems too early to fix a guide/limit value for DEHP, the more as there are no toxicological needs for such a measure.

Finally, it can be concluded that there are possibilities to extend the list of pollutants when the Directive 86/278/EEC will be revised on the basis of the experience gained in different countries. On the other hand it is clear that in some fields there are still gaps in knowledge which have to be filled in by research and by examination programs to come to reasonable and safe decisions when a further revision of the directive will be discussed. There is no need for urgent measures because the use of sludge in agriculture up to now following legal regulations has caused no negative effects on soil, food and man in the last decade. This must be made more often and intensively public.

Table 5 summarises the conclusions and recommendations presented.

Table 5 – Proposal summary for the regulation of organic pollutants in sewage sludge

Organic compounds	*Analyzing frequency*		*Guide/limit values span (mg/kg dm)*	*Remarks*
	like heavy metals	*examination upon request*		
AOX	+	+	400-600	
PCB	—	+	0,6 – 1,0	
LAS	—	—	—	
NP/NPEO	+	+	50 – 100	
DEHP	—	+	—	Subject to further research
PAH	—	+	—	Number of PAH subject to further discussion
Benzo[a]pyrene	+	+	1,0 – 3,0	
PBDE	—	—	—	Subject to further research
PCDD/F (ng TE/kg dm)	—	+	50 - 200	Environmental importance decreases

European Commission

EUR 22265 EN – DG Joint Research Centre, Institute for the Environment and Sustainability
Luxembourg: Office for Official Publications of the European Communities
2006 – 30 pp. – 21.0 x 29.7 cm
Scientific and Technical Research series
ISBN 92-79-021222
ISSN 1018-5593

Abstract

This report summarises the conclusions of two independent, JRC coordinated studies concerning background values of relevant organic micro-pollutants in sewage sludges as well as background values of some trace elements and organic matter in (uncontaminated) soils, in view of a revision of the Sewage Sludge Directive /278/EEC.
The first study, which was conducted by DIN e.V. (R. Leschber) and the second study, which was conducted by BGR (J. Utermann, O. Düwel, I. Nagel), were presented and discussed during two JRC workshop sessions 15-16 April 2004.), done in close collaboration with DG ENV. Attendees of these events were invited via the official channels involving the Environmental Departments and Authorities of the Member States. Numerous additional comments were received and the draft versions of the aforementioned reports were adopted accordingly.
In the following, the conclusions of the JRC concerning the main findings of the two studies as well as recommendations and observations are presented. It has to be stressed that these recommendations and observations are not necessarily those of the authors of the respective studies.

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ISBN 92-79-02122-2
9 789279 021220